THE EVALUATION OF AGRICULTURAL PRACTICES AND THE USE OF CONSTRUCTED WETLANDS FOR TREATMENT OF PESTICIDE RUNOFF IN COMMEWIJNE, SURINAME

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FACULTY OF **BIOSCIENCE ENGINEERING**

The evaluation of agricultural practices and the use of constructed wetlands for treatment of pesticide runoff in Commewijne, Suriname

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Thesis submitted in the fulfillment of the requirements for the degree of Doctor (PhD) in Applied Biological Sciences

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De evaluatie van landbouwkundige praktijken en de toepassing van biologische vijvers voor de behandeling van pesticiden runoff in Commewijne, Suriname

Cover
Cover picture by Mahabali Shirley Sahina:
Constructed wetland planted with *Nymphaea amazonum* and *Echinocloa polystachya*

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GOD:

YOU raise me up, so I can stand on mountains;

YOU raise me up to walk on stormy seas:

I am strong, when I am on YOUR SHOULDERS:

YOU raise me up……To more than I can be.

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May GOD bless all of you

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LIST OF ABBREVIATIONS

A	Acaricide
AA-EQS	Annual Average Environmental Quality Standard
A.I.	Active Ingredient
ADI	Acceptable Daily Intake
AHFSU	Agricultural Health and Food Safety Unit
ANOVA	Analysis of Variance
ARfD	Acute Reference Dose
AZP	Azinphos Methyl
Avnonveg	Average nonvegetated
Avveg	Average vegetated
BCL	Boscalid
BGB	Brilliant Green Bile
BMP	Best Management Practices
BW	Bodey Weight
CAC	Codex Alimentarius Commission
$CaCl_2$	Calcium chloride
CCME	Canadian Council of Ministers of the Environment
CPF	Chlorpyrifos
C_{ss}	Concentration in Suspended Solids
CW	Constructed Wetland
CWs	Constructed Wetlands
D	Depth
d	Days
DBPs	Disinfection By-products
DEA	Di Ethyl Atrazine
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
DOM	Dissolved Organic Matter
DW	Dry Weight
CDPR	California Department of Pesticide Regulation
DT_{50}	Half-life
EC	Emulsifiable Concentrate
EC_{cond}	Electrical Conductivity
E. coli	Escherichia coli
EHC	Environmental Health Criteria
E.mutata	Eleocharis mutata
EQS	Environmental Quality Standard
ESA	Ethane Sulfonic Acid

EU	European Union
F	Flow counts
FAO	Food and Agriculture Organization of the United Nations
FC	Fecal Coliforms
fDOC	Fraction Dissolved Organic Carbon
ffree	Fraction unbound pesticide
FOCUS	FOrum for the Co-ordination of pesticide fate models and their USe
fPOC	Fraction Particulate Organic Carbon
FSMS	Food Safety Management System
G.A.P.	Good Agricultural Practices
GC-MS	Gas Chromatography-Mass Spectrometry
GDP	Gross Domestic Product
GlobalG.A.P.	Global Good Agricultural Practices
GUS	Groundwater Ubiquity Score
GW	Groundwater
h	Hours
H	Herbicide
ha	Hectare
HPLC	High Performance Liquid Chromatography
HLR	Hydraulic Loading Rate
HRT	Hydraulic Residence Time
I	Insecticide
IPU	Isoproturon
IESTI	International Estimated Short-Term Intake
IQR	Inter Quartile Range
IUPAC	International Union of Pure and Applied Chemistry
KAP	Knowledge Attitudes and Practices
K_d	Soil-Water Partitioning Coefficient
K_f	Freunlich Sorption Coefficient
K_{oc}	Organic-Carbon Partitioning Coefficient
K_{ow}	Octanol-Water Partitioning Coefficient
L	Length
LC_{50}	Lethal Concentration 50
LOD	Limit of Detection
MAC-EQS	Maximum Acceptable Concentration-Environmental Quality Standard
2,4-MCPA	2-methyl-4-chloro-phenoxyacetic acid
MeP	MethylParathion
min	Minutes
MIRA	Milieurapport Vlaanderen
MPN	Most Probable Number
MRL	Maximum Residue Limit

MUG	4-methylumbelliferyl-β-D-glucuronide.
MUMA	Multiple Use Management Areas
N.amazonum	Nymphaea amazonum
NaN3	Sodium azide
NC	Not Calculated
ND	Not Detected
N.D.	Not Determined
NIOSH	The National Institute for Occupational Safety and Health
NOEC	No Observed Effect Concentration
NONVEG	Non Vegetated
NPP	Napropamide
NPS	Non Point Source
NT	Not Tested
NVWA	Nederlandse Voedsel en Waren Authoriteit
o.a.	Onder andere
OC	Organic Carbon
OECD	The Organization for Economic Cooperation and Development
OM	Organic Matter
ORP	Oxidation Redox Potential
OXA	Oxanillic acid
PCZ	Prochloraz
pH	acidity
PMRA	Pest Management Regulatory Agency
PPCPs	Pharmaceuticals and personal care products
PPDB	Pesticide Properties Data Base
PPEs	Personal protective equipment's
ppm	Parts per million
PRT	Pesticide Retention Time
PVDF	Poly Vinylidene Difluoride
rpm	rates per minutes
RQ	Research Question
S	Salinity
SB	Sediment Basin
SETAC	Society of Environmental Toxicology and Chemistry
SFCW	Surface Flow Constructed Wetlands
SFO	Single First Order
SSFCW	Subsurface Flow Constructed Wetlands
Si	Silt
SI-units	International System of Units of Standards
SL	Soluble Concentrate
S-MET	S-metolachlor

Sol_{H2O}	Water Solubility
SS	Suspended Solids
SSFCW	Subsurface Flow Constructed Wetlands
SW	Surface Water
T	Temperature
TBZ	Tebuconazole
TC	Total Coliforms
TDS	Total Dissolved Solids
TEA	Terminal Electron Acceptors
TKN	Total Kjeldahl Nitrogen
T-RFLP	Terminal restriction fragment length polymorphism
USDA	United States Department of Agriculture
USDA-NRCS	United States Department of Agriculture-National Resources Conservation Service
US EPA	United States Environmental Protection Agency
v	Velocity
Vvar	Variability factor
VEG	Vegetated
V_p	Vapor Pressure
WHO	World Health Organization
WLA	Water Loopkundige Afdeling
WRP	Wetlands Research Program
WTO	World Trade Organization
WWTP	Waste Water Treatment Plant
Yr	Year

EXECUTIVE SUMMARY

Worldwide an increase is expected in the use of pesticides for crop protection e.g. because of a projected increase in the world's population of 30% by the year 2050. The intensive use of pesticides leads to an substantial risk of contamination of the environmental compartments and of harmful effects on humans. With the lack of appropriate legislation and control mechanisms and misuse of pesticides in developing countries such as Suriname, it is expected that humans and the environment remain at risk. Farmers in the Commewijne district in Suriname rely on surface and groundwater for irrigation purposes. Mostly small drainage ditches are constructed in which pesticides and other contaminants like pathogens by means of runoff move to surface water and/or leach to groundwater. During the dry season, water from these ditches is used for irrigation purposes and poses an additional risk, because crops can become contaminated with e.g. pesticides and pathogens. Constructed wetland systems were proven to reduce and eliminate the exposure risk of the aquatic environment because of conversion (into less toxic compounds) and removal or retention of pesticides and other harmfull components such as nutrients, trace metals and pathogens. This research conducted in Commewijne, Suriname is focused on 2 main parts: 1) good agricultural practices (G.A.P.) of pesticide use and its associated risk and 2) the use of constructed wetland systems to mitigate pesticide pollution in agricultural runoff. By means of a literature review, the state-of-the-art of constructed wetland systems to reduce/remove the main constituents/contaminants in agricultural runoff is presented. **Appendix 1, Table a**, provides specific details on the different constructed wetland (CW) systems used in mitigation of pesticide pollution. A set of 4 research questions (RQ) are formulated which are related to the specific research objectives described in **chapter 1**:

RQ1: Do farmers in the Commewijne district, Suriname apply good agricultural practices (G.A.P.) in the use of pesticides and do vegetables comply with applicable maximum residue limit (MRL) values? What is the risk of human exposure to pesticides?

RQ2: What is the status of the pesticide contamination of the different environmental compartments (surface water and sediment) and the bacteriological quality of irrigation water, and what are the types of wetland plants present in the main ditches of the research area?

RQ3: What is the potential of vegetated and non-vegetated wetland mesocosms to remove selected pesticides in agricultural runoff?

RQ4: What is the potential of a field scale wetland to mitigate pesticide pollution originating from agricultural runoff under two conditions: 1) wetland planted with a monoculture and 2) wetland planted with multiple plant types?

In the literature review presented in **chapter 2 and Appendix 1**, it was found that different types of constructed wetland systems exist, and that the fate and removal/retention of pesticides is dependent on the pesticide properties such as polarity, solubility, the log organic carbon

partitioning coefficient (log K_{oc}), volatility and half-life times. Other factors of importance are the type of design (batch, continuous, single stage, multiple stage, surface flow, subsurface flow, and hybrid), the effect of other contaminants, and the presence of vegetation and substrate properties. Water soluble pesticides mainly disappear by microbial degradation in the water phase or are retained by the water-plant interphase. Some pesticides, however, have depending on their properties (e.g. relatively high K_{oc}) very good sorption capabilities. For the more hydrophobic pesticides such as pyrethroids, sedimentation/adsorption to particles that are high in organic carbon content is one of the main mechanisms. Accumulation of pesticides in sediment of field scale systems might be influenced by wetland vegetation and plant density. Desorption or remobilization of pesticides has also been observed in different wetland studies.

In the present work, one of the aims is to investigate (by means of a face to face questionnaire) the good agricultural practices of pesticide use within farmers community in the research area, district Commewijne and to perform a pesticides risk assessment study (**chapter 3**). The face to face survey was conducted under a total population of (78) farmers in the region. A minor group of farmers made use of non-authorized pesticides and cocktails, despite the G.A.P.-program which has been implemented in 2003. Parts of the personal protective equipment's (PPEs) such as a mouth cap, protective clothes and shoes were used as prescribed in the G.A.P. procedures. However, often the use of gloves and safety glasses was not according to G.A.P. guidelines. Residue analysis of vegetables was conducted and residues of the pesticides imidacloprid and chlorothalonil were measured. The comparison of the International Estimated Short Term Intake (IESTI) for imidacloprid with the European Union-Acute Reference Dose (EU ARfD) showed that in all types of vegetables, the residue is lower than the EU ARfD. Pesticide misuse was related to different factors such as the low educational level of farmers, the easy access to non-authorized pesticides from e.g. neighbouring countries and the lack of legislation related to pesticide use and handling and control mechanisms.

The next step of this research **(chapter 4)** was to investigate whether the most frequently used pesticides (assessed in the study described in chapter 3) could be detected in different environmental compartments (surface water, sediment and water from small drainage canals near farmer's fields) in the research area. The microbial quality of irrigation water (surface and phreatic groundwater) was also determined by a Most Probable Number (MPN) technique. To be able to select plants for future wetland studies, a vegetation diversity study was performed involving the main drainage canals in the Alkmaar area in district Commewijne. Results showed that surface water was contaminated with the fungicide chlorothalonil and the insecticides lambda-cyhalothrin and α-endosulfan. Sediment and drainage ditches near farmer's fields were also contaminated with lambda-cyhalothrin and imidacloprid, while chlorothalonil was below the limit of detection (LOD). The microbiological water quality did not comply to the WHO norm for phreatic ground- and surface water and to a lesser extent for surface water. Total and fecal coliforms and *Escherichia coli* were present in numbers higher than this norm. The vegetation diversity assessment showed the abundance of different wetland plants and a detailed overview is

presented in **Appendix 2**, **Table b**. It can be concluded that a pesticide problem exists for all investigated compartments and that water used for irrigation purposes does not comply with the WHO norm regarding microbial quality. The latter can pose an additional risk, because irrigated crops can become contaminated with these pathogens. Different plants which were detected in the research area, can e.g. based on their abundance in the main drainage ditches of Commewijne, be used in wetland mesocosm experiments or field trials.

The study described in chapter 4, showed that different environmental compartments such as drainage canals contained the insecticides lambda-cyhalothrin and imidacloprid. Therefore, the dissipation/retention of these insecticides was investigated in wetland mesocosms planted with different plant monocultures, *Nymphaea amazonum* and *Eleocharis mutata* (**chapter 5).** Mesocosms were exposed to different pesticide concentrations (low (batches 1 and 2), high (batches 3 and 4) and extra high for imidacloprid (batch 5) in repeated batches. Different concentrations were used, to resemble the different application rates used by farmers in the research area. A faster and complete removal of both pesticides from the water phase in both types of mesocosms was observed when mesocosms were exposed to lower pesticide concentrations. During repeated batches (batch 2 was a repetition of batch 1 and batch 4 a repetition of batch 3), an enhanced removal of both pesticides was observed. For batch 5 also higher imidacloprid removal rates were observed at the start of the experiment, which indicate a possible enhanced biodegradation due to repeated exposure of mesocosms with this pesticide. However starting from time 72 h, a higher removal was observed for batch 1 (lowest target concentration). Statistical tests also show significant differences between the DT_{50} of batch 5 and batch 1, indicating that for Batch 5, the half-life is significantly higher. The latter contradict the previous statement of an enhanced biodegradation, which would be characterised by lower DT_{50} values for batch 5.
The main dissipation pathway for lambda-cyhalothrin was sedimentation/adsorption in both types of mesocosms as shown by the amounts found in the sediment of both mesocosms. The amounts detected in plant, however, indicated different uptake mechanisms, and was high for imidacloprid in *N.amazonum*. The half-life in water was determined for both pesticides and was much lower for lambda-cyhalothrin in both types of mesocosms. Less than 50% from the amount of lambda-cyhalothrin added could be quantified in sediment and plants. The major pathway for imidaclorpid, in the *Nymphaea amazonum* mesocosms was accumulation in plant tissue and sediment particles (around 96.4%), while for the *Eleocharis mutata* mesocosms this value was much lower (around 15.9%). The differences indicate different removal mechanisms of plants and interactions with the sediment phase. They might be explained by differences in pesticide properties and fate characteristics. These include, the low water solubility and high log octanol-water partitioning coefficient (log K_{ow}) of lambda-cyhalothrin and the high water solubility, accumulation by plants and photolytic degradation potential of imidacloprid.

In chapter 5, vegetated mesocosms were exposed to increasing concentrations of lambda-cyhalothrin and imidachloprid. It was therefore decided to perform these tests with a constant

and high dose of pesticides during 5 consecutive batches (**chapter 6**). This, instead of the two batches performed for each exposure concentration. It may be possible that a prolonged exposure might have a different effect on pesticide removal. To assess the influence of vegetation (*P. australis)* on pesticide's removal, non-vegetated mesocosms were used besides vegetated ones. Complete dissipation of chlorothalonil from the water phase was obtained at the end of the experiment, with the exception of batch 4 (the third repetition of the experiment). Comparable results were obtained for lambda-cyhalothrin for the batches 1, 2 (first replication) and 5 (fourth replication), while a decrease in removal was observed in batches 3 and 4. This might be related to a temporary toxicity towards microorganisms capable of degrading lambda-cyhalothrin.
Complete dissipation of imidacloprid from the water phase was obtained for the last batch in the end of the experiment, for both vegetated and non-vegetated mesocosms. This indicates a possible microbiological enhancement due to repeated exposure. Pesticide half-life times were the highest for lambda-cyhalothrin. However, these half-life times were lower than the half-life times obtained for this pesticide in previous mesocosm study (chapter 5). The highest and fastest removal was observed for chlorothalonil (volatile pesticide), followed by imidacloprid and lambda-cyhalothrin, with no significant differences between vegetated and non-vegetated mesocosms. This finding is also in agreement with reported DT_{50} values for these pesticides. The present study also shows that *P. australis* has a relatively good accumulation potential towards lambda-cyhalothrin, compared to the *N. amazonum* and *E.mutata* plants in mesocosm studies described in chapters 5 and 6 and can be considered for use in future wetland experiments.

The potential of a field scale wetland to mitigate lambda-cyhalothrin and imidacloprid pollution from agricultural runoff during two phases (phase 1, with a monoculture and phase 2, with multiple plant types) was the last experiment conducted and described in **chapter 7**. Design considerations were based on outputs from mesocosm experiments described in chapter 5 and 6 (reaction rate constant, half-life time values, pesticide accumulation in different plants) and from flow measurements (chapter 6). Lambda-cyhalothrin was not detected in water (phase 1), while few detections were done in phase 2. This was related to the longer duration of the simulated rainfall and the conditions of the soil (dry during phase 2). For both, phase 1 and 2, a good removal of imidacloprid was observed from the water phase of the surface flow wetland, which showed to be somewhat higher and more constant for phase 2. More lambda-cyhalothrin was found in the roots of *Nymphaea amazonum* plants during phase 2 compared to phase 1, while for the mesocosm experiment described in chapter 5 no detection was done in the roots of this plant. It seems like with a combination of two plants a certain competition is present, which should be further investigated. For imidacloprid a somewhat higher uptake (*N.amazonum*) was observed for roots (phase 1) compared to roots (phase 2). Also compared to phase 1 a much lesser amount is found in roots compared to the amounts found in shoots and leaves. More imidacloprid was detected in both the plant upper parts and roots of *Echinocloa polystachya* plants, indicating a higher accumulation of this pesticide compared to *N. amazonum* plants. Lower amounts of both lambda-cyhalothrin and imidacloprid were found in sediment during phase 2 compared to phase 1. The different and preferential accumulation potential of plants and amounts found in sediment

was not observed in mesocosm studies (chapter 5 and 6).In these studies the accumulation of pesticides in sediment (chapter 5) was not measured at regular time-intervals and with multiple plant types, which was the case in the field scale study. Therefore it was difficult in those studies to draw a conclusion that with a higher vegetation density lesser amounts of pesticides are found in sediment. In the mesocosm study described in chapter 5, lambda-cyhalothrin was not detected in the roots of *N. amazonum*, while in the field-scale study lambda-cyhalothrin was measured in some sampling points during both phase 1 and phase 2. Also less imidacloprid was measured in the roots of *N. amazonum* in phase 2 compared to phase 1. Contrary to the high uptake of lambda-cyhalothrin by *P. australis* (chapter 6), *E. polystachya* (phase 2 of the field-scale experiment), showed a much higher uptake towards both lambda-cyhalothrin and imidacloprid, which was however much higher for imidacloprid. For both types of pesticides lesser amounts were found in sediment during phase 2 compared to phase 1 and might be explainded by the presence of a more dense vegetation during phase 2. Sorption experiments using substrates from the research area showed that the highest distribution coefficient (K_d) was found for lambda-cyhalothrin in substrate with the lowest clay and organic matter content. Sorption kinetics for lambda-cyhalothrin showed a constant accumulation in time, while for imidacloprid an irregular pattern was found. This might indicate desorption of pesticides to occur and was also observed during the field scale wetland experiment. Based on the high removal efficiencies for high initial imidacloprid concentrations, it is expected that with this field scale wetland actual (much lower) effluent concentrations (presented in chapter 4) in ditches will be further reduced to acceptable environmental levels.

In conclusion, the results reported in this doctoral thesis have provided insight in the existing pesticide problem in district Commewijne and in the different processes occurring in wetland treatment systems on various scales. Results indicate a good removal of pesticides from agricultural runoff and wetlands can provide a sustainable and environmentally sound approach in abetement of pesticide pollution. Results can be used for future studies making use of other types of pesticides (in combination with other contaminants) and other types of wetland configurations with the ultimate goal to provide farmers, government agencies and researchers with preliminary design guidelines for the efficient abatement of agricultural runoff in Suriname. Further recommendations are provided in **chapter 8**.

GEDETAILLEERDE SAMENVATTING

Wereldwijd wordt een toename verwacht in het gebruik van bestrijdingsmiddelen voor de gewasbescherming, onder andere vanwege de (geprojecteerde) toename van de wereldbevolking met 30% in het jaar 2050. Dit intensief gebruik van bestrijdingsmiddelen zal ook zorgen voor een toenemende risico op contaminatie van milieu compartimenten en kan een schadelijk effect hebben op mens, waterbronnen en biodiversiteit. Met een gebrek aan geschikte wetgeving en controle mechanismen, alsook onjuist gebruik van bestrijdingsmiddelen in ontwikkelingslanden zoals Suriname, is het te verwachten dat de mens en het milieu een risicogroep blijven. Landbouwers in district Commewijne, Suriname maken gebruik van oppervlakte en grondwater voor irrigatie doeleinden. Meestal worden kleine drainagesloten aangelegd, welke er ook voor zorgen dat pesticides en andere contaminanten zoals pathogenen meegevoerd worden naar oppervlakte water of kunnen uitlogen naar grondwater. Tijdens de droge tijd, wordt het water van deze drainage sloten gebruikt voor de irrigatie, welke dan een bijkomend risico oplevert, namelijk dat gewassen gecontamineerd kunnen raken met o.a. bestrijdingsmiddelen en pathogenen. Biologische vijversystemen hebben bewezen dat ze deze blootstellingsrisico's voor o.a. het aquatisch leven kunnen mitigeren. Dit middels transformatie (omzetting in minder toxische vormen), verwijdering of retentie van bestrijdingsmiddelen, nutrienten, spore elementen en pathogenen. Dit onderzoek welke uitgevoerd is geworden in Commewijne, Suriname bestaat uit 2 fasen namelijk: 1) een onderzoek naar goede landbouwkundige praktijken (G.A.P.) van bestrijdingsmiddelengebruik en gerelateerde risico's voor de consument en 2) onderzoek naar de afbraak van bestrijdingsmiddelen in landbouw runoff middels biologische vijvers. Middels een literatuuronderzoek is inzicht verkregen in 'the state-of-the-art' van biologische vijvers om de belangrijke contaminanten/stoffen in landbouw runoff te verwijderen en of op te nemen. In bijlage 1, wordt een gedetailleerde overzicht gegeven over verschillende (biologische) vijversystemen welke gebruikt zijn voor het mitigeren van bestrijdingsmiddelen in landbouw runoff. In totaal zijn 4 onderzoeksvragen geformuleerd welke mede gerelateerd zijn aan de specifieke doelstellingen (**hoofdstuk 1**) van dit doctoraatsonderzoek:

1: Maken landbouwers in distrikt Commewijne, Suriname gebruik van goede landbouwkundige praktijken m.b.t. het gebruik van bestrijdingsmiddelen en voldoen groentestalen aan de geldende Maximale Residu Limieten (MRLs)? Indien dit niet het geval is, wat is het potentiële risico voor de consument?

2: Wat zijn de gehalten aan bestrijdingsmiddelenresiduen in verschillende milieucompartimenten (oppervlakte water en sediment) en wat is de microbiologische kwaliteit van het irrigatiewater en welke water planten zijn aanwezig in de belangrijkste drainage kanalen van het Commewijne gebied?

3: Wat is het vermogen van biologische vijvers om geselecteerde bestrijdingsmiddelen te verwijderen in beplante en onbeplante mesocosms?

4: Wat is het vermogen van een biologische vijver op veldschaal om geselecteerde bestrijdingsmiddelen in landbouw runoff te verwijderen gedurende twee fasen namelijk fase 1: vijver beplant met een monocultuur van planten en fase 2: vijver beplant met meerdere planten soorten?

Uit de resultaten van de literatuurstudie welke gepresenteerd is in **hoofdstuk 2 en bijlage 1, Tabel a**, is gebleken dat er verschillende typen van biologische vijversystemen bestaan, en dat het lot en verwijdering van bestrijdingsmiddelen afhankelijk is van onder andere hun eigenschappen en gedrag in het milieu zoals polariteit en oplosbaarheid, de log organisch koolstof-water partitie coëfficiënt (log K_{oc}), vluchtigheid en halfwaarde tijd. Andere factoren van belang zijn het type ontwerp (batch, continue, enkel of multi stappen systeem, oppervlakte of ondergrondse stroming en hybride systemen), de invloed van andere contaminanten, aanwezigheid van planten en eigenschappen van het substraat.
Het is gebleken dat de verwijdering van wateroplosbare bestrijdingsmiddelen voornamelijk gebaseerd is op microbiologische afbraak in de waterfase en retentie door de waterplant interfase, alhoewel sommige polaire bestrijdingsmiddelen, afhankelijk van hun eigenschappen zoals een relatieve hoge K_{oc} waarde, ook een goede sorptievermogen bezitten. Voor de meer hydrofobe bestrijdingsmiddelen zoals pyrethroids, vindt voornamelijk sedimentatie/adsorptie aan deeltjes (die met voorkeur een hoog gehalte aan organisch koolstof hebben) plaats. Accumulatie van bestrijdingsmiddelen in sediment in systemen op veldschaal kunnen beïnvloed worden door de planten in de vijver en plantdichtheid. Ook werd desorptie of remobilisatie beschreven in verschillende studies.

In de volgende stap van dit doctoraatsonderzoek, was het doel om middels een “face to face” enquête na te gaan indien bestrijdingsmiddelen werden toegepast volgens goede landbouwkundige praktijken. Dit geschiedde onder een totale populatie (78) van landbouwers in de regio Alkmaar **(hoofdstuk 3)**. Een kleine groep landbouwers bleek gebruik te maken van verboden bestrijdingsmiddelen en cocktails, dit ondanks het G.A.P. programma dat in 2003 werd geïmplementeerd. Het gebruik van ‘personall protective equipment’s’ of PPEs (mondkap, beschermde kledij en schoenen) was volgens G.A.P. richtlijnen, met uitzondering van handschoenen en veiligheidsbrillen. Voor de pesticide risico evaluatie, vond residu onderzoek van gewassen plaats, waarbij residuen van imidacloprid en chlorothalonil werden gevonden. Waarden voor imidacloprid bleken hoger te zijn dan de maximale residu limieten van de Europese Unie (EU-MRLs). Vergelijking van de IESTI ‘(International Estimated Short Term Intake)’ voor imidacloprid met de EU ArfD ‘(Acute Reference Dose)’, toonde dat in alle gecontamineerde groentesoorten, deze waarde lager was dan de EU ArfD. De hoofdredenen voor het onjuist omgaan met bestrijdingsmiddelen waren voornamelijk, de lage scholing van de landbouwers, de makkelijke toegang tot verboden bestrijdingsmiddelen aangeleverd vanuit de buurlanden, het ontbreken van

adequate wetgeving en controle mechanismen alsook een registratie systeem voor het gebruik van bestrijdingsmiddelen.

De volgende stap van het doctoraatsonderzoek was om na te gaan indien de meest gebruikte bestrijdingsmiddelen (gehaald uit de resultaten van de G.A.P. survey beschreven in hoofdstuk 3) ook terug te vinden waren in de verschillende milieucompartimenten zoals oppervlakte water en sediment. Daarnaast werd de microbiologische kwaliteit van irrigatie water bepaald via een 'most probable number (MPN)' techniek. Verder werd ook een vegetatie onderzoek uitgevoerd in de 2 belangrijkste drainage kanalen in het onderzoeksgebied Alkmaar (**hoofdstuk 4**). Vegetatie, zoals beschreven in o.a. hoofdstuk 2, speelt een belangrijke rol bij het verwijderingsproces van bestrijdingsmiddelen in biologische vijvers. Ook kan een selectie van planten plaatsvinden die gebruikt kunnen worden voor toekomstig onderzoek met biologische vijversystemen. Resultaten toonden dat oppervlakte water gecontamineerd was met de fungicide chlorothalonil en de insecticiden lambda-cyhalothrin en α-endosulfan. Echter konden alleen 6 soorten bestrijdingsmiddelen worden gekwantificeerd middels o.a. 'Gas Chromatography Mass Spectrofotometry (GCMS)'. Ook sediment en water afkomstig van drainagesloten dichtbij de velden van lanbbouwers, bleken gecontamineerd te zijn met lambda-cyhalothrin en imidacloprid. Chlorothalonil, werd niet gedetecteerd in water of residuen daarvan waren beneden de detectielimiet.
Een slechte microbiologische kwaliteit werd geconstateerd voor zowel freatisch grond- en oppervlaktewater. De slechte microbiologische kwaliteit werd bevestigd door aanwezigheid van totale en fecale coliformen en *Escherichia coli* in hoeveelheden hoger dan de WHO norm. De diversiteitsstudie gedaan voor planten, toonde de aanwezigheid van verschillende wetland planten, waarvan een gedetailleerd overzicht is gegeven in **Appendix 2, tabel b**. Selectie van planten voor wetland studies heeft o.a. via de resultaten van deze assessment plaatsgevonden. Geconstateerd kan worden dat er wel sprake is van bestrijdingsmiddelen contaminatie is in het onderzoeksgebied en wel voor de verschillende compartimenten. De kwaliteit van het water gebruikt voor irrigatie doeleinden blijkt een slechte microbiologische kwaliteit te hebben, welke een bijkomend probleem met zich meebrengt namelijk het risico, dat gewassen via irrigatie gecontamineerd kunnen raken met o.a. pathogenen. Verschillende planten welke gedetecteerd zijn in het onderzoeksgebied kunnen op basis van hun voorkomens in de drainagesloten van Commewijne, gebruikt worden in experimenten met mesocosms of veldproeven.

Uit de studie beschreven in hoofdstuk 4, was het duidelijk dat in verschillende milieu compartimenten zoals drainage sloten, residuen van de insecticiden lambda-cyhalothrin en imidacloprid zijn gedecteerd. Het was nu ook mogelijk om planten te selecteren die in mesocosm experimenten gebruikt konden worden. Daarom werd besloten om de verwijdering/retentie van de twee insecticiden nader te onderzoeken. Dat gebeurde in mesocosms beplant met verschillende plant monoculturen (*Nymphaea amazonum* en *Eleocharis Mutata*) en blootgesteld aan verschillende concentraties van bestrijdingsmiddelen (laag (batches 1 en 2)), hoog (batches 3 en 4) en extra hoog voor imidacloprid (batch 5)) **(hoofdstuk 5**). Verschillende concentraties

werden gebruikt om de verschillende applicatie hoeveelheden aan bestrijdingsmiddelen in het veld na te bootsen. Resultaten tonen een veel eerdere en een volledige verwijdering van bestrijdingsmiddelen uit de waterfase voor beide mesocosms blootgesteld aan de laagste dosering. Dit vond plaats gedurende herhaaldelijke blootstellingen (batch 2 was een herhaling van batch 1 en batch 4 van batch 3). De oorzaak hiervan kan gelegen hebben aan een verbeterde microbiele degradatie van bestrijdingsmiddelen, veroorzaakt door de herhaaldelijke blootstelling aan bestrijdingsmiddelen. Voor batch 5 werden ook hogere verwijderingssnelheden voor imidacloprid waargenomen, welke ook een verbeterde degradatie zouden kunnen aanduiden. Echter blijkt dat for batch 1 (laagste docering) vanaf 72 h, een veel hogere verwijdering is in vergelijking met die voor batch 5. Statistische analyse toont aan dat er significante verschillen zijn tussen de halfwaarde tijden van batch1 en batch5, waarbij die voor batch 5 significant hoger zijn. Dit laatste weerspreekt het fenomeen van een verbeterde microbiele degradatie, welke door lagere halfwaarde tijden zou worden gekarakteriseerd.
De belangrijkste dissipatie route voor lambda-cyhalothrin was sedimentatie/adsorptie in beide typen mesocosms en werd bevestigd door hoeveelheden welke teruggevonden werden in het sediment van beide mesocosms. Aan de hand van de hoeveelheden aan bestrijdingsmiddelen gedetecteerd in planten, bleken er verschillendde opname mechanismen te zijn, met name voor imidacloprid in *N.amazonum* mesocosms. De halfwaarde tijden in water werden bepaald voor beide bestrijdingsmiddelen en bleken veel lager te zijn voor lambda-cyhalothrin in beide typen mesocosms. Verder bleek dat minder dan 50% van de toegediende hoeveelheid aan lambda-cyhalothrin teruggevonden werd in sediment en planten. De belangrijkste dissipatie route voor imidacloprid in de *Nymphaea amazonum* mesocosms was accumulatie in plantweefsel en sediment deeltjes (ongeveer 96.4%), terwijl dit slechts 15.9% was in *Eleocharis mutata* mesocosm. Deze verschillen indiceren verschillende verwijderingsmechanismen van planten en interacties met het sediment en zijn gerelateerd aan verschillen in physico-chemische eigenschappen en dissipatie routes van bestrijdingsmiddelen. Deze zijn o.a. de lage oplosbaarheid in water en hoge log octanol-water partitie coefficient (log K_{ow}) van lambda-cyhalothrin. Voor imidacloprid zijn deze o.a. de hoge oplosbaarheid in water, accumulatie door planten en fotolytisch afbraak.

In het voorgaande experiment (hoofdstuk 5), werden beplante mesocosms blootgesteld aan bestrijdingsmiddelen met toenemende concentraties van lambda-cyhalothrin en imidacloprid. Er werd daarom besloten om mesocosm experimenten uit te voeren, waarbij blootstelling geschiedde aan een constante en hoge concentratie bestrijdingsmiddelen gedurende 5 opeenvolgende batches (**hoofdstuk 6).** Dit in tegenstelling tot de 2 batches uitgevoerd voor elke blootstellingsconcentratie in voorgaand experiment. Het is mogelijk dat door langdurige blootstelling er een ander effect op de pesticide verwijdering zal zijn. Verder werden ook niet beplante mesocosms gebruikt om het effect van vegetatie op pesticide verwijdering na te gaan. Volledige verwijdering van chloorthalonil vond plaats aan het einde van het experiment voor alle batches met uitzondering van batch 4 (derde herhaling van het experiment). Vergelijkende resultaten werden gevonden voor de verwijdering van lambda-cyhalothrin voor de batches 1 en 2 (eerste herhaling) en 5 (vierde herhaling), terwijl een terugval in verwijdering te merken was voor

de batches 3 en 4. Dit kan mogelijk te wijten zijn aan een tijdelijke toxiciteit tegenover de microorganismen verantwoordelijk voor lambda-cyhalothrin afbraak. Complete verwijdering van imidacloprid werd in batch 5 verkregen voor zowel beplante als niet-beplante mesocosms aan het einde van het experiment en indiceert een verbeterde microbiologische afbraak als gevolg van herhaaldelijke blootstelling van de mesocosms. Halfwaarde tijden voor bestrijdingsmiddelen waren het hoogst voor lambda-cyhalothrin, terwijl in hoofdstuk 5 een veel lagere waarde werd gevonden. De hoogste en snelste verwijdering was voor chlorothalonil (welke ook vluchtig is), gevolgd door imidacloprid en lambda-cyhalothrin, echter zonder significante verschillen voor beplante en niet-beplante mesocosms. Deze resultaten stemmen overeen met de gevonden halfwaarde tijden. Deze studie resulteerde in een relatieve hoge opname potentiaal van *P. australis* voor lambda-cyhalothrin, vergeleken met de *N. amazonum* en *E. mutata* planten uit voorgaand mesocosm experiment (hoofdstuk 5) en resultaten kunnen gebruikt worden voor o.a. vervolg onderzoek.

Het vermogen van een biologische vijver op veldschaal om lambda-cyhalothrin en imidacloprid concentraties te verlagen in een gesimuleerde landbouw runoff, gedurende twee fasen (fase 1, vijver beplant met een monocultuur en fase 2, met meerder plantensoorten) was het laatst uitgevoerde experiment (**hoofdstuk 7)**. Het ontwerp van de vijver, was o.a. gebaseerd op de output (reaktiesnelheidsconstante, halfwaarde tijden en pesticide accumulatie in plantmateriaal) verkregen uit de mesocosm experimenten (beschreven in hoofdstuk 5 en 6) en de debiet bepaling (hoofdstuk 6). Overeenkomstig met het voorgaand mesocosm experiment (hoofdstuk 6), werden ook op veldschaal slechts in enkele waterstalen (fase 2) residuen van lambda-cyhalothrin gemeten. De detectie bleek o.a. afhankelijk te zijn van de duur van de kunstmatige irrigatie alsook de condities waaronder het landbouwareaal verkeerde (heel droge bodems tijdens fase 2). Voor zowel fase 1 als fase 2 werd een goede afname van imidacloprid gevonden uit de waterfase van de vijver, welke echter een wat hoger en stabiel verloop vertoonde voor fase 2, namelijk in de vijver beplant met 2 typen van planten. Meer lambda-cyhalothrin werd gevonden in de wortels van *N. amazonum* planten in fase 2, vergeleken met fase 1, terwijl in het mesocosm experiment beschreven in hoofdstuk 5, geen detectie plaatsvond in de wortels van deze plant. Het lijkt alsof met een combinatie van twee plantensoorten een bepaalde competitie aanwezig is voor de opname van lambda-cyhalothrin, welke nader onderzoek vereist.
Voor imidacloprid werd in tegenstelling tot de bevindingen in fase 1, veel minder teruggevonden in de wortels ten op zichte van het bovenste plant gedeelte. In fase 2 werd ook meer imidacloprid gedetecteerd in zowel de wortels als het bovenste plantmateriaal van *Echinocloa polystachya in vergelijking met N. Amazonum,* welke een hogere opname indiceert ten op zichte van de *N. amazonum* planten. Het verschil in opname alsook de voorkeur bij opname door planten werd niet waargenomen in de voorgaande mesocosm experimenten (hoofdstuk 5 en 6). Hetzelfde kan ook gezegd worden voor de hoeveelheiden aan bestrijdingsmiddelen teruggevonden in sediment. In de mesocosm experimenten (hoofdstuk 5) werd de accumulatie van bestijdingsmiddelen niet met de tijd opgevolgd en ook niet vergeleken met een mix cultuur van planten. Hierdoor kon er geen duidelijke conclusie worden getrokken indien met meer vegetatie dichtheid er minder aan

bestrijdingsmiddelen wordt teruggevonden in sediment. Zoals eerder beschreven werd in het mesocosm experiment beschreven in hoofdstuk 5 geen detectie gedaan voor lambda-cyhalothrin in de wortels van *N. Amazonum*, terwijl bij onderzoek of veld-schaal dit wel het geval was. In tegenstelling tot de hoge accumulatie van lambda-cyhalothrin in *P. australis* (hoofdstuk 6) werden voor de *E. polystachya* plant veel hogere waarden gemeten voor beide bestrijdingsmiddelen, welke echter veel hoger was voor imidacloprid. Voor beide pesticiden (lambda-cyhalothrin en imidacloprid) werden lagere hoeveelheden teruggevonden in sedimentstalen genomen in fase 2., die mogelijkerwijs gerelateerd zijn aan de hogere plantdensiteit voor die fase.

Het resultaat van sorptieproeven waarbij gebruik werd gemaakt van substraten uit het onderzoeksgebied, toonde aan dat de hoogste distributie coefficient (K_d) werd gevonden voor lambda-cyhalothrin en wel in het substraat met het laagste percentage aan klei en organisch materiaal. De sorptie kinetiek experimenten toonden aan dat er een constante opname verloop met de tijd was voor lambda-cyhalothrin, terwijl voor imidacloprid een onregelmatig patroon werd gevonden. Dit onregematig patroon geeft mogelijk het fenomeen van desorptie aan en werd ook waargenomen voor de biologische vijver proef. Lettende op het hoog verwijderingsrendement voor initiële hoge concentraties van het bestrijdingsmiddel, kan gesteld worden dat werkelijke effluent concentraties van imidacloprid (die veel lager zullen zijn) in drainagesloten verder gereduceerd zullen worden tot aanvaardbare concentraties.

Concluderend kan gesteld worden, dat met dit doctoraatsonderzoek inzicht is verkregen in de bestaande pesticide problematiek in district Commewijne alsook in de verschillende processen die plaatsvinden in biologische vijversystemen op verschillende schaalniveaus. De verkregen resultaten tonen een optimale verwijdering van bestrijdingsmiddelen uit landbouw runoff in Suriname. Biologische vijversystemen zijn een duurzame en milieuvriendelijke alsook relatief goedkope systemen om water vervuild met o.a. bestrijdingsmiddelen te zuiveren. Resultaten kunnen gebruikt worden voor het uitvoeren van vervolgonderzoek, waarbij gebruik gemaakt kan worden van andere typen bestrijdingsmiddelen (in combinatie met andere contaminanten) en andere biologische vijver configuraties, met als uiteindelijke doel de landbouwer, overheidsinstanties en onderzoekers te voorzien van preliminaire ontwerp richtlijnen voor het succesvol mitigeren van landbouw runoff in Suriname. Verdere aanbevelingen zijn weergegeven in **hoofdstuk 8**.

Chapter 1 Background and general introduction

This chapter provides an overview of the use of pesticides in general and in Suriname. In particular, health and food safety issues are also presented, together with the concepts of G.A.P. (Good Agricultural Practices) and GLOBAL G.A.P. in Suriname. The processes governing pesticide movement from the agricultural field to receiving water bodies (e.g. ditches, creeks, swamps, rivers) are described. In addition, an overview is given of the framework of this research, together with the research objectives and the thesis outline.

1.1 Pesticide use in Agriculture

Pesticides are widely used to ensure high crop yields and their use is expected to increase based on a growing world population and the need for more food supply (Ecobichon 2001; Zhang et al. 2007; Popp et al. 2013). The dependence on pesticides for crop protection has been increased in developing countries, where many older, more toxic and inexpensive chemicals are used (Ecobichon 2001). About 30% of pesticides marketed in developing countries with an estimated market value of USD 900 million annually do not meet internationally accepted quality standards such as those from the FAO/WHO (Popp et al. 2013). With the lack of appropriate legislation and control mechanisms, especially in developing countries such as Suriname, this trend increases pesticide risks of pesticide exposure and poses a health threat to humans and the ecosystem. Another aspect is the loss of export opportunities for developing countries especially for horticultural crops as the developed countries are tightening maximum allowable residue levels (Popp et al. 2013). According to the World Health Organization (WHO 2008), 25% of the world pesticide production is used in developing countries. The WHO estimated that annually, a million people are poisoned with 20,000 cases resulting in death (WHO 1990). The main causes of poisoning are insufficient knowledge of the pesticides used and failure in using the appropriate PPEs (personal protective equipment's) such as gloves, boots and respiratory equipments (Snelder et al. 2006). Studies to assess the Knowledge, Attitudes, and Practices (KAP) regarding pesticide use were conducted worldwide to understand the occupational settings and work conditions in which pesticides are handled and applied by farmers (Pasiani et al. 2012). Repeated pesticide applications may put farmer's health and the environment at risk and cause adverse effects. These effects depend on several factors such as toxicity and persistence of the pesticide, the measures taken during its application and the dosage applied (Damalas and Eleftherohorinos 2011). Several tools (surveys, pesticide residue monitoring, dietary risk assessments) can be used to assess good agricultural practices (G.A.P.) regarding pesticide use.

1.2 Agriculture in Suriname

Agriculture employs 70% of the economically active population in Suriname. People are mainly involved in the production of staple grains such as corn and rice and bananas. Besides rice and bananas, other important crops are vegetables, plantains, citrus fruits and cassava. These crops together account for 61% of the total value of agricultural production in the 2006-2010 period in Suriname but represents a relatively small share of 9% of the Gross Domestic Product (GDP) in 2012 (Derlagen et al. 2013). Compared to the relatively high number of small scale farmers (10,000-12,000), less than 10 fruit and vegetable exporters are noted. Export of agricultural commodities (Figure 1-1) takes place to different regions namely the European Union (EU) and the Caribbean. The largest export products are rice and bananas and the lowest are vegetables (see 'Other' in Figure 1-1). Production of rice takes place on medium to large scale farms and makes use of advanced production techniques such as aereal pesticide spraying. (Milton 2009).

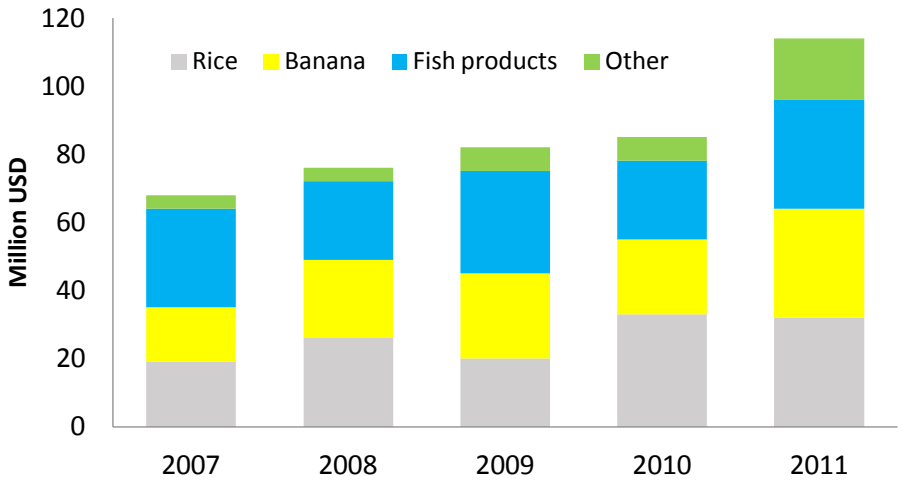

Figure 1-1: Value of main agricultural export products of Suriname for the period 2007-2011 and expressed in million USD. Other refers to vegetables, plantains, citrus fruits and cassava (Derlagen et al. 2013)

Developing countries mostly lack data on the actual amounts and type of pesticides used (Fleischer and Waibel 2003; Abate et al. 2000). Based on the available data for Suriname, it is observed that in general, in the period 2010-2014, the import of pesticides (insecticides, herbicides and fungicides) increased (see Table 1-1).

Table 1-1: Pesticide import numbers (ton/year) for Suriname, from 2010 up to 2014 (Van Sauers 2016)

Pesticide Ton/year	2010	2011	2012	2013	2014
Insecticides	244.7	213.3	351.0	73.1	250.0
Herbicides	705.9	681.3	461.2	277.2	716.0
Fungicides	429.0	684.4	474.3	447.4	415.9
Rodenticides	11.0	8.3	1.4	3.5	13.0
Molluscides	31.0	15.0	3.1	5.2	20.3

Information about pesticide use in Suriname, concentrations in different environmental compartments and mitigation techniques is scarce. Two studies (Graafsma et al. 2006; Van Spijker et al. 2009) on pesticide exposure in the Nickerie "rice" district report on the increasing numbers of suicide and attempted suicide using pesticides. These reports highlight the easy accessibility and distribution of pesticides in Suriname. Locally, one report of importance (Van de Lande 2001) is focused on the role of Surinamese women in agriculture and highlighted the use of pesticides in the Commewijne district.

1.3 Health and food safety in Suriname

The general policy document of the Ministry of Agriculture, Animal Husbandry and Fisheries about the agricultural sector in Suriname consists of seven strategic objectives. Objective two of the policy document aims at securing agricultural health and food safety. To secure agricultural health and food safety, an Agricultural Health and Food Safety Unit (AHFSU) was established in 2001, with the aim to coordinate health and food safety issues and to comply with the World Trade Organisation (WTO) sanitary and phyto-sanitary measures. It aims to facilitate production and trade of crops and has to establish a food and analytical laboratory (Ministry of Agriculture in Suriname 2013). The AHFSU organizes G.A.P. trainings for vegetable and fruit producers regarding pesticide use. In addition, a trace-back system for farmers, to identify the "polluters" in cases where MRLs (Maximum Residue Limits) are exceeded was established (Ministry of Agriculture in Suriname 2008).

1.3.1 Global Good Agricultural Practices and Food Safety

The GLOBAL G.A.P. (Global Good Agricultural Practices) previously called the EurepG.A.P. is a farm assurance program that helps producers to comply with globally accepted criteria for food safety, sustainable production methods, worker and animal welfare, responsible use of water, compound feed and plant propagation materials. This program also helps in harmonized certification, which means savings for producers, as they do not need to undergo several audits against different criteria every year. Different modules (from plant and animal production to plant propagation material) exist, for which different guidelines are applicable. These guidelines are subdivided in different parts. It is up to the users to make the appropriate selection of the parts, which are needed in undergoing the certification process. This process consists of

fulfilment of general and national G.A.P. requirements related to food safety and quality. Also of importance are checklists, guidelines, supporting documents, and harmonization instruments**.** In connection to food safety, countries need to establish a Food Safety Management System (FSMS) by means of the implementation of various quality assurance and legal requirements related to production, organization and the environment (Luning et al. 2008). FSMS are based on good agricultural and good hygiene practices and require following national and international, public and private standards and guidelines (CAC 2010a). According to Unnevehr (2000) and Kirezieva et al. (2015), certain countries (e.g. of the European Union) have put more stringent requirements related to food safety and are difficult to reach by export companies from less developed countries. In general, EU companies own more information about food safety issues because of their sampling and sector monitoring capacities (Jaffee and Henson 2005; Abhilash and Singh 2009). Companies in developed countries have put many efforts in the management of pesticides; e.g., pesticide residues are nowadays regulated by the European Commission and by the Codex Alimentarius Commission (EC 2005; CAC 2010b; Popp et al. 2013). Other drawbacks in developing countries are the failures of local institutional environments to support companies in setting and implementing their own FSMSs and the poor legislative framework in these countries, which still requires improvements in the set-up and enforcement (Kirezieva et al. 2015).

1.3.2 Good Agriculture Practices in Suriname

The Good Agricultural Practices (G.A.P.) project started in 2003 in Suriname, by the department for agriculture of the Ministry of Agriculture, Animal Husbandry and Fisheries of Suriname. According to this ministry, G.A.P. involves a set of measures, actions and activities, which are or can be undertaken during cultivation, harvest, separation and packaging of crops. The focus is on eliminating and reducing the possible risks of microbial and chemical contamination (Grauwde 2010). Eleven export-based crops are registered under this G.A.P. project in Suriname (Table 1-2).

Table 1-2: "G.A.P." registered crops of Suriname with their scientific and Dutch or traditional name (Ministry of Agriculture, Animal Husbandry and Fisheries in Suriname, G.A.P. department 2010)

Scientific name	Dutch and traditional name
Vigna unguiculata subsp. sesquipedalis	Kouseband
Momordica charantia	Sopropo (Paré)
Solanum macrocarpon	Antroewa
Solanum melongena	Boulanger (Aubergine)/eggplant
Abelmoschus (Hibicus) esculentus	Oker/ocher
Capsicum Chinense	Peper/pepper
Cestrum latifolium	Bitawiri
Allium Ampeloprasum var.porrum	Prei /leek
Dolichos lablab	Sim
Apium graveolens	Soepgroenten/soup vegetables
Cucurbita moschata	Pompoen/pumpkin

1.4 Pesticide movement to aquatic systems

The major problem in chemical crop protection is pesticide movement away from the treated crop and into the different environmental compartments (e.g. surface and ground water, sediment, biota and air). This thesis focuses mainly on water contamination and agricultural runoff.
The losses of pesticides from the field into aquatic systems are:

1) losses during direct application;
2) losses by runoff: pesticides can be in a dissolved, granular or adsorbed state;
3) losses due to aerial drift;
4) losses due to volatilization from the crop followed by atmospheric deposition;
5) contamination due to uptake by biota and subsequent movement of pesticides to aquatic systems.

These transport routes not only depend on physical and chemical properties of the pesticides such as water solubility, but also on factors such as the pesticide application technique, weather and climate conditions and characteristics of the land such as the type of soil and the slope (US EPA 1984; Damalas and Eleftherohorinos 2011). One of the solutions to overcome the problem of pesticide runoff from the field is a constructed wetland, which is the main study object of this thesis.

1.4.1 Point sources versus diffuse sources of contamination

Pesticides enter aquatic systems from the farmland by means of a) point sources and b) diffuse sources. Point sources are easier to control depending on the farmers will to apply good agricultural practices regarding the use and handling of pesticides. Point sources of pesticide pollution may include storage, handling, mixing and cleaning areas for pesticides. Spillages, leakages of spray equipments or at pesticide storage facilities, rinsing of spray leftovers and spray drift of small droplets are main contributors to this type of pesticide pollution (De Wlde et al. 2006; MIRA (Milieu Rapport Vlaanderen) 2013). Point sources from non-agricultural use e.g. application of pesticides on roads are also an important source of contamination (Reichenberger et al. 2007). According to Holvoet et al. (2007), the contribution of point sources in river systems in Europe ranges from 20-80% of the total contamination of pesticides in water. According to MIRA (2013), these point sources can be avoided by adequate education of farmers.
Diffuse or non-point sources of pesticide pollution arise from agricultural applications in the field. Examples of diffuse inputs are drain flow, base flow seepage, surface and subsurface runoff and soil erosion, spray drift and atmospheric deposition (Reichenberger et al. 2007; Holvoet et al. 2007; Gregoire et al. 2009; Vymazal and Březinová 2015).

1.4.2 Pesticides in surface runoff

Most articles related to the treatment of water contaminated with pesticides refer to surface runoff, agricultural runoff or non-point source (NPS) runoff. Pesticide losses by means of surface runoff mostly represent less than 1% of the applied active substance and rarely exceed 10% (Gregoire et al. 2009). The amount of pesticides lost through runoff depends on the quantity present on the soil surface and the occurrence of rainfall. The time between application of a pesticide and the first (intense) rainfall is also a very important factor, because the shorter this period, the higher the amount of pesticides in agricultural runoff. Several other factors influences the amount of pesticides in runoff and are based on watershed characteristics such as soil properties, vegetation cover, infiltration capacity of the soil and pesticide characteristics. According to Holvoet et al. (2007), soluble compounds are more susceptible towards losses by means of runoff than soil erosion. Soil erosion is mostly linked to strong sorbing pesticides. Spanoghe et al. (2005) conducted an experiment to investigate runoff of a mixture of three herbicides from hard surfaces (e.g. cement, asphalt and concrete). These pesticides (glyphosate, diuron and diflufenican) had different adsorption properties. Results showed that application of rainfall (60 mm/h) immediately after pesticide application to asphalt, resulted in 70% of glyphosate loss in runoff, compared to only 5% for diflufenican. The high amount of glyphosate loss was attributed to its high water solubility and the apolarity of the (asphalt) surface.

1.5 Framework of research and thesis outline

The world's population is projected to increase by 30% to 9.2 billion by the year 2050 (Popp et al. 2013). Worldwide pesticides are increasingly used for crop protection (Ecobichon 2001; Zhang et al. 2007; Snelder et al. 2008). The Surinamese government enforced several measures to comply with new trade agreements arising from globalization. With the establishment of the Agricultural Health and Food Safety Unit (AHFSU) in 2001, the government aimed to ensure food quality and safety. This AHFSU has to coordinate health and food safety issues and guide farmers to comply with the World Trade Organization (WTO) sanitary and phyto-sanitary measures. Despite these efforts, several notifications from the Netherlands (one of the biggest export markets) show pesticide residues with levels higher than the Maximum Residue level (MRL) in fresh produce originating from Suriname. With the lack of appropriate legislation and control mechanisms and misuse of pesticides in developing countries such as Suriname, it is expected that humans and the environment remain at risk. Farmers in the Commewijne district in Suriname rely on surface and ground water for irrigation purposes. Mostly small drainage ditches are constructed through which pesticides by means of agricultural runoff are transported to surface water. Pesticide leaching to groundwater can also occur. During dry periods when irrigation water is scarce, water from the drainage ditches is used for irrigation purposes. Wastewater from e.g. households enters these ditches by means of self-constructed drainage channels. This may increase human exposure towards pesticides and pathogens. To minimize and eliminate this risk, the implementation of guidelines for pesticide best management practices (BMPs) are needed. In this context the construction of a wetland might be a solution (Moore et al. 2006; Matamoros et al. 2007; Reichenberger et al. 2007; Budd et al. 2009; Gregoire et al. 2009; Elsaesser et al. 2011; Elsayed et al. 2014; Vallée et al. 2015A; Vallée et al. 2015B; Vymazal and Březinová 2015). Constructed wetland systems have proven to reduce and eliminate the exposure risk to the aquatic environment because of conversion (into less toxic compounds) and removal or retention of contaminant (see chapter 2).

Based on the above, the following research questions (RQ) were formulated:

RQ1: Do farmers in district Commewijne, Suriname apply good agricultural practices (G.A.P.) in the use of pesticides and do vegetables comply with applicable MRL values? What is the risk of human exposure to pesticides?

RQ2: What is the status of the pesticide contamination of the different environmental compartments (surface water and sediment) and the bacteriological quality of irrigation water and what are the types of wetland plants present in the main ditches of the research area?

RQ3: What is the potential of vegetated and non-vegetated wetland mesocosms to remove selected pesticides in agricultural runoff?

RQ4: What is the potential of a field scale wetland to mitigate pesticide pollution originating from agricultural runoff under 2 conditions: 1) wetland planted with a monoculture and 2) wetland planted with multiple plant types?

The first part of this doctoral thesis investigates G.A.P. in the use of pesticides, with the aim to obtain more information on the used pesticides, the amount of pesticides applied, justification of their use and the use of personal protective equipment's (PPEs). This allowed to select the pesticides of concern for a more targeted sampling and analysis of different environmental compartments and for performing wetland studies. In the second part, wetland experiments are first performed on micro scale, to better understand ongoing processes, followed by validation making use of a field scale wetland. Half-life times (DT_{50} values), uptake of selected pesticides by different wetland plants in mesocosm systems and flow measurements are used for design of the field scale wetland.

Based on the research questions the following objectives were formulated:

1) To present by reviewing the literature, the state-of-the-art of constructed wetland systems to reduce/remove the main constituents/contaminants present in agricultural runoff. This is addressed in **chapter 2**.
2) To assess the agricultural practices of pesticide use and its associated risk of human exposure. This is addressed in **chapter 3**.
3) To determine the amount of pesticides present in different environmental compartments, the bacteriological quality of irrigation water and the vegetation diversity of the main drainage ditches in the Alkmaar area. This is addressed in **chapter 4**.
4) To assess the efficiency of the removal of selected pesticides in vegetated and non-vegetated mesocosms, which enables the determination of pesticide half-life degradation times in wetland systems and which provides input data for the design of a field scale wetland. This is addressed in **chapter 5 and chapter 6**.
5) To assess the feasibility of a field scale surface flow wetland to mitigate pesticide pollution originating from agricultural runoff and to determine the amount of pesticides in wetland plants and sediment, under two conditions, 1) wetland planted with a monoculture and 2) wetland planted with multiple plant types. This is addressed in **chapter 7**.
6) To assess the adsorption of selected pesticides on different sediments from the wetland research area. This is addressed in **chapter 7**.

The main answers to the research questions and the discussion and conclusions are presented in **chapter 8**, together with a number of aspects that still deserve attention for further investigation.

1.6 Research criteria

The motivation for the selection of 1) the research area, 2) the methodology and 3) types of pesticides used in this study.

1.6.1 Selection of the area

The Commewijne area was selected as research site, because of the following:

- The Alkmaar and Tamanredjo regions are main agricultural production areas and are two of the six resorts within the Commewijne district located near the capital Paramaribo.
- Willingness of the head and members of the Ministry of Agriculture and the farmers in this area to participate in this study is high to facilitate agricultural research.
- Almost all farmers are G.A.P.-registered and it is therefore of interest to conduct the first part of the research (G.A.P. analysis of pesticide use) within this group.
- The only scientific article on pesticide use in agriculture in Suriname is based on research conducted in this area. The results of that work indicate bad agricultural practices concerning the use of pesticides.
- The researcher was familiar with the area and farmers because of previously conducted surveys.
- In Commewijne there is a lack of potable and surface water for irrigation purposes. Pumping wells and rainwater are used for personal hygiene, drinking purposes and for humans and animals. By investigating the potential of wetland systems to mitigate pesticide pollution, results can be used for improving water quality and to allow the reuse of water for e.g. irrigation purposes.
- There is no sewer system for grey wastewater, which mostly flows to the drainage ditches. During dry periods, this water is also used for irrigation purposes. Wetlands can be used to improve not only the chemical (e.g. removal of pesticides) but also the microbial quality of this water.
- In the past there were several plantations (sugar and coffee) in Commewijne and the infrastructure (drainage ditches) in the field is still in place, allowing low cost experiments.

1.6.2 Selected methodology

The output of the different sequential steps and their mutual relations is presented in Figure 1-2.

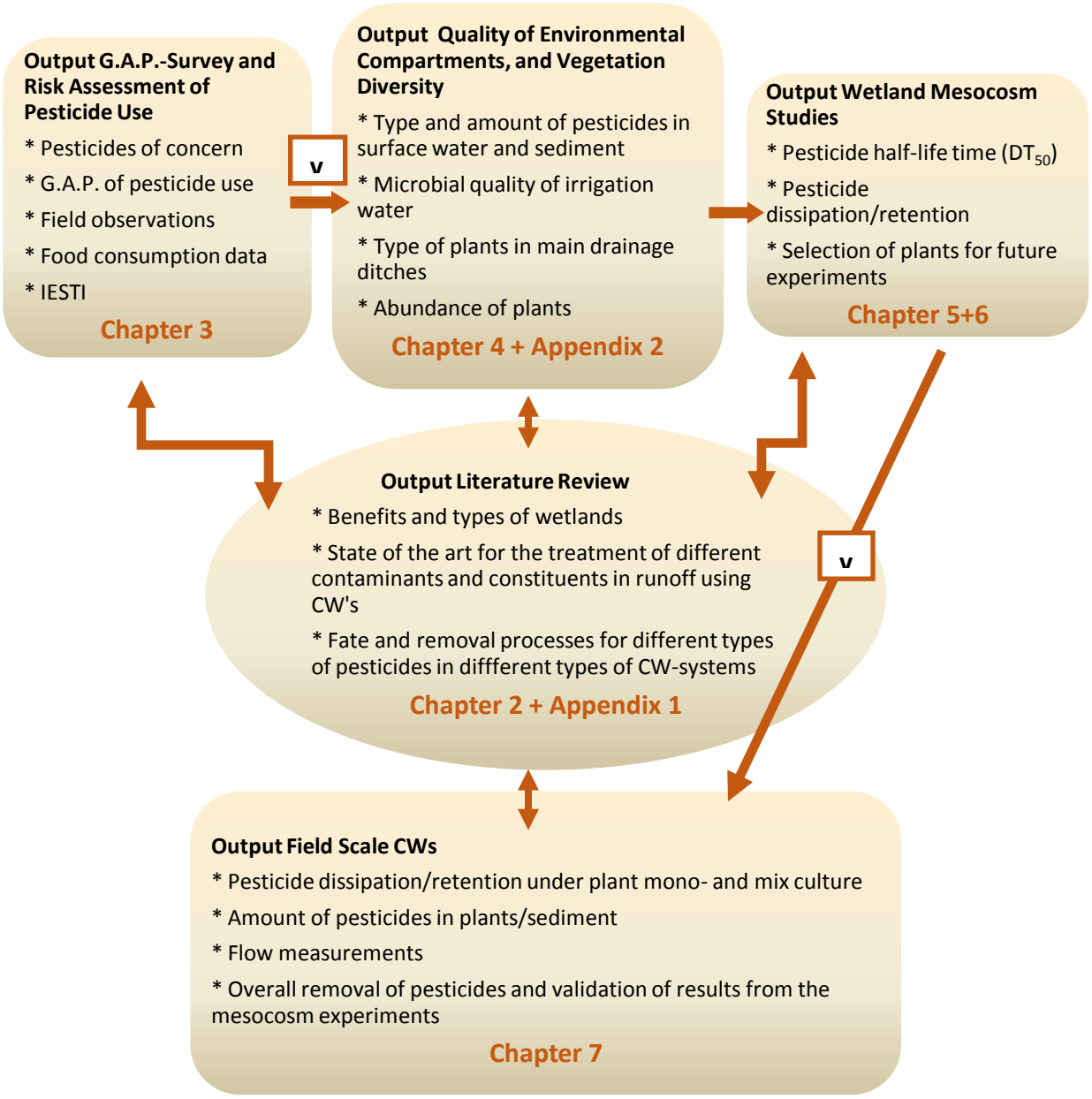

Figure 1-2: Output of different sequential steps in the methodology of this research and their mutual connectivity. Results from chapter 3 are validated (v) in chapter 4, while those of chapter 5+6 are validated (v) in chapter 7

The output of the G.A.P. survey, residue analysis, literature review and vegetation diversity is used as input data for the execution of wetland mesocosm experiments. The output of these mesocosm experiments is validated by means of field scale constructed wetland experiments.

1.6.3 Selected pesticides

Based on the G.A.P. survey, lambda-cyhalothin and imidacloprid belong to the main pesticides applied in the research area. Selection of these pesticides for this doctoral thesis was based on:

1. Their frequency of use in the research area.
2. Physical and chemical properties of pesticides; e.g. lambda-cyhalothrin is a hydrophobic pesticide, while imidacloprid is highly water-soluble.
3. The availability of a validated analytical method for the identification and quantification of these pesticides in different environmental matrices.
4. Pesticide-plant combinations and the novelty of the research. The pesticide-plant combinations used in present wetland studies (chapter 5, 6 and 7) have not yet been tested in other wetland treatment systems.

1.6.4 Pesticides half-life in water-sediment systems

FOCUS (FOrum for the Co-ordination of pesticide fate models and their USe) has established a number of work groups to develop procedures for estimating concentrations of plant protection products and their metabolites in various environmental compartments including the soil-water systems. They also develop recommendations for calculating degradation kinetics in the EU registration process, which is a fundamental component of environmental risk assessments of plant protection products.
According to FOCUS (2006), the DT_{50} describes the time taken for a 50 % decline in mass or concentration of a substance to occur by dissipation or degradation from the environment or an environmental compartment after it has been applied to, formed in, or transferred to, an environmental compartment. The half-life should further be derived from fitting single first-order (SFO) kinetics to data.

Degradation includes processes, such as microbial degradation, hydrolysis and photolysis, breakdown of substances in different environmental compartments by transforming them into degradation products, oxidation and transformation into microbial biosynthetates or polymerization products. Dissipation is defined as the overall process leading to the eventual disappearance of substances from the environment, or an environmental compartment.
Dissipation comprises two main types of processes: transfer processes, such as volatilization, leaching, plant uptake, run-off or erosion and degradation processes such as microbial degradation, hydrolysis and/or photolysis transforming substances into degradation products (FOCUS 2006).
A standard procedure to conduct water-sediment studies for pesticides that are non-volatile or slightly volatile is described in “The Organization for Economic Cooperation and Development (OECD) Guideline 308 (OECD, 2002b) and by the Society of Environmental Toxicology and Chemistry (SETAC) (1995). In these guidelines, a minimum of six sampling times (including zero time) is considered necessary to estimate kinetic endpoints over an experimental period not normally exceeding 100 days, or when 90% of the test substance has dissipated by transformation and/or volatilization.

Chapter 2 Treatment of agricultural runoff in constructed wetlands: state of the art

In this chapter, a classification of wetlands according to Ramsar (2010) is given, followed by a brief overview of natural wetlands in Suriname and the current status on CWs used for the treatment of agricultural runoff. The different types of wetlands, factors affecting wetland performance and removal mechanisms are described. The main group of constituents/contaminants in agricultural runoff and their removal/retention in CWs systems are also presented. In addition, general design considerations and management options are discussed.

2.1 Classification of wetlands

A wetland is a complex assemblage of water, substrate or sediment, plants (vascular and algae), litter (primarily fallen plant material), invertebrates (mostly insect larvae and worms) and an array of microorganisms (most important bacteria) (US EPA 1995). The world's wetlands area ranges between 8.3 and 10.1 million km^2, of which 1.3-1.5 million km^2 are rice paddies (Vymazal et al. 2011; Mitsch and Gosselink 2015). Wetlands are classified according to Ramsar (2010) in three main categories, i.e., (1) marine/coastal wetlands; (2) inland wetlands; (3) human-made wetlands. Within the marine/coastal wetlands a code system of 12 types of CWs exists, ranging from type A, i.e., permanent shallow marine waters which in most cases are less than 6 m deep up to type Zk (a), i.e., karst and other subterranean hydrological systems. The inland wetlands are divided into i.e., type L (permanent inland deltas) up to type Ski (b) (karst and other subterranean hydrological systems, inland) (Ramsar 2010). The human-made wetlands are subdivided into 10 types (Table 2-1).

The Surinamese terrain consists of young and old coastal plains interspersed with brackish and freshwater wetlands. More than two-thirds of Suriname's mangroves and other coastal wetlands are protected in nature reserves or managed as 'Multiple Use Management Areas' (MUMA). From West to East the following MUMA are present: Bigi Pan MUMA (area: 67,900 ha), North Coronie MUMA (area: 27,200 ha), North Saramacca MUMA (area: 88,400 ha) and North Commewijne-Marowijne MUMA (61,500 ha). These MUMA are natural wetland systems and are maintained to optimize long-term natural productivity and conservation.
Suriname has ratified the Ramsar convention in 1985 and designated the Coppename monding (Saramacca) as wetland (Ramsar site no. 304, area 12,000 ha) of international importance. This importance was based on the findings of Morrison and Ross (1989) that Suriname supported over half (52% or 1.53 million) of the total shorebirds they counted around the entire coastline of South America.

Table 2-1: Human-made wetlands subdivided in different hydrological systems (Ramsar 2010)

	Human-made	**wetlands**
Type	**Description**	
1	Aquaculture (e.g., fish/shrimp) ponds.	
2	Ponds, including farm ponds, stock ponds, small tanks; (**generally less than 8 ha**).	
3	Irrigated land, including irrigation channels and rice fields.	
4	Seasonally flooded agricultural land (including intensively managed or grazed wet meadows or pasture.	
5	Salt exploitation sites: salt pans, salines, etc.	
6	Water storage areas: reservoirs/barrages/dams/impoundments (**generally over 8 ha**).	
7	Excavations: gravel/brick/clay pits, borrow pits, mining pools.	
8	Wastewater treatment areas: sewage farms, settling ponds, oxidation basins, etc.	
9	Canals and drainage channels, ditches.	
Zk(c)	Karst and other subterranean hydrological systems, human-made.	

The human-made wetlands of Suriname are mostly of type 1-4 and type 9 (Table 2-1). No scientific data are, however, present on the use of constructed wetlands for the purpose of water quality improvement e.g. for the treatment of pesticide runoff.

2.2 Pros and cons of Wetlands

Wetlands are important features in the landscape that provide numerous beneficial services for people and for fish and wildlife (US EPA 2015). Some of these services are:

- protecting and improving water quality;
- providing fish and wildlife habitats; many species of birds and mammals rely on wetlands for food, water and shelter, especially during migration and breeding;
- storing floodwaters and maintaining surface water flow during dry periods;
- maintaining the atmosphere by storage of carbon within their plant communities and soil instead of releasing it to the atmosphere as carbon dioxide;
- providing shoreline erosion control and opportunities for recreation;
- providing aesthetic appreciation and natural products for human use at no cost.

Besides these services, coastal wetlands, including mangrove forests are also of regional importance as fish and shrimp nurseries, and of local importance for fishery, agriculture, forestry, and tourism. Threats to these areas are (illegal) mining activities and pollution of water with chemicals such as pesticides, and salt intrusion (UNDP 2013). Some cons are that wetlands can host annoying insects such as disease-carrying mosquitoes, which can sometimes have social, cultural, and economic impacts by limiting community activities (Rey et al. 2012). For the human made systems, the major disadvantages are their relatively large land requirement and

the long period to achieve the optimum treatment efficiency. However, when considering all of their ecological services, they should be promoted as Best Management Practice (BMP) within the farmland (O'Geen et al. 2010).

2.3 CWs for the treatment of agricultural runoff

CWs used for the treatment of agricultural runoff, are man-made engineered systems that have been designed to emphasize specific characters of a natural wetland with the aim of improved treatment efficiency. They are a recommended practice for buffering pollutant source areas and receiving waters. They are an integral part of many landscapes, often serving as transition zones between upland areas and most water bodies (Lange et al. 2011; Locke et al. 2011). Diffuse agricultural pollutants entering wetlands include organic and inorganic chemical contaminants and biological contaminants (Wauchope 1978). Pesticides belong to the organic chemical constituents, together with the organic and inorganic chemicals such as trace elements/metals and salts. CW systems for treatment of agricultural runoff are divided into two main types: 'Surface Flow Constructed Wetlands' (SFCW) and 'Subsurface Flow Constructed Wetlands' (SSFCW).

CWs with surface flow consist of basins or channels, with a suitable medium to support the vegetation and water at a relatively shallow depth of less than 0.6 m, flowing through the basin (Wallace and Knight 2006). At shallow depths and in the presence of vegetation and litter, the wetland exhibits a low water flow. In long, narrow channels, plug-flow conditions may occur. Most removal processes occur in the water column or in the litter layer. This system uses all types of macrophytes: free floating, submerged or emergent macrophytes.

SSFCW are classified according to the direction of the flow to 'Horizontal Flow Constructed Wetlands' (HFCW) and 'Vertical Flow Constructed Wetlands' (VFCW). Figures 2-2 and 2-3, present a schematic view of respectively a HFCW and a VFCW. In HFCW, wastewater is continuously fed at the inlet and flows slowly through the porous medium under the surface of the bed in a horizontal path until it reaches the outlet zone. During this passage, wastewater will come into contact with a network of aerobic, anoxic and anaerobic zones. Aerobic zones occur around roots and rhizomes that leak oxygen into the substrate. The filtration bed is, however, mostly anoxic or even anaerobic (bound and free oxygen are absent). Vertical flow systems usually consist of a filtration bed filled with graded gravel or sand planted with macrophytes. VFCW are fed intermittently with a large volume of wastewater, which then gradually percolates down through the bed and is collected by a drainage network at the base. The bed drains completely free and this allows air to refill the bed (Kadlec and Wallace 2009; Vymazal and Březinová 2015). In general, the depth of the substrate in a SSFCW is restricted to approximately the rooting depth of plants which enables the plants to be in contact with the flowing water.

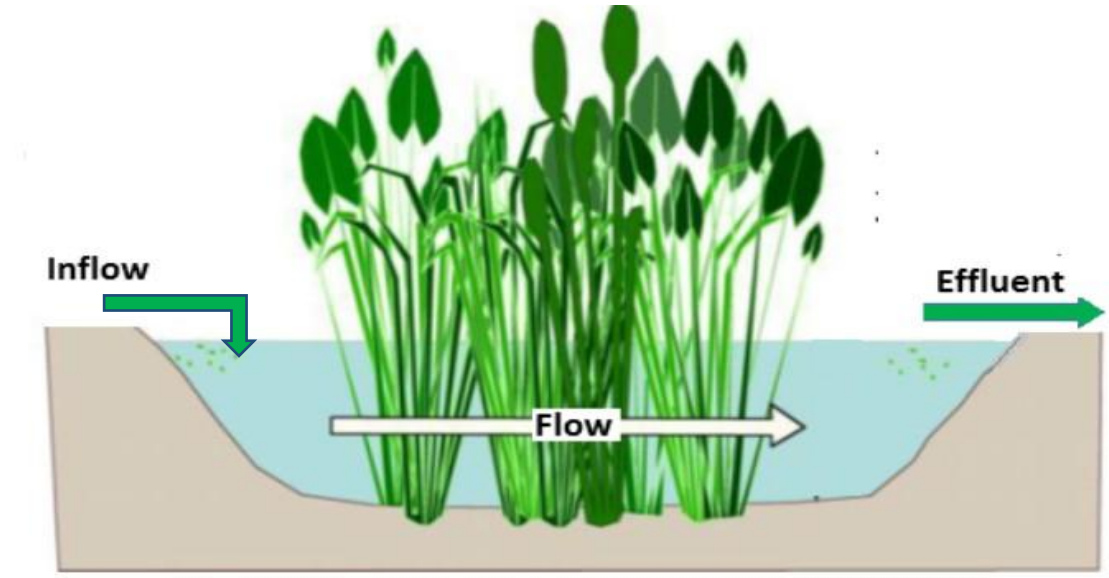

Figure 2-1: Schematic view of a surface flow constructed wetland (SFCW), with incoming wastestream or inflow and the treated water or effluent (Oki and White 2012)

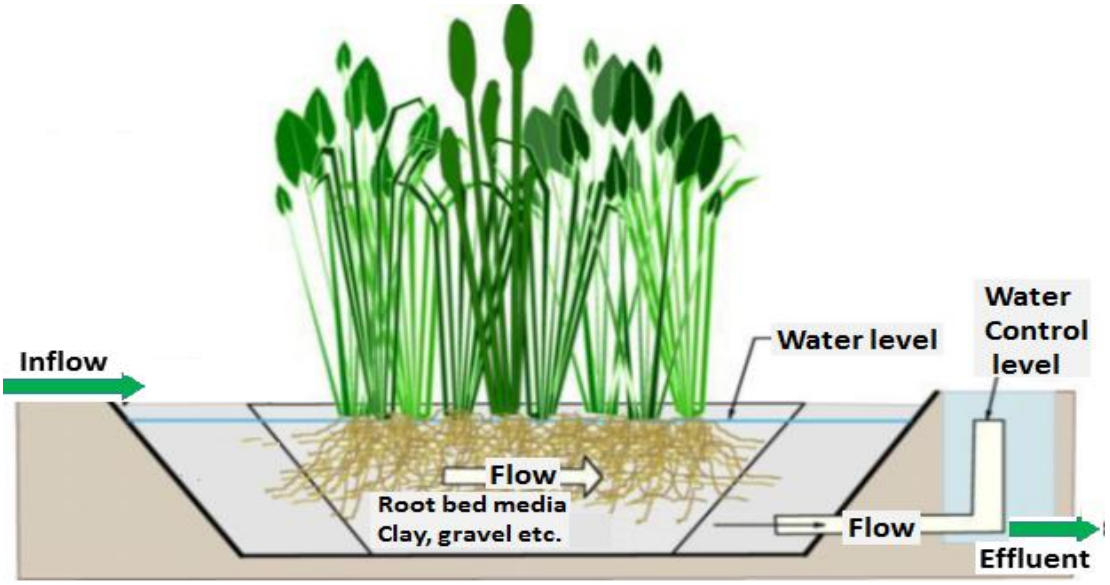

Figure 2-2: Schematic view of a horizontal subsurface flow constructed wetland (HFCW) with different possibilities for root bed matrix, a water control structure, the incoming wastestream or inflow and the treated water or effluent (Oki and White 2012)

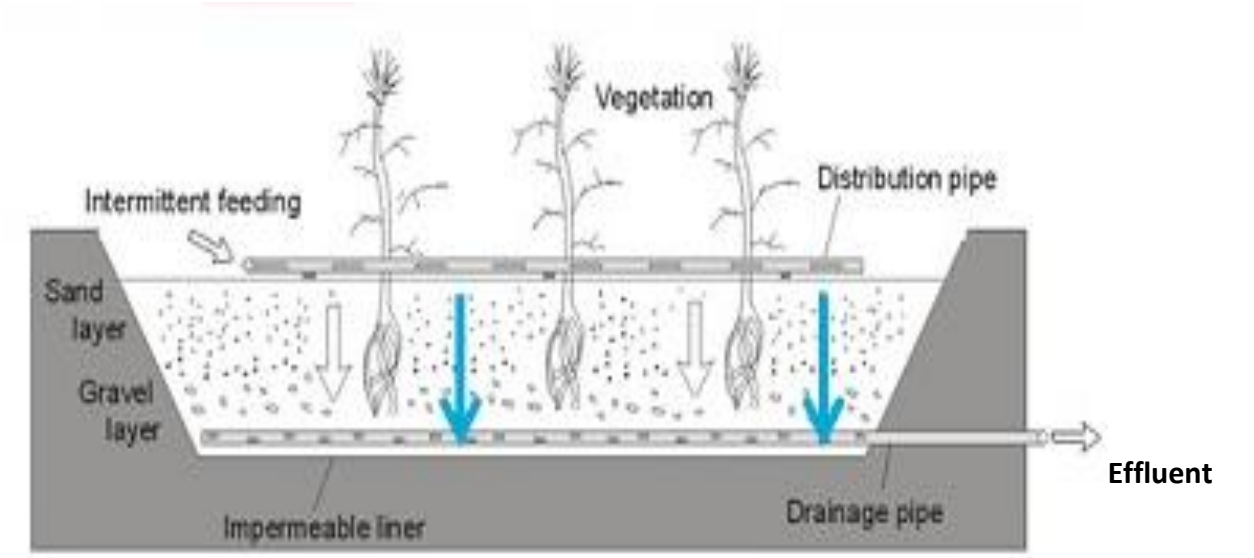

Figure 2-3: Schematic view of a vertical subsurface flow constructed wetland (VFCW) with different possibilities for root bed matrix, the incoming wastestream represented by the intermittent feeding and the treated water or effluent (Langergraber 2008)

SSFCW are not common in agricultural settings because of the high maintenance costs associated with the clogging of porous media (Laurent et al. 2015; Liu et al. 2015). This clogging arises mostly from a high amount of particulate matter in runoff. The vertical subsurface flow constructed wetland (VSSFCW: Figure 2-3) exhibits a good oxygen transfer and increases the ability to nitrify, a process which is limited in a HSSFCW (Vymazal 2010). A third system is called a hybrid CW. In that system, various types of CWs are sequentially combined. The most common one is a combination between HFCW and VFCW (Vymazal 2013A; Vymazal and Březinová 2015).

Current research perspectives focus on the combination of different CWs types or the integration of other (e.g. conventional) treatment systems with CWs (Araña et al. 2008; Wang et al. 2014; Wu et al. 2014). Possible combinations are presented in Figure 2-4. A combination of a membrane bioreactor and a VFCW was used to treat synthetic municipal wastewater. That combination, in particular the membrane reactor, prevents the clogging of the CWs because of the removal of particulates prior to entering the CW (Liu et al. 2015).

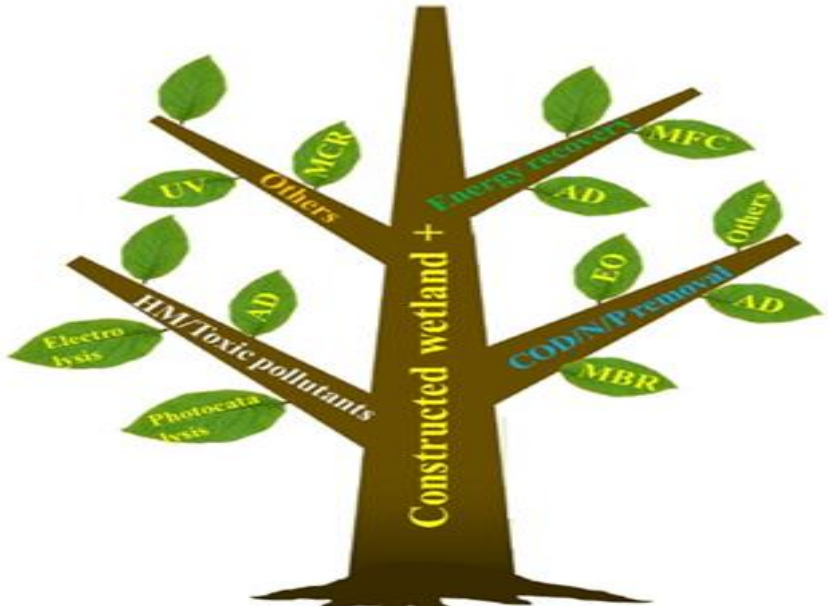

Figure 2-4: Integrations of constructed wetlands (CWs) with other treatment systems directed to either removal of heavy metals (HM) and/or toxic pollutants, COD/N/P removal, energy recovery or others (such as pathogen removal). Treatment systems/processes include: Membrane Chemical Reactor (MCR), Electrochemical Oxidation (EO), Membrane Bioreactor (MBR), Microbial Fuel Cell (MFC) and Anaerobic Digester (AD) (Liu et al. 2015)

A special need in agriculture is the removal of pathogens from agricultural waste streams or other water bodies (Figure 2-5) with the aim to reuse the treated water for irrigation. For that purpose, CWs and the use of UV light or Ozonation/Chlorination have been combined. However, most of the combined systems integrating CWs are still under investigation and might be a problem for implementation in developing countries because of high operational and management costs.

Other challenges are to evaluate the removal of pharmaceuticals and personal care products (PPCPs) (Ávila and García 2016; Hijosa-Valsero et al. 2016) as well as nanomaterials (Adeleye et al. 2016) in CWs.

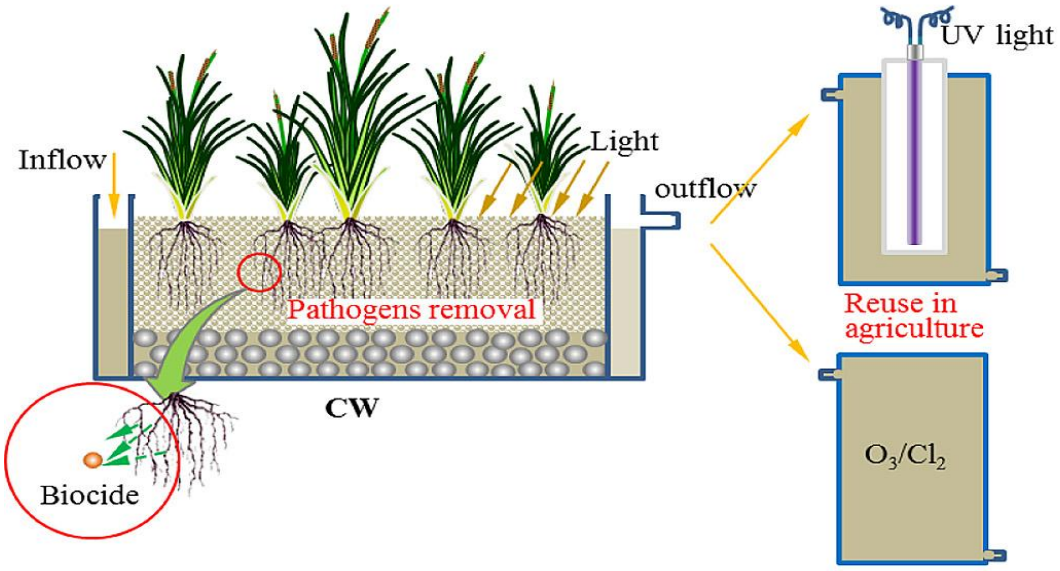

Figure 2-5: Treatment system for the removal of pathogens from wastewater by combining or 'integrating' a constructed wetland and a disinfection step (use of UV light or ozonation (use of O_3)/chlorination (use of Cl_2)). Biocides are defined here as root excretions of wetland vegetation, capable to kill pathogens (Liu et al. 2015)

2.4 Factors influencing the performance of constructed wetlands

The prevailing factors influencing the performance of constructed wetlands are climate (section 2.4.1) and inflow (section 2.4.2).

2.4.1 Climate

Temperature (T) plays an important role in removal rate of pollutants in CWs, because it influences the biogeochemical reaction rates of some removal processes, especially nitrogen conversion processes. Cold temperatures severely affect microbial processes and the water purification performance of constructed wetlands. Low aeration and oxygen levels are among the limiting factors for a better performance of CWs, mainly because of the die-off of aquatic macrophytes that transport oxygen to their rhizospheres for microbial respiration (Kadlec and Reddy 2001). Subsequently, oxygen dependent removal processes such as nitrification are impaired (Kadlec and Wallace 2009). Results from water quality of 169 full scale VFCW systems, which were in operation in France, for a period of 12 years, show only minor differences in removal efficiency, even for TKN (Total Kjeldahl Nitrogen) which removal is known to be hampered in CWs (Paing et al. 2015). Removal efficiencies for different parameters were compared over two seasons, i.e. summer (mean T, 20 ^{0}C), and winter (mean T, 7 ^{0}C). The main reason for this is that in vertical flow CWs systems, aerobic conditions are maintained due to the intermittent feeding and resting periods, and a good oxygen transfer through the drain aeration system (see Figure 2-3). Temperature is influenced by solar radiation, which also

affects the primary productivity and evapotranspiration, and determines the wetland energy balance. During warm temperatures (e.g. summer), the largest energy gain of CWs is solar radiation and the largest energy loss is evapotranspiration (Kadlec and Wallace 2009).

2.4.2 Inflow

The inflow of CWs receiving agricultural runoff is highly variable in time, because of variation in irrigation and cultivation practices and hydrological patterns. Hydrological patterns strongly influence the biotic community, biogeochemical processes and the fate of pollutants (Zhang et al. 2015). Pollutant removal is often accomplished by manipulating the system's hydraulic and hydrologic conditions and by selecting the appropriate type of (dominant) vegetation (Vymazal 2007; Kadlec and Wallace 2009). Contaminant pulses in the inflow are related to several factors such as timing of pesticide application, crop rotation and drift, processing of the soil. Seasonal patterns in contaminant flux occur as a result of land use, storm events or snow melt, discharge from drainage, and/or irrigation runoff (Brauer et al. 2009; Vymazal 2014). The inflow also serves as part of the wetland water budget, this together with the outflow, rainfall, evapotranspiration and infiltration or seepage (Kadlec and Wallace 2009).

2.5 Removal processes

The major removal processes of contaminants in SFCW are sedimentation and sediment burial, (bio) degradation, filtration, aggregation, hydrolysis and surface adhesion (for suspended solids). Sediment burial is considered to be the major long-term phosphorus storage in wetlands (Reddy et al. 1999). Organic compounds (e.g. pesticides and organic matter) can be removed by deposition, filtration, aerobic and anaerobic microbial degradation and photo degradation. Nitrogen compounds undergo nitrification and denitrification (O'Geen et al. 2010). Removal rates of phosphorus are considered low compared to other constituents and are removed by means of adsorption, absorption, complexation and precipitation. For SSFCW the continuous saturation of the filter bed leads to anoxic/anaerobic processes, especially in the case of heavy loadings, which subsequently limits nitrification, aerobic hydrolysis and the removal of ammonia. The oxygen limited conditions allow biogeochemical transformations such as denitrification and methanogenesis to occur (Vymazal 2014). Because of limited contact between the water column and the soil, precipitation with Al, Fe and Ca-ions is limited. The filter bed (in the case of gravel or crushed rock) has a low adsorption capacity towards phosphorus and thus contributing minimally (Vohla et al. 2011). Phosphorus removal is improved by the use of materials with a high P-adsorption capacity such as minerals with reactive Fe or Al hydroxide or oxide groups such as furnace steel slag (Blanco et al. 2016). A major sink for total suspended solids is adsorption on the biomass film present on the gravel bed of the CW (USEPA 1999). Other important processes are biotic uptake of nutrients, redox processes, pathogen removal, hydrolysis and photochemical degradation. The rate of hydrolysis generally increases with increasing pH (O'Geen et al. 2010). Factors which can influence these processes are the prevailing environmental conditions such as temperature, pH, redox

conditions, oxygen availability, microorganisms and vegetation (Kadlec and Wallace 2009). Microorganisms have a positive influence on pollutants removal, especially in vertical flow systems. Sludge accumulation in these systems increases the hydraulic retention time (HRT) and favors the establishment of a diverse microbial community (Paing et al. 2015).

Major contributions of plants in wetlands are to remove or retain contaminants by means of filtration, assimilation or accumulation and to facilitate sedimentation. They increase the surface area of the substrate, which facilitates microbial attachment, allows oxygen penetration from the atmosphere to the rhizosphere and subsequently promotes mineralization and redox processes. They are capable of UV-light interception which subsequently decreases the amount of pathogens and allows photochemical degradation (Vymazal et al. 2013; Zheng et al. 2015). Dense vegetation present in wetlands increases the hydraulic residence time and affects the removal of pollutants and also reduces sedimentation and the toxicity of pesticides towards aquatic organisms present in the sediment (Elsaesser et al. 2011; Brogan and Relyea 2014; Vallée et al. 2015A).

2.6 Contaminants in agricultural runoff

Agriculture belongs to one of the major contributors of surface water pollution in the United States (USEPA 1994) and the EU (Gregoire et al. 2009). Runoff is the process by which transport of constituents/contaminants (in a dissolved or particulate form) takes place along the surface of sloping agricultural land (Vymazal and Březinová 2015). Figure 2-6 presents the main constituents/contaminants in agricultural runoff according to Rickerts (1993). These include nutrients, pesticides, salts, pathogens, particulate matter, and trace elements (e.g. heavy metals and micronutrients). Nutrients such as nitrogen and phosphorus are natural parts of aquatic ecosystems and have many ecological benefits such as supporting the growth of algae and aquatic plants and providing food for aquatic organisms. However, when the concentrations of these nutrients become too high, the water becomes polluted and this results in euthrophication, which is characterized by algal blooms. These blooms can severely reduce or eliminate oxygen in water, leading to illnesses in and death of fish. Some algal blooms are also harmful to humans (US EPA 2016).
Pesticides belong to the chemical contaminants together with organics (e.g. pharmaceuticals, personal care products (PPCPs) and dissolved organic matter (DOM) and inorganics (e.g. trace elements and salts). They belong to the most important non-point source (NPS) pollutants, causing problems in receiving water bodies.

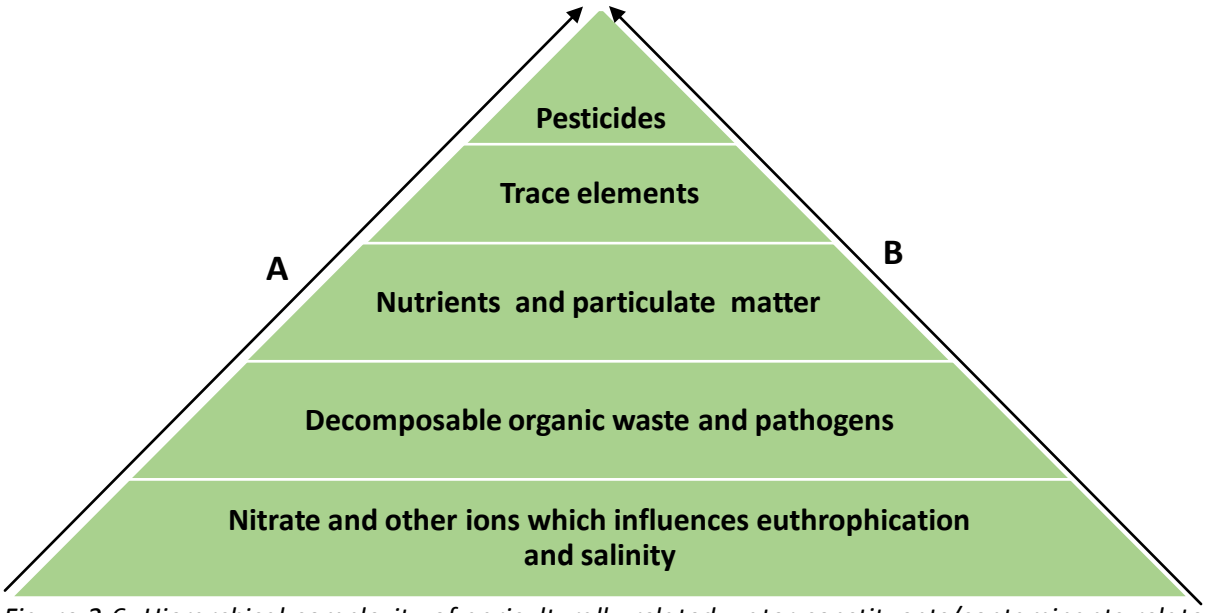

Figure 2-6: Hierarchical complexity of agriculturally-related water constituents/contaminants related to different environmental problems, with A) increasing cost for treatment and B) increasing scientific complexity caused by a decreasing knowledge base (Rickerts 1993)

The costs to society of diffuse water pollution from agriculture can include environmental and ecosystem damage, reduced aquaculture and fisheries income, and increased treatment costs for drinking water (Smith et al. 2015). Agriculture consumes, globally, about 70% of the world water withdrawals, this by means of irrigation. Therefore, any improvement in e.g. waste water removal efficiencies to levels which allow reuse of water for irrigation purposes is valuable. Especially, by using sustainable technologies such as CWs in water-stressed areas (Kundzewicz 2007). Several studies (Gregoire et al. 2009; García et al. 2010; O'Geen et al. 2010; Stehle et al. 2011; Vymazal and Březinová 2015) provide an overview of the main contaminants and removal mechanisms occurring in constructed wetlands used for treatment of agricultural non-point source pollution. It is estimated that worldwide, 100,000 CWs currently treat over billion liters of water per day (Türker et al. 2014). For the main constituents/contaminants e.g. sediment, pesticides and nutrients, studies performed in CWs systems indicate good removal capabilities (Vymazal 2007; Maynard et al. 2009; Stehle et al. 2011; Mitsch et al. 2014; Vymazal and Březinová 2015; Zheng et al. 2015).

2.6.1 Suspended solids

Settling and sedimentation (largest particles), aggregation (flocculation), and interception by surfaces in the water column (e.g. bacteria and colloids) are the main removal mechanisms for suspended solids in CWs. Sedimentation is the physical process of solid particles settling in water. The rate of sedimentation is determined by the particle size, particle density, water

velocity and turbulence, salinity, temperature, and water column depth (Kadlec and Wallace 2009). Sediment is also considered as indirectly toxic, since pollutants (metals, pesticides, and pathogens) are often adsorbed to particulates within sediment, and if desorbed, can become toxic to aquatic organisms. In SFCW, resuspension may occur by means of wind mixing and is influenced by phytoplankton (algae) and animal activity (foraging). In SSFCW, resuspension is minimal, because of the absence of wind, waves and animal induced activities. These include bioturbation or predation of suspended solids by rotifers and other higher organisms grazing on the plankton (Wallace and Knight 2006). Removal of suspended solids in CWs results in improved water quality and increased water clarity. It subsequently increases plant accessibility to sunlight, nutrient retention and carbon sequestration (Mitsch and Gosselink 2015). Carbon sequestration occurs when carbon associated with sediment is buried in anaerobic soil environments and stays preserved because of slow decomposition (Smith et al. 2002). In the study of Mitsch et al. (2014), the results of five separate sedimentation studies during 15 years (following wetland creation in 1994) of two experimental flow-through wetlands with a size of 1 ha are presented. The average sedimentation rate for the two created wetlands as measured by the sedimentation/erosion table (SET) method in 2009-2010 was 5.0 cm/yr with a range from -3.4 cm/yr to 17.2 cm/yr. In agricultural settings, sedimentation rates are highly variable and can be as high as 85 kg m^{-2}/yr (Maynard et al. 2009). The average fluctuation rate or difference between the sedimentation rates (-6.41 cm/yr) was found to be higher than the net sedimentation rate, which indicates that sediment undergoes significant resuspension. The highest rates of sedimentation were found using sediment bottles as collection technique, indicating that the method used for quantifying the amount of sediment is also important. Sedimentation was influenced by erosion, water depth, the presence of emergent vegetation and bioturbation. The effect of particle size is presented in Maynard et al. (2009). In that study, fine sediments (silts and clay) were distributed by means of preferential pathways through wetlands with low hydraulic retention times. This resulted in a decreased removal of particle bound-P, and can also be the case for other contaminants such as pesticides, metals, and pathogens.

2.6.2 Pesticides

Several factors affect loading rates of pesticides to constructed wetlands. They range from soil structure and chemistry, type of irrigation, pesticide formulation, and time of application to rainfall events (Vymazal and Březinová 2015). Chapter 1 presents information about the transport of pesticides into aquatic systems and the different sources. When pesticides enter a CW, several processes (Figure 2-7) may occur, depending on

- pesticide characteristics;
- organic matter content;
- clay content;
- substrate;
- pH;
- redox conditions;

- presence and/or absence of water;
- light penetration;
- retention time;
- inflow;
- pesticide mass;
- presence and type of macrophytes;
- presence of microbial communities;
- type of CWs;
- mode of operation (batch vs. continuous);
- hydrological conditions and
- hydraulic properties of the CW.

(Kadlec and Wallace 2009; O'Geen et al. 2010; Elsayed et al. 2014: 2015; Vallée et al. 2015; Vymazal and Březinová 2015; Maillard et al. 2016).

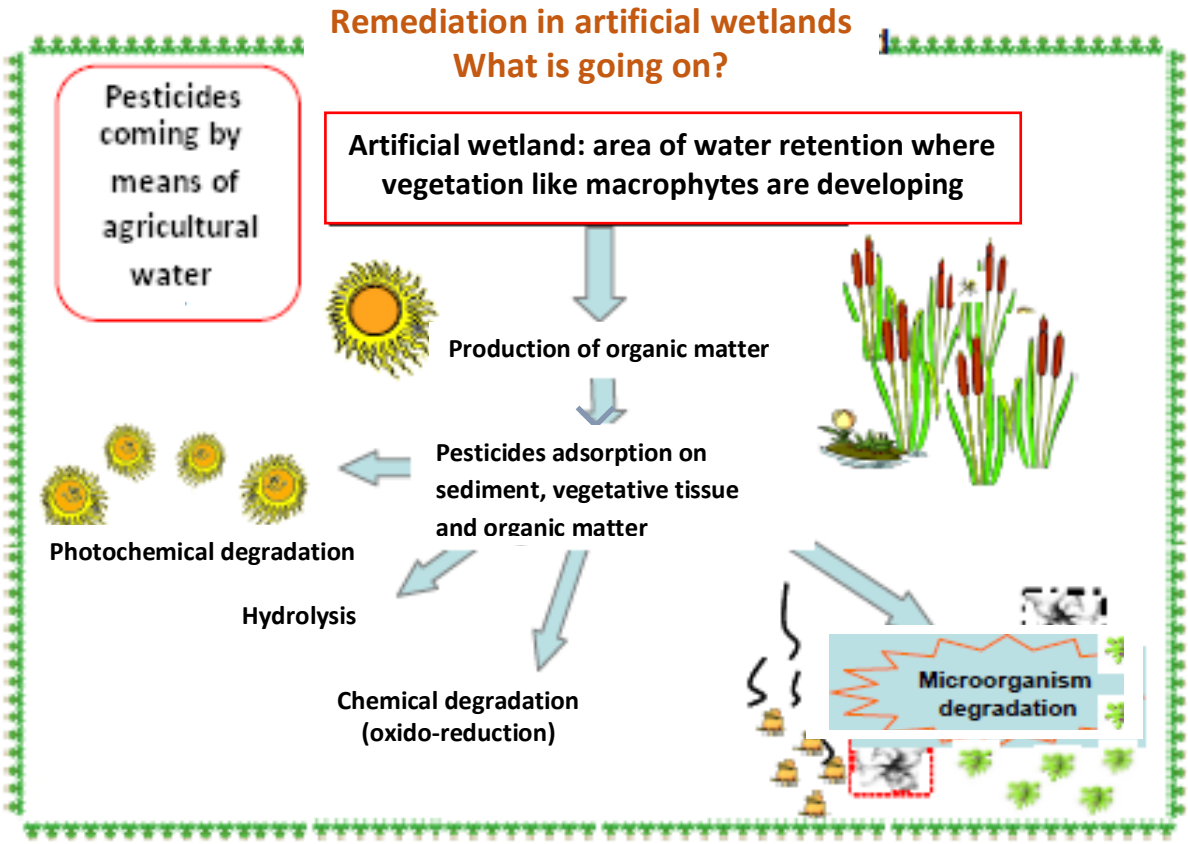

Figure 2-7: Several processes which may occur when pesticides enter a CW (Gregoire 2010)

2.6.2.1 Pesticide fate and removal derived from different wetland studies

Since the 1950s, there is an increasing trend in the number of articles published in the literature on the effective removal of different types of pollutants in CWs systems. The first studies

involving the removal of pesticides in CWs were carried out in the 1970s, but only in the last decade, this became more prominent. One of the first researches performed in the USA, Mississippi assessed the influence of plants on the removal of mevinphos from water. The removal efficiency of this organophosphate pesticide was 100% and 86% in, respectively, vegetated (*N. odorata* and *P. distichum*) and non-vegetated CW mesocosms after 12 days from the start of the experiment. For mesocosms without soil and plants, only 26% of mevinphos was removed (Vymazal and Březinová 2015). Dordio and Carvalho (2013) describe the influence of plants on pesticide removal. Vegetated mesocosms (*P. australis*), in that study, removed between 26.2 and 36.9% more of the herbicide MCPA (2-methyl-4-chlorophenoxyacetic acid) compared to non-vegetated mesocosms. The mesocosm substrate, which consisted of a mixture of gravel and light expended clay aggregates (LECA), also showed a high adsorption capacity, especially of LECA towards MCPA.

Moore et al. (2013) studied the fate and removal of the insecticides diazinon (organophosphate), permethrin (pyrethroid) and the herbicide atrazine after an initial runoff and an additional flushing in vegetated and non-vegetated CW mesocosms. Mesocosms were exposed to concentrations of 20 µg/l of atrazine and diazinon and 10 µg/l of permethrin. The highest load reduction for diazinon was 69 ± 4%, for atrazine 61 ± 7%, for *cis*-permethrin 88 ± 2% and for *trans*-permethrin 89 ± 6%. The *L. oryzoides* mesocosms showed the highest decrease but were not significantly different in removal from the non-vegetated mesocosms. Mass retention and sorption to plants was found to be reversible for atrazine and diazinon. This was derived from the amount of pesticides (%) released after flushing (48-51 h after the start of the experiment). Amounts were found to be between 8-29% for atrazine and diazinon and between 1.0-2.0% for permethrin. The results obtained for permethrin indicate strong sorption, which is related to the high K_{oc} value of permethrin. Appendix 1 provides additional information on the dimensions and other operational parameters under which pesticides were investigated in CWs. These show that pesticide properties such as K_{oc} and solubility in water are decisive in the sorption of a mixture of six pesticides, three herbicides (2-methyl-4-chlorophenoxyacetic acid or 2,4-MCPA, isoproturon (IPU), and napropamide (NPP) and three fungicides (boscalid (BCL), prochloraz (PCZ) and tebuconazole (TBZ) on four substrates (two soils, sediment and straw) found in ponds and ditches in France (Vallée et al. 2014). Adsorption of all types of pesticides was higher on the straw substrate. This was linked to the higher organic content (20 to 30 times higher) compared to that of the sediment and the soils. Retention was greater for pesticides with hydrophobic properties (low solubility and high K_{oc}). The adsorption capacity was in the following order; PCZ >> TBZ-BCL > NPP > MCPA-IPU, with '>>' indicating a much higher adsorption of prochloraz compared to 'TBZ-BCL', 'NPP' and 'MCPA-IPU'. Comparable adsorption capacities were observed between tebuconazole and boscalid (noted as: TBZ-BCL) and between MCPA and isoproturon (MCPA-IPU).

In Elsayed et al. (2015) the concentrations of chloroacetanilide herbicides (rac-metolachlor, acetochlor and alachlor), their ethane sulfonic acid (ESA) and oxanilic acid (OXA) depredates, and enantiomeric fractions of metolachlor were determined in lab-scale wetlands, and related to the hydro-chemical conditions (dissolved oxygen and amount of nutrients) and bacterial

composition. The mean mass removal was 29 ± 19%; 61 ± 14% and 52 ± 12% and dissipation was highly variable and between 8-70%, 56-90% and 32-64%, respectively. Dissipation occurs mainly in the root part, while uptake of pesticides by roots was not significant. Biodegradation of chiral metolachlor resulted in the detection of enantiomeric fractions of metolachlor, namely the ESA (Ethane Sulfonic Acid) and the OXA (oxanillic acid) degradation product. At the outlet (60 cm from inlet), higher amounts of herbicides were found than 55 cm from the inlet. Findings were consistent with lower concentrations of TEA (terminal electron acceptors). This is related to preferential flows and root channelling. Specific microbial communities are found (Elsayed et al. 2014) under different redox conditions, while S-enantiomeric degradation of metolachlor relates to specific enzymes and uptake by proteins. Further information on exposure concentrations and system dimensions are given in Appendix 1.

The influence of batch versus continuous mode of operation of SSFCW on pesticides and tracers' fate is investigated in Maillard et al. (2016). Wetlands were exposed to 960 g/l S-MET (87% of the *S*-enantiomer of Metolachlor) during two time intervals of 14 days with water amended with S-Met and tracers (Bromide, Uranine and Sulphorodhamine B). In between a 14 days interval, without pesticide, only water was monitored. For the batch mode: 4 flood-drain cycles of 14 days of saturated conditions with the *S*-MET-tracers mix were applied, followed by a 7 days drainage for each cycle. For the batch mode, DO concentrations (and redox potential) were 3.1 ± 1.8 mg/L (320mV) at the start of the experiment and dropped to <0.02 mg/l (<−400 mV) following drainage. Lower nitrate and higher manganese (II) concentrations at the outlet of the continuous-flow wetland indicated a reduction of TEAs, which was not observed for the batch-mode wetland. Removal of S-MET was high and constant in the batch operated wetland (93-97%), while in the continuous-flow mode the removal varied highly (40-79%). The higher removal obtained for the batch mode related to a higher aerobic degradation caused by oxygen replenishment during the dry operations (drain cycle). The uptake by vegetation was low for both types of CWs. However, it was more pronounced, e.g. less than 5% (62.5 mg) of the total supplied amount of S-MET, for the batch operated wetland compared to, below the LOD (limit of detection), the continuous mode CWs. Different metabolites were formed during different modes of operation. The tracer mass budget revealed that plant uptake, sorption, photo- and biodegradation (the main dissipation pathway) were prominent under batch mode and were in agreement with the dissipation of S-MET (around 90%) under the operating conditions.

Different studies (Bouldin et al. 2005; Budd et al. 2009; Budd et al. 2011; Moore et al. 2013 and Lizotte et al. 2014) assess the fate and removal of pyrethroids (lambda-cyhalothrin, *cis*-permethrin and *trans*-permethrin) in CWs (see Appendix 1). They are discussed in the next chapters on wetland mesocosms and field experiments. In general, these studies show that the major pathway in CWs systems is through sedimentation and uptake by vegetation. Retention (mass uptake by vegetation or sediment) varies between 34% (Budd et al. 2009) up to 100% (Lizotte et al. 2014), with the lowest retention being related to a non-vegetated system. The hydrophobicity and low solubility of pyrethroids are the main reasons for a rapid dissipation from the water phase in CWs. Sorption to soil particles are also related to the high K_{oc} values and to the particle size. Bouldin et al. (2005) studied the influence of a mixture of pesticides

compared to a treatment with only one type of pesticide. The results show a higher uptake of lambda-cyhalothrin for mesocosms amended with atrazine, while amendment of mesocosms with nutrients (Lizotte et al. 2014) does not influence the removal of permethrin and results in comparable retentions of cis- and trans-permethrin.

2.6.2.2 Pesticide properties

Different classes of pesticides investigated in CWs systems are mostly limited to herbicides and insecticides (e.g. organophosphates and pyrethroids). They experience different fate and removal mechanisms in CWs. These mechanisms are mostly related to their physico-chemical properties such as solubility, octanol-water (K_{ow}) and organic-carbon (K_{oc}) partitioning coefficient, vapor pressure, their persistence (chemical half-life) in water and soil, and their vulnerability towards photolysis (Gregoire et al., 2009; Stehle et al. 2011; Tournebize et al., 2013). According to Vymazal and Březinová (2015), many factors influence the occurrences of processes in CWs. Some of these factors are the organic matter content, clay content, filtration material quality, pH, redox conditions, the presence and/or absence of water, retention time, pesticide mass in the inflow, the presence and type of macrophytes, or type of CWs. Vymazal and Březinová (2015) reviewed the removal of pesticides in agricultural runoff and drainage in 47 wetland studies, involving the removal of 87 pesticides. One of their main findings was that mostly free water surface wetlands (SFCW) were used and that pesticide removal of individual pesticides was highly variable. Regression analysis between the removal efficiency and log K_{oc} resulted in a very low R^2 (correlation coefficient). However, when pesticides were grouped, the highest average removal was found for the pesticides with a very low solubility, very high K_{ow} and high K_{oc} (pyrethroids and strobilurin). Findings based on average removal efficiencies were 97% for pesticides from the organochlorine group (namely endosulfan and pentachlorophenol), followed by 96% for pesticides from the strobilurin/strobin group (kresoxim methyl, trifloxystrobin and azoxystrobin), 94% for the organophosphate pesticides (azinophos methyl, diazinon, dimethoate, glufosinate, chlorpyrifos, methyl parathion, mevinphos, omethoate, parathion, prothiofos) and 84% for pyrethroids (bifenthrin, cyhalothrin, cypermethrin, esfenvalerate, permethrin). The lowest removal efficiencies were 24% for triazinone pesticides (metamitron, metribuzin), 35% for the aryloxyalkanoic acid group (dichlorprop, MCPA, mecoprop) and 50% for urea-based pesticides (diuron, fluorometuron, chlorotoluron, isoproturon, and linuron). For these poorly removed pesticides, results did not show any clear relationship between removal and solubility, K_{ow} or K_{oc}. Values for these parameters were highly variable. In the review of O'Geen et al. (2010), which focused on the removal efficiencies of 27 pesticides (12 organophosphates and 15 herbicides) it was observed, with one exception, that log K_{ow} values of less than 4.2 resulted in more than 50% reduction in pesticide concentrations. This indicates that sorption is the primary driving force of removal for highly hydrophobic chemicals. Sorption leads to contaminant removal by making the contaminant less reactive or by removing it from the system (water phase) through sedimentation and burial. Sorption is limited by the amount of sorptive surfaces, the chemical and mineralogical nature of the particles and the nature (e.g. charge) of contaminants. For pesticides with log K_{ow} values between 1 and 4, there are large variations in the removal efficiencies, indicating that chemicals

in this range are removed by wetlands, but that the wetland performance depends on system characteristics. Pesticides with log K_{ow} < 1 and low water solubility, lower removal efficiencies are found. Between the different pesticides groups, the organophosphates and pyrethroids have the highest removal, while for herbicides the high variability in data did not allow to observe a specific trend in removal.

2.6.2.3 Role of vegetation

Pesticide uptake and decontamination by plants is a biological/microbiological process, which occurs predominantly in the rhizosphere. Several mechanisms of pesticide degradation are still unknown (Crowly et al. 1997; Mc Kinlay and Kasperek 1999). The reviews of Vymazal (2011; 2013c) provide a detailed overview of the role of plants in horizontal flow constructed wetlands and in surface flow constructed wetlands. Vymazal (2013c) gives an overview of the different types of plants used in different countries. The metabolic fate of pesticides depends on abiotic environmental conditions (pH, moisture, soil and temperature), microbial community, plant species, pesticide characteristics and biological and chemical reactions (Van Eerd et al. 2003). Pesticide metabolism in plants involves three phases. They are: oxidation, hydrolysis and reduction (phase 1), further detoxification of phase 1 metabolites into products, which are less mobile and toxic (phase 2) and conjugation and further detoxification of phase 2 metabolites to non-toxic and immobile biopolymers (phase 3) (Van Eerd et al. 2003; Yuan et al. 2007). Plants also degrade the products of pesticides transformed by microorganisms (Weinberger and Bollag (1972) and Cerniglia (1992) and they bind pesticides to their cell walls. That binding results in bound pesticide residues, which makes pesticides difficult to extract and to hydrolyse. Rose et al. (2008) successfully calibrated a graphical model to field data and described pesticide loss from pounded aquatic systems. The results show that aquatic plants enhanced sedimentation, thus directly contribute to the removal of sediment-bound pesticides, and accelerate biofilm contact and photolysis. By means of the removal of suspended solids, which are responsible for the uptake and scattering of light, more UV light will be perceived, resulting in pesticides photolysis. Pesticide (fluometuron) removal was found to be faster in a vegetated pond (60% of the initial mass load (5.6 µg/l) removed) than in an open pond (30% of the initial mass load removed) after 10 days of operation. Vegetation also influenced the hydrology of wetlands systems, especially in channelized aquatic environments such as ditches. Vegetation within the channel exerted roughness, drag and friction on flowing water and reduced the flow rate, increased the water depth and hydraulic retention time (HRT) and subsequently the pesticide residence time (PRT) and pesticide removal (Gregoire 2010). Plants also bio-accumulate pesticides. However this amount is highly variable. For instance, in Moore et al. (2009a) it was found that 49% of the insecticide lambda-cyhalothrin was associated with vegetation (mix of vegetation), while in Mahabali and Spanoghe (2014) this was about 1% in both *N. amazonum* and *E. mutata* plants. Systemic herbicides are more capable of being removed through plant uptake and rhizosphere sorption (Vymazal and Březinová 2015). The accumulation of pesticides in wetland plants is observed in several earlier conducted laboratory studies (Hinman and Klaine 1992; Feurtel-Mazel et al. 1996; Crum et al. 1999). The accumulation is found to be based on the type, the concentration and the solubility of the pesticides in relation to sorption.

In a more recent study (Guo et al. 2014), it was observed that less hydrophobic (low K_{OW}) organochlorine pesticides were more easily accumulated and transported in the tissues of *Phragmites australis*, *Typha sp*. and *Ceratophyllum demersum*.
Another important point of view is presented in Maillard and Imfeld (2014), in which the partitioning, retention and degradation is described, based on a mass budget of 12 pesticides in the water phase of a wetland, receiving contaminated runoff. The authors conclude that throughout the year different storage compartments and dissipation processes prevail. During spring, bed sediment and plants are the prevailing storage compartments and plant uptake the main dissipation process, while during summer and late summer these compartments are water, suspended solids and bed sediment and the main processes, biodegradation and sorption to bed sediment.

2.6.2.4 Pesticide sorption and degradation

Pesticide degradation in CWs mainly takes place by photolytic and microbial degradation, while other mechanisms such as plant uptake and sorption also occur. Sorption includes both adsorption and absorption. Adsorption can be physical or chemical, with physical adsorption belonging to the reversible type of adsorption, because the pesticide is weakly bound to the adsorbent. Reaction kinetics is usually in the order of minutes to hours before a sorption equilibrium is established (Reddy and DeLaune 2008). The adsorption potential of a pesticide is known to depend on the properties of both the pesticide (e.g. solubility and log K_{oc}) and the sorbent phase. Several types of adsorbents have been used such as soil, straw, sediment and furnace steel slag (Vallée et al. 2014; Blanco et al. 2016). For hydrophobic chemicals, sorption to soil or sediment is influenced not only by the quantity of organic matter, but also by the binding characteristics of the organic matter (O'Geen et al. 2010; Vallée et al. 2014). Adsorption belongs to one of the main processes of pesticide removal in CWs. However, in horizontal flow constructed wetlands in which washed gravel of crushed rock (filtration media with low organic matter content) is mostly used, adsorption is limited. Adsorption occurs mainly in older mature systems where organic matter concentration increases due to sedimentation of suspended solids and formation on biofilms (Vymazal and Březinová 2015). Pesticide sorption by plants is also a main mechanism as indicated by several studies (Dordio and Carvalho 2013; Moore et al. 2013; Guo et al. 2014; Maillard and Imfeld 2014; Vallee et al. 2014).

Repetitive exposure to the same pesticides over time, due to induction and adaptation of microbes, leads to the establishment of organisms capable to enhance the degradation of pesticides in wetlands. Sometimes bio-augmentation or enrichment with microorganisms is needed to enhance microbial degradation (Kadlec and Wallace 2009; O'Geen et al. 2010). Runes et al. (2001) showed that atrazine degradation in wetland sediment is enhanced by means of mixing of sediment soil with soil from an atrazine spill site known to contain atrazine mineralizing bacteria. Racke and Coats (1988studied degradation of organophosphorus insecticides in soil enriched with microorganisms with a laboratory experiment. In that study, however, degradation is only enhanced for one organophosphate. Ahmad et al. (2012) grew ryegrass in soil spiked with chlorpyrifos (CPF) and inoculated with a pesticide degrading

bacterial strain. The highest CPF degradation was 97% and, compared to non-inoculated plants, significantly less CPF accumulation was observed in roots and shoots of inoculated plants. Enhanced degradation occurred when a population of soil microorganisms, which is adapted due to previous exposure to a pesticide, rapidly degraded a subsequent application of the pesticide (Felsot et al. 1982; Racke and Coats, 1988). Microbial degradation of pesticides was negatively affected by strong (irreversible) bindings (e.g. sorption) of pesticides or by wetland aging (O'Geen et al. 2010).

2.6.3 Nutrients

Wetland plants are very productive and considerable amounts of nutrients and other chemicals can be bound in their biomass (Vymazal 2011; Vymazal and Březinová 2016A). Nutrients are of concern because of their link to eutrophication and hypoxia within aquatic systems. CWs often show limited capacity for nutrient (especially phosphorous) reduction (Vymazal 2007). Agricultural N fertilizer is the single largest source of reactive N (e.g. ammonia, ammonium, nitrate, nitrite, organic N etc.) in the world and can be responsible for eutrophication and hypoxia of waterways, as well as loss of biodiversity, habitat degradation, shifts in food chain structure and fisheries impairment (Moore et al. 2010). Forms of nitrogen in irrigated agricultural runoff include nitrate, ammonium and organic N (dissolved and particulate). However, the dominant form of N in CWs that is received by agricultural runoff is nitrate (O'Geen et al. 2010). As mentioned in section 2.5, N-compounds in CWs undergo several processes. They include nitrification (limited in SSFCW), denitrification, plant assimilation, sedimentation and burial of particulate N and ammonia volatilization, with denitrification being the most dominant process (Kadlec and Knight 1996). In SFCW nitrogen removal occurs primarily through nitrification (in the water column) and subsequently by means of denitrification (in the litter layer) and ammonia volatilization. Volatilization occurs under higher pH values caused by algal photosynthesis (Vymazal 2010). In SFCW, plant uptake is considered as the primary mechanism for nitrogen reduction (Vymazal, 2007). The vegetation is usually not harvested and the litter provides organic carbon necessary for denitrification, which may proceed in anaerobic zones within the litter layer. Nitrogen removal in SSFCW is affected by the HRT, temperature, vegetation type and properties of the soil medium. Intermittent feeding leads to an increased transfer of oxygen and a higher NH_4-N removal in VSSFCW compared to HSSFCW (Zhang et al. 2015).

Phosphorus (P) entering wetlands is typically present in both organic and inorganic forms that are either dissolved (<0.45 mm) or particulate (>0.45 mm). In most agricultural soils, 50-75% of P is inorganic, with 60-90% of P transported from cultivated fields as particulate phosphorus (PP), which is not readily bioavailable such as dissolved inorganic phosphorus (DIP) (O'Geen et al. 2010). Phosphorus is removed at relatively slow rates by means of adsorption, absorption, complexation and precipitation. Phosphorus (P) removal in CWs occurs through a combination of several processes: peat/soil accretion (vertical increase in the elevation of the soil surface; (Reed 1995), plant uptake, microbial growth, substrate adsorption and chemical precipitation. From these processes, adsorption and chemical precipitation play the largest role, particularly

in saturated subsurface flow CWs (SSFCW), where the contact between wastewater and substrate is enhanced (Vymazal 2007; Blanco et al. 2016). Phosphorus removal in FWSCW is variable and is largely dependent on both the hydraulic loading rate (HLR) and size of systems (Braskerud et al. 2005). CWs with subsurface flow (SSFCW) have a major potential for phosphorus removal, and among those systems, horizontal SSFCW (HSSFCW) have an even higher potential as the substrate is constantly flooded and there is little fluctuation in redox potential in the bed (Vymazal, 2007). A high Al or Fe content provides effective phosphorus adsorption. Phosphorus in SSFCW is removed primarily by ligand exchange reactions, whereby phosphate displaces water or hydroxyls from the surface of Fe and Al hydrous oxides (Davies and Cottingham 1993). Other factors which influence P removal are vegetation, types of substrate and influent loadings. For example, chronic high nutrient loadings can reduce the capacity of a wetland to store P, because the sediment at the inflow becomes saturated.

In Ghermandi et al. (2007) the performance of 38 tertiary treatment wetlands worldwide using SFCW was assessed. On average, these CWs removed 75.3% NO_3-N, 62.31% total nitrogen (TN), 68.54% (range 28%-96%) NH_4-N and 47.92% (range 13%-75%) total P (TP). The ranges indicate the high variability in removal efficiencies for NH_4-N and TP. In a review of 25 CWs worldwide in tropical and subtropical regions, Zhang et al. (2015) found that removal efficiencies of TN in SSFCW generally ranged from 40 up to 55%. From these types of wetlands the vertical subsurface flow CWs (VSSFCW) have been reported to remove NH_4-N (66.02%) slightly ($p > 0.05$) more efficiently than do horizontal subsurface flow CWs (62.57%). Low removal efficiencies of NO_3-N were found in HSSF (42.46%), compared to VSSFCW (73.33%), while higher TP removal efficiencies (69.75%) were found for HSSFCW when compared to VSSFCW (60.08%). Based on the different performances toward N-removal and to their inability to provide both aerobic and anaerobic conditions simultaneously, single-stage CWs cannot achieve a high removal of total N (Vymazal 2007). In general, HSSFCW can provide good conditions for denitrification. The ability to nitrify ammonia is, however, very limited. In contrast, vertical SSFCW (VSSFCW) can remove NH_4-N successfully, but denitrification hardly takes place in these systems. To deal with this problem it is advisable to use a combination of systems e.g. a hybrid or multi-stage CWs system. The most frequently used are VSSFCW-HSSFCW combinations. The average efficiencies of 11 hybrid systems shown in Zhang et al. (2015) were: 80.71% for NH_4-N, 80.76% for NO_3-N and 75.41% for total nitrogen (TN) (Zhang et al. 2015).

In the two years study of Zheng et al. (2016), involving a SFCW and a SSFCW, the nitrogen uptake in the first year by the aboveground parts of *P. australis* and *T. orientalis* in the SFCW were 31.7 g N/m^2 and 20.8 g N/m^2, respectively. That value increased to 80.0 g N/m^2 and 37.6 g N/m^2, respectively in the second year. The phosphorus uptake rates increased from 3.0 g P/m^2 and 2.5 g P/m^2, respectively, in the first year to 7.3 g P/m^2 and 4.2 g P/m^2, respectively, in the second year. In the SSFCW, however, the proportion attributable to plants for TN removal was decreased from 6.2% to 5.8%, while the proportion attributable to plants for TP removal was increased from 4.6% to 4.8%. This was related to the reproduction of plants in the SSF wetland and an enhanced nitrification-denitrification process around the rhizosphere of the plants. The sorption capacity of gravel towards P decreased, because of wetland aging (Kadlec and Wallace

2009; Hallin et al. 2015; Zheng et al. 2016). All these processes are temperature dependent and are enhanced in tropical regions compared to temperate regions e.g. ammonia volatilization increases 1.3-3.5 times with each 10°C rise in temperature from 0°C to 30°C, and denitrification rates almost double (1.5-2.0) with each 10°C increment (Zhang et al. 2015). Another importance consideration is presented in Vymazal (2016). In that study it was stated that the concentration of nitrogen and phosphorus is always higher in leaves than in stems. However, when the stem biomass (often much higher in robust emergent species such as *Phragmites australis*) is higher than that of leaves, more nutrients can be accumulated in stems and vice versa.

2.6.4 Trace metals

According to O'Geen et al. (2010), the term trace metal is often interchanged with micronutrients, microelements, and heavy metals; although not all trace metals are heavy metals. Toxic trace elements can include metals that have a toxic effect on species. They are naturally occurring and at low concentrations in soils and water. Examples of common metal pollutants in aquatic ecosystems are lead, cadmium, mercury, chromium, nickel and arsenic. These are metals of concern because of their toxic properties that induce adverse effects in humans and aquatic organisms (Duruibe et al., 2007). Compared to other pollutants trace metals are especially of concern because they are less mobile, they do not degrade and can accumulate in soil. Concentrations can increase incrementally each year and can become elevated above background levels and threaten plant, animal, and environmental health. Erosion of metal-rich soil particles via surface runoff can threaten surface water quality, contaminate river sediment, and lead to long-range transport of associated trace metals (Kadlec and Wallace 2009; O'Geen et al. 2010). The removal efficiency of trace metals in constructed wetlands is difficult to predict, because it is highly variable and dependent on factors such as plant species and the target trace element (Kara 2005). High concentrations of heavy metals can be phytotoxic to wetland vegetation and subsequently cause a decrease in pesticide removal.

Historical use of pesticides and application of biosolids, including sewage sludge and animal manures, has increased trace metal content in agricultural soils. The levels of trace metals in sewage sludge are typically higher than animal manures and can vary greatly depending on the source and treatment (He et al. 2004). The two most important factors controlling trace metal speciation in soils, sediments, and wetlands are redox potential and pH (Reddy and DeLaune 2008; Du Laing et al. 2009). The solubility of trace metals increases under reducing conditions or low pH, as can be derived from the different Pourbaix (Eh-pH) diagrams of specific metals. Methylation of mercury (biotic and abiotic) and arsenic occurs also in low soil redox conditions, while with high salinity and high sulphate loads in combination with reduced environments, mobility of trace metals increases and decreases respectively due to the formation of soluble chloride complexes and sulfide precipitations. Formation of methyl mercury results in its bioaccumulation in the food chain and amounts may increase up to unacceptable and toxic levels and affect the health of humans e.g. by consumption of contaminated fish (Driscoll et al. 2013). Also of importance are the reduced forms of iron and manganese. When oxidized, the

reduced form is transformed into amorphous hydrous oxides, which have a large surface area, suitable to function as a sorbent for trace- or heavy metals (Reddy and DeLaune 2008).

Wetland soils are generally characterized as having reducing conditions and high levels of organic matter, serving as a source of ligands to bind trace metals and to facilitate microbial transformations in the soil. The most important processes occurring are sedimentation/precipitation, sorption and complexation. Sedimentation and sorption are responsible for the metals to settle and to become sequestered in soils, while binding occurs to clay minerals, metal oxides and organic matter fractions (Reddy and DeLaune 2008). The release of trace metals within wetlands occurs primarily via organic matter decomposition or microbial catalyzed reduction of Mn and Fe-oxides. Micro-organisms play an important role in the redox processes and in the speciation of trace metals in soils and aquatic environments. Examples are the conversion of inorganic mercury into methyl mercury by anaerobic bacteria and the influence of sulphate reducing bacteria (SRB) to facilitate the precipitation from the water phase and subsequent sequestration of metals (O'Geen et at. 2010).

The use of wetland plants such as *Typha latifolia, Typha angusfolia* and *Phragmites australis* have been demonstrated as an effective method to remove a range of metals (e.g., Cd, Cu, Pb, Zn, Hg, Se) from wetland ecosystems (Liu et al. 2007; Rai 2009; Ali et al. 2013). Different phytoremediation mechanisms have been observed for plants in CWs such as accumulation, dissipation, immobilization, cation exchange, and root induces chemical changes as well as serving as carbon source for bacterial metabolism or degradation (Rai et al. 2015). Upadhyay et al. (2016) showed that a moderate to high accumulation of metals was achieved in a three staged horizontal flow wetland systems, by two macrophytes (*P. crispus* and *H. verticillata*). The highest accumulation in the tissue of *P. crispus*, was obtained for Mn (86.36 µg/g DW), followed by Cr (54.16 µg/g DW), Pb (31.56 µg/g DW), Zn (28.06 µg/g DW) and Cu (25.76 µg/g DW). For *H. verticillata*, the obtained values were: Zn (45.29), Mn (42.64), Pb (22.62), Cu (18.09) and Cr (16.31 36 µg/g DW). In that study, plants were harvested to prevent metals from re-entering the wetland. Recently, the green algae *Chlorella sp*. was reported to remove and transfer trace elements from biogas fluid (Yan et al. 2014 and Yan and Zheng 2014).

2.6.5 Pathogens

The effluent in advanced water treatment systems is mostly disinfected by using chemicals such as chlorine. This, however, raises health related concerns because of the formation of by-products such as trihalomethanes. Therefore, attention has been shifted to test the capacity for human pathogen removal in eco-sustainable systems such as constructed wetlands (Toscano et al. 2013). Microbial indicators for fecal contamination in the wastewater sources are quantified to evaluate pathogen removal performances of wastewater treatment and sanitation processes, and typically include *Escherichia coli* (*E. coli*), total coliforms (TC) and fecal coliforms (FC), fecal streptococci (FS) (=fecal enterococci), sometimes staphylococci (ST), and *Clostridium perfringens* (CLP), which spores are considered conservative surrogates for *C. parvum* and *Giardia lamblia* (oo)cysts (Wu et al. 2016). Sources of these pathogens are diverse as they are

shed in the feces of wildlife, humans, livestock, and pets. According to Cooley et al. (2007), nonpoint sources have become the primary source of microbial pathogens in waterways, with agricultural activities being the single largest contributor. Pachepsky et al. (2011) states that there is sufficient data available on indicator organisms in irrigation water. However, no data exist on a comprehensive study of pathogens in irrigation water, mainly because of high costs for extensive sampling and the cultivation of human pathogens, which has to occur in designated laboratories.

Different removal efficiencies are reported in the literature. For instance Kadlec and Wallace (2009) found removal efficiencies of pathogens above 70% in SFCW, primarily bacteria and viruses from waste water streams. In SFCW, retentions of 80-99% have been seen for pathogen indicators such as *E. coli* and fecal coliforms from municipal and livestock wastewater, while scarce information is available for removal of pathogens in agricultural runoff (Díaz et al. 2010; O'Geen et al. 2010). Physical removal mechanisms for pathogens include filtration, sedimentation, soil and biofilm adsorption and aggregation. Biological elimination mechanisms include predation (protozoa and/or viral), bacteriophage activity, lytic bacteria, release of antibiotics by plants and other microbes, and natural death. Chemical elimination mechanisms include oxidative damage, UV irradiation, and toxins excreted by other bacteria and plants. Environmental factors such as pH, sunlight, temperature, vegetation type and density, and redox potential play a role in pathogen survival and elimination (O'Geen et al. 2010; Pachepsky et al. 2011). The reduction of pathogens in SFCW has shown to be critically dependent on internal flow patterns in the wetland, sunlight intensity and exposure time (Mayo 2004). Small elements of flow (hydraulic short circuiting) can carry enough organisms in the outlet of the wetland without treatment. In general, first order models forecast a decline of pathogens in the wetland if: a) the incoming levels exceed the regrowth and b) reintroduction of pathogens is minimal. For the coliforms, many organisms in this broad group are not limited to fecal sources. SFCW eliminates high numbers of these organisms, with reductions between 1 to 2 log10. *Escherichia coli* are usually considered harmless. However, several strains are able to cause gastroenteritis and other dangerous diseases. The most affected are the elderly, and therefore *E. coli* has found favor over other indicator organisms. Problems observed in CWs are regrowth and reintroduction, while large reductions (up to 3.77 log10) may occur when dealing with a high inflow concentration e.g. for an inlet concentration of 248,172 CFU/100ml the reduction was 3.77 log10 units, while for 150 CFU/100 ml a 0.17 log10 reduction was found (Kadlec and Wallace 2009). In SSFCW, fecal coliform removal from 130 wetlands was on average 1.82 log10, total coliforms 2.04 log10 (average of 54 systems) and *E. coli* 2.53 log10 (average of 44 systems) and is enhanced under longer HRT and lower hydraulic loading, finer bed material, higher water temperatures and shallower designs (García et al. 2004; Kadlec and Wallace 2009).

According to a review of 28 studies (Wu et al. 2016), horizontal subsurface flow CWs (HSSFCW) have in general a better capacity than free water surface flow CWs (SFCW) for the removal of *E. coli* (+1.1 log10 CFU/100 mL), FC (+0.2 log10 CFU/100 mL), fecal streptococci (FS) (+0.9 log10 CFU/100 mL), C. perfringens (+0.6 log10 CFU/100 ml) and staphylococci (+0.8 log10 CFU/100 mL), with the exception of total coliforms (TC) (−0.9 log10 CFU/100 mL). The positive influence of plants in CWs on pathogen removal has been observed, but mostly in HSSFCW (García et al.

2013; Wu et al. 2016). However, it is not certain whether plants affect system hydraulics, increase the surface area availability at plant roots or release root exudates (biocides, Figure 2-5), which might be toxic to pathogenic microorganisms (Tunçsiper et al., 2012; Avelar et al. 2014).

Wastewater composition can affect the disinfection and bacterial composition in CWs. Regrowth of bacteria can be stimulated by an increase in organic concentration and nutrients. Microbes may survive longer or replicate faster in the presence of available nitrogen, while nutrients provide resources for the metabolism of microorganisms (Díaz et al. 2010; O'Geen 2015). Also for wastewater loaded with organics, particulate matter and surfactants, a reduced removal of indicator bacteria is found. These chemicals compete for adsorption sites in the porous media and can shield the bacteria from treatment, by means of binding (adsorption) and sedimentation (Stevik et al. 2004; Boutilier et al. 2009). Besides the water composition, the hydraulic regime is also of importance especially for SSFCW systems. In free water surface flow CWs, the hydraulic conductivity of both the gravel bed and the rhizosphere zone is negligible (García et al. 2008). In SSFCW systems, however, the hydraulic conductivity of the substrate is an important design parameter. Drying and wetting cycles subject the wetland system to alternate saturated and unsaturated flow regimes. Rapid drainage might enhance atmospheric oxygenation of the treatment bed, creating a better condition for biofilm development (Tunçsiper et al. 2012). Hydraulic overloading also reduces the removal efficiency towards bacteria, potentially because of a decreased adsorption to the biofilm. Different studies (Tanner et al. 1995; Sawaittayothin and Polprasert 2007; Diaz et al. 2010; Tunçsiper et al. 2012) found a positive correlation between the hydraulic retention time (HRT) and the removal efficiency of bacteria. Tanner et al. (1995) obtained removals of 1.3, 1.3, 1.9 and 2.4 log10 CFU/100 mL of thermo tolerant coliforms in horizontal flow CWs planted with *Schoenoplectus validus.* In unplanted CWs with HRT values of 2, 3, 5.5 and 7 days, the removals were respectively 1.0, 1.2, 1.1 and 2.0 log10 CFU/100 mL. According to García et al. (2004) microbial inactivation reaches saturation values when the HRT was approximately 3 days.

Overall good removal efficiencies are presented for bacteria removal in both types of wetland systems (SFCW or SSFCW). However, single stage CWs are usually not enough to reach the standards recommended for wastewater reuse. It is therefore suggested that combining different wetland based systems may increase indicator bacteria removal rates (García et al. 2013; Liu et al. 2015). An example of such a system is presented in Figure 2-5.

2.7 Design and management

CWs are designed to take advantage of naturally-occurring processes involving wetland vegetation, soils and associated microbial assemblages for environmental cleanup (Vymazal 2007; Kadlec and Wallace 2009). Various design criteria such as retention time, organic load, substrate type and flow of CWs have been implemented to enhance the removal efficiency (Kadlec and Wallace, 2009; Wu et al. 2014: 2015). Aquatic vegetation affects the performance efficiency of CWs significantly, through adsorption, filtration, absorption, complexation, oxygen

transport. Vegetation also provides a suitable environment for microbial growth and therefore, selection of plant species is of vital concern in CWs designing (Rai et al. 2015). Other key design factors are hydraulic loading rate (HLR) and hydraulic retention time (HRT). According to Wallace and Knight (2006), one of the first steps to take into consideration is that the wetland is large enough in order to produce an effluent of acceptable quality.

2.7.1 Hydrology

It is difficult to optimize hydrological characteristics of CWs receiving agricultural runoff, because flows are not continuous and originate from many sources. Possible sources are surface runoff, stream and river runoff, tile drainage or irrigation return flows (O'Geen 2010). In general, water enters wetlands by means of different inputs which are stream flow, runoff, groundwater discharge and precipitation, while wetlands lose water by means of stream flow, groundwater recharge and evapotranspiration (Kadlec and Knight 1996). Hence, the water budget is given by means of Formula 2-1:

$$Qi - Oo + Oc - Qb - Qgw + Qsw + PA - ETAv - E\ (A - Av) = \frac{dV}{dt} \qquad \textit{Formula 2-1}$$

In which:

A: wetland surface area (m^2)
Av: wetland vegetated surface area (m^2)
ET: evapotranspiration rate (m/d)
E: evaporation rate (m/d)
P: precipitation rate (m/d)
Qb: bank loss rate (m^3/d)
Qc: catchment runoff rate (m^3/d)
Qgw: infiltration to groundwater (m^3/d)
Qi: input flow rate (m^3/d)
Qo: output flow rate (m^3/d)
Qsw: surface water inflow rate (m^3/d)
t: time (d)
V: water storage volume in the wetland (m^3)

One of the important factors which affect hydrology is the composition of plant species (Kadlec and Wallace 2009), soil characteristics and nutrient cycles (Kadlec and Knight 1996). On the other hand, vegetation in CW can alter the hydrology by roughness, drag and friction on flowing water by which mean the flow rate is reduced and water depths and hydraulic retention time are increased (Gregoire 2010). The ability to control water depths is critical for the operation of treatment wetlands. The flow and storage volume determines the time the water stays in the system as well as the degree of mixing, which influences the interactions between pollutants and wetland ecosystem (Kadlec and Wallace 2009).

2.7.2 Hydroperiod and water depth

The hydroperiod refers to the duration of flooding. This duration and water depth accounts for the water regime in CWs (Kadlec and Wallace 2009). Hydroperiod is governed by inflow, outflow, and storage capacity. It is one of the most important hydrologic design considerations, because it affects the wetland surface area, vegetation, particle settling and resuspension, biodiversity, soil redox status, soil mineralogy, and ultimately pollutant removal. CWs that receive water from irrigated agriculture often have stable hydroperiods during the growing season, but have highly variable ones in the off season due to flooding or drying down (O'Geen 2010). Fluctuating hydroperiods in CWs result in heterogeneous biotic and abiotic conditions and consequently in the establishment of a high microbial diversity able to promote both anaerobic- and aerobic-removal of a wide range of pollutants such as pesticides, heavy metals and nutrients. According to Gregoire (2010) an optimal CW size is linked to an optimal water volume to be intercepted and to an optimal water depth of around 0.5 m. According to Wallace and Knight (2006), depths greater than 0.6 m are rarely tolerated for emergent wetland vegetation and result in die-off of vegetation and subsequently promote the growth of algae.

2.7.3 Hydraulic loading rate (HLR) and hydraulic residence time (HRT)

The hydrologic loading rate is calculated by dividing the flow rate by the wetland surface area. It is a way to size a CW relative to its input water flow (Kadlec and Wallace 2009). The hydraulic loading rate equals (see Formula 2-2):

$$q = \frac{Qi}{LW} = \frac{\varepsilon h}{\mathrm{T}n} \qquad \textit{Formula 2-2}$$

In which:

q: hydraulic loading rate (m/d)
Qi: inlet flow rate (m^3/d)
L: wetland length (m)
W: wetland width (m)
ε: porosity of wetland bed media (dimensionless)
h: water depth (m)
T_n: nominal hydraulic residence time (d)

The hydraulic loading rate in agricultural settings is partly predetermined by the input flow rate. Therefore, design considerations have to modify the wetland area to optimize the hydrologic loading rate. A general rule of thumb is that the size of CWs covers 3% to 6% of its contributing watershed area. That, however, depends on the climate and the nature of runoff. In case of a wetland which is too small, excessive loading rates will limit the HRT and subsequently the

wetland removal efficiency. In case the CW is too big, expansive dry regions may occur (O'Geen 2010). According to the United States Department of Agriculture-National Resources Conservation Service (USDA-NRCS 2008), it is possible to estimate the hydraulic loading rate, if input data (rainfall, runoff curve number, type of soil) are available and in case surface runoff, is the main water supply to the CW.
Increasing hydraulic retention times in agricultural settings will mostly result in higher pollutant removal rates (Gregoire 2010). Alternatively, HRT is managed by decreasing the input or output flow, but this process can be costly or impractical to implement. Very long HRTs can have adverse effects by increasing the export of dissolved organic carbon (DOC) and associated disinfection byproducts (DBPs) or by increasing salinity via evapoconcentration effects in semi-arid regions (Diaz et al. 2008).
The porosity of a wetland (ε) is the fraction of the volume available for water to flow through. Wetland porosity has proven difficult to be accurately measured in the field. As a result, wetland porosity values reported in literature are highly variable (Bendoricchio et al. 2000). Kadlec and Knight (1996) report that average wetland porosity values are usually greater than 0.95, and ε=1.0 can be used as a good approximation.

2.7.4 Dimension and design

According to Gregoire (2010), the wetland area should be designed so that it has a very shallow sloping edge and a permanent pool. This configuration provides a variety of hydrological conditions, with some areas permanently and others temporarily flooded and favors the growth of wetland plants and microbes and pollution metabolism under both aerobic and anaerobic conditions. Wetland type, size and operation rules must be adapted to the runoff expected at the wetland site.
Efficient CWs have a variety of shapes and sizes. In general, the larger the wetland, the greater the potential for contaminant removal. To encourage parallel flow paths, with the aim to minimize stagnant zones, these large wetlands need multiple inlets and outlets. CWs designs with good hydraulic efficiencies have shapes and/or barriers to facilitate complete mixing throughout the wetland without persistence of stagnant zones and input and output locations positioned on opposite ends of the wetland (O'Geen 2015).

WRP (1994) presents a first order model approach (Figure 2-8) to estimate the wetland which is needed for efficient removal of pesticides. According to that study one of the first steps in design of a CW is to estimate the pesticide half-life time applicable to wetlands. That half-life time is chemical dependent and varies with wetlands characteristics, such as vegetative cover, vegetation type, and climatological conditions. As can be seen from Figure 2-8, the DT_{50} can be estimated by means of mesocosm studies or from the literature. However, literature values for chemical half-lives can be unreliable for CW design, because few of the available data are developed from actual wetland studies. The half-life time determines the HRT needed for efficient removal of pollutants. In this approach, the HRT is the basis for a hydraulic design and can be interpreted as the time needed for a parcel of water to pass through the wetland. Its selection is based on the objectives of the treatment (water quality or water treatment to reach

a certain target). The HRT is underestimated in the case of short circuiting (Kadlec and Wallace 2009), which results in low removal efficiencies. The actual time water spends in the wetland, can be accessed from tracer studies (Chang et al. 2011; Maillard et al. 2016), by injecting dissolved inert tracer material into the wetland inlet, and then measuring the tracer concentration as a function of time at the wetland outlet. Pollutants to be removed, should be identified and the design storm or flow must be estimated. The design flow can be selected or determined from a design storm event. A storm event is a maximum event which is used to determine the size of a wetland and possible control structures. The design will differ depending on the type of pollutants e.g. herbicides require a longer retention time for removal than suspended solids (WRP 1994).

After the HRT is determined, a wetland configuration is chosen which depends primarily on the availability of land and consists of single or multiple stages. Multiple wetland treatment units or stages have the advantages of providing greater flexibility in the design and operation. They enhance the performance of the system by decreasing the potential for short-circuiting (Bendoricchio et al. 2000).

Since wetlands are shallow (especially SFCW), the total wetland area is usually used for the purpose of design and not the volume (Kadlec and Wallace 2009). The area is calculated by means of a rearrangement of Formula 2-2, to provide the required HRT (also see chapter 6, Formula 6-1 up to 6-3). The formula used to calculate the wetland area (Formula 6-3) does not differ much from Formula 2-2 (adapted from Kadlec and Wallace 2009), if length multiplied by width in Formula 2-2, is rewritten as area and a value of 1 is used for the porosity (Kadlec and Knight 1996). For a SFCW, the nominal wetland water volume is the volume enclosed by the upper water surface and the bottom and sides of the impoundment, while for a SSFCW the porosity of the media is multiplied by the nominal water volume (Kadlec and Wallace 2009). That is clearly an indication that porosity influences SSFCW systems more than SFCW.

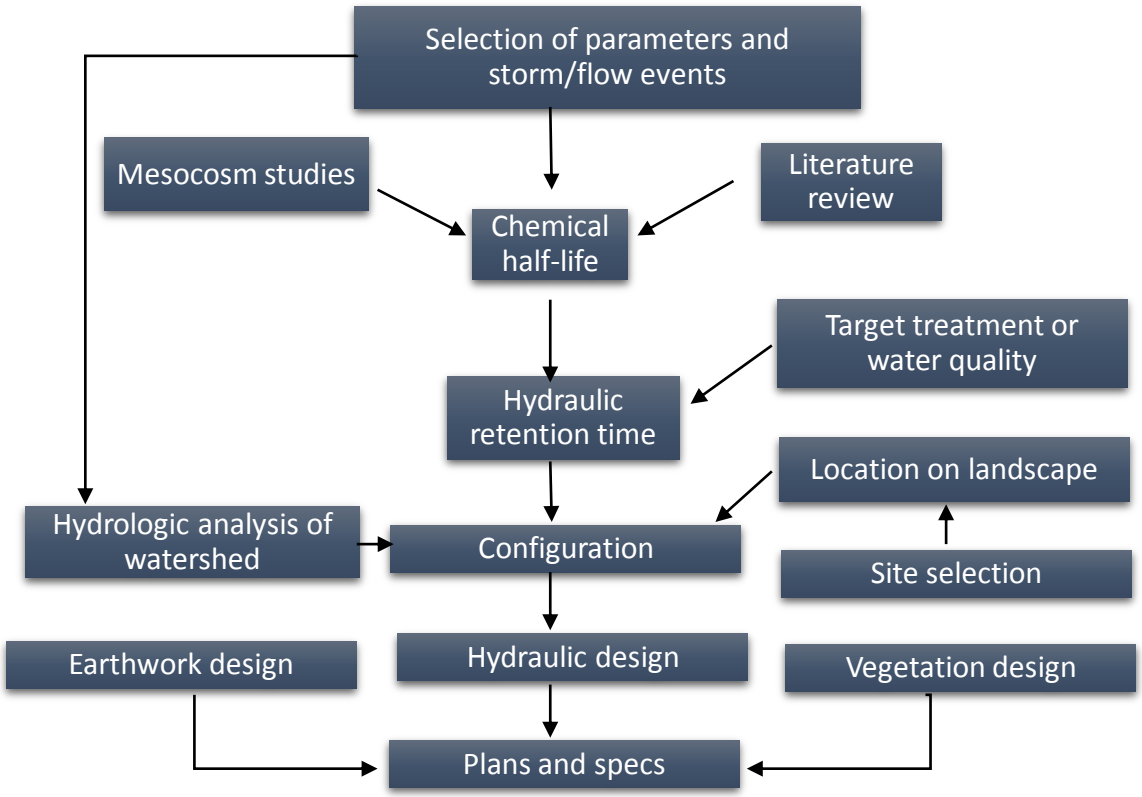

Figure 2-8: Design sequence used for constructed wetland systems (WRP 1994)

2.7.5 Selection of vegetation

Not all wetland plants are able to survive within a certain depth. It is, therefore, important to evaluate water levels at normal and peak flow rates. Not all plants grow under every climate condition and planting stocks should be available within a reasonable distance from the wetland site (Kadlec and Wallace 2009). Plants can be introduced in the wetland by natural regrowth or by planting. Planting allows control of the species mix and is less time consuming. In many countries non-native species are not allowed and native plants reasonable in quantity and costs should be considered. Furthermore larger plants which do not colonize and can resist the water quality are desired and should be outweighed by their costs and available finances (Langergraber 2004). Planting densities range from 1,000-40,000 plants per hectare. Vegetation management after planting is maintained by water depth control. The best technique for rapid plant cover is to avoid flooding. Flooding limits soil oxygen which is needed for root metabolism and effective use of nutrients (Kadlec and Wallace 2009). According to Bendoricchio et al. (2000), the water depth of reed beds should not exceed 20 cm within the first year and should be gradually increased up to 40 cm during the second year, with periods in between of drainage and shallow water depths. Harvesting of emergent vegetation is only required to maintain the hydraulic capacity, to promote active growth and to avoid mosquito outbreaks. Furthermore, having a diverse wetland vegetation community is recommended and a priority in most

mitigation studies. That is because monocultures are less tolerant to different environmental conditions and easily become stressed and might die off.

2.7.6 Management, operation, and maintenance of wetlands

Besides the considerations given for plants in the previous section, it is of importance to work with a “ramp-up” from fairly good to an impaired water quality, instead of a direct exposure of the CW to a highly polluted waste stream during the start-up of the wetland (Kadlec and Wallace 2009).

Operational and management plans of wetlands address operation and cleaning of inlet and outlet structures, cleaning of devices for primary treatment (grids, traps for sediment), biomass harvesting, berm maintenance and monitoring. Monitoring focuses on the removal effectiveness and the quality of the inflow and effluent. Treatment effectiveness should be based on pollutant mass balances and as such will require monitoring the inflow, influent pollutant concentrations, outflow, and effluent pollutant concentrations. Vegetation should also be monitored for coverage, health and diversity (WRP 1994).

2.7.7 Nuisance and nuisance control of wetlands

Several animals can bring damage to wetland structures or may interfere with the operational conditions of wetlands. Some examples are birds, fish, rodents, muskrats, beavers, insects and mosquitoes (Kadlec and Wallace 2009). A variety of mosquito abatement methods can be used including: (1) chemical treatments; (2) biological treatments, such as the use of *Bacillus thuringiensis* variety *israelensis* (Bti) and *Bacillus sphaericus* (Bs); (3) larvivorous fish such as *Gambusia affinis*; and (4) CW design features that discourage habitat and/or facilitate access by predators. Mosquitoes normally proliferate in densely vegetated wet areas, which protect mosquito larvae from predators and inhibit biological control efforts. CW design features to control mosquito larvae attempts to discourage these densely vegetated areas, by means of preventing stagnant areas, and encourage mosquito fish habitats. Those habitats are created by (1) including the creation of steep walled basin margins, (2) maintaining episodes of water depths greater than 80-150 cm to discourage establishment of dense emergent macrophytes and (3) creation of deeper areas for fish with access to shallow areas where larvae prevail (O’Geen et al. 2010). Other possibilities are draining and flooding of the wetland. Draining of the wetland can be important for many reasons: it aids the establishment of vegetation after planting, it allows supplementary planting if initial planting results in poor survival rates, it can be used to control weeds, particularly floating species, it can help in mosquito and fish management and it facilitates the reduction of erosion (Bendoricchio et al. 2000).

Chapter 3 Risk assessment of pesticide usage by farmers in Suriname: a pilot study for the Alkmaar and Tamanredjo regions

This chapter has been partially compiled from:
Mahabali, S.S. and Spanoghe, P., 2015. Risk assessment of pesticide usage by farmers in Commewijne, Suriname, South America: a pilot study for the Alkmaar and Tamanredjo regions. Environmental monitoring and assessment, 187 (3): 153, pp. 12.

Abstract
Limited scientific work is available on pesticide use and exposure in Suriname. Therefore, research was conducted to assess the application, safety practices (use of personal protective equipment's (PPEs), and potential risks of pesticide use. A face-to-face questionnaire was worked out and the consumer exposure to pesticides was calculated by means of the International Estimated Short-term Intake (IESTI). The amount of pesticides used by farmers was compared with the guidelines on the label and the authorized dosage in the EU. The majority of the farmers was male and between 41 to 60 years old. Most of them had only a primary school education. Less than 5% of the farmers used non-authorized pesticides. Results show that most farmers (58-100%) apply an amount of pesticide for spraying which is within 0-100% of the regulated dose. Fairly good results were obtained for the use of personal protective equipment's (PPEs) during spray applications. Statistical analysis did not reveal a significant difference between the different age classes, the different levels of education, and the use of PPEs. Pesticide residue analysis of four major crops measured during two separate assessments showed that between 40% (first assessment) and 65% (second assessment) of samples contained pesticides. The results for imidacloprid exceeded the maximum residue level (MRL). Assessing consumers exposure showed that the highest observed exposure value (IESTI) was not higher than 74.5% of the EU acute reference toxic dose (ARfD). In Suriname, educational programs, G.A.P. training, sustainable agricultural practices, food monitoring studies, and legislative control mechanisms are needed to protect farmers' health and the environment and to ensure food safety.

3.1 Introduction

In Suriname, an increasing trend is observed for the import of pesticides (insecticides, herbicides, and fungicides). Because of repeated detections of high pesticide residue concentrations and sometimes non-authorized pesticides, the Netherlands wants to stop the import of fresh produce from Suriname (Mijland 2012). De Putter and Van Sauers-Muller (2008) presented monitoring results from the Netherlands in the period 2004-2006. In this report, a value as high as 1800 times the MRL was found for the organophosphate methamidophos in

spinach. The Surinamese government enforced several measures to comply with new trade agreements arising from globalization and to ensure food quality and safety (see chapter 1). Several laws and resolutions exist on the use and storage of pesticides. For persistent pollutants (e.g., organochlorine pesticides), the Stockholm Convention has been ratified in Suriname in September 2011. As from April 16, 2012, the import, sale, storage, and use of harmful pesticides such as endosulfan, dimethoate, and carbofuran were banned. Despite these efforts, several notifications from the Netherlands, which is one of the biggest export markets, show pesticide residue levels higher than the MRL values (NVWA 2013). This chapter provides data on pesticide application, safety practices (use of personal protective equipment's (PPEs)), and potential risk of human exposure. That, by means of residue analysis and calculation of the International Estimated Short-term Intake (IESTI) in the resorts Alkmaar and Tamanredjo of the Commewijne district. A dietary exposure assessment combines food consumption data with residue data in food. It results in the International Estimated Short-term Intake (IESTI), which is compared with the toxicity value (the Acute Reference Dose or ARfD) to predict the human risk for acute exposure which commonly covers a period of 24 h (FAO/WHO 2009).

3.2 Material and methods

3.2.1 Study area

This study took place in district Commewijne, Suriname. Suriname is situated along the north coast of South America, the area is 163,820 km^2 and the population is 541,638 (Census statistieken 2012). The country has a typical tropical climate with 2 rainy and 2 dry seasons, a mean daily temperature of about 27°C and an annual average rainfall of 1500-3000 mm (National Profile Suriname 2006). The study was carried out in 2 regions of district Commewijne namely Alkmaar with an area of 81 km^2 and a population of 4,213 and Tamanredjo with an area of 512 km^2 and a population of 5,510 people. Agriculture is the main source of income in these areas. The majority of farmers (46) were from the Alkmaar region, while 32 farmers were from Tamanredjo.

3.2.2 Questionnaire

For this cross-sectional study, a total population of 78 registered farmers was taken into consideration to assess pesticide application and safety practices (use of PPEs). The formula used (Israel 1992) for determination of the sample size was:

$$N_0 = \frac{Z^2 pq}{e^2}$$ *Formula 3-1*

In which:

N_0: *sample size (N_0 = 73)*
Z^2: *abscissa of the normal curve that cuts off an area α at the tails. Note: 1-α*

equals the desired confidence level (95%) ($Z^2 = [1.96]^2 = 3.84$)

e: the desired level of precision (e = 0.05)

p: the estimated proportion of an attribute present in the population (p = 0.95; 95% of G.A.P. registered farmers equals 78 farmers)

q: equals 1-p (q = 0.05)

According to Israel (1992), the sample size is increased by 10% to compensate for persons the researcher cannot contact. With this 10% increase, the sample size becomes 80. However, only 78 farmers were G.A.P. registered farmers and therefore this number was chosen to execute the survey.

The questionnaire on G.A.P. and pesticide use (see Appendix 5) was developed, based on information provided in the local G.A.P. Control Points and Compliance Criteria, All Farm Base (2012). The questionnaire consisted of 46 questions (some divided into sub questions). The survey was carried out from March up to June 2010 (4 months) by making use of multiple observers (students and agricultural extension officers). These officers were familiar to the farmers and helped well in receiving a good response to the questions, especially when farmers could not express themselves using the official language of Suriname (Dutch). Legislation concerning non-authorized pesticides, was used to validate answers given by farmers on the types of pesticides used. Farmers responses related to the pesticide dose applied in kg per hectare, had to be transformed into SI units. The application rate of frequently used pesticides was compared with the label information and with the information from the authorization in the EU. The governmental website of Belgium (www.Fytoweb.be) and the website of the European Commission (http://ec.europa.eu/food/plant/pesticides) were consulted on May 12, 2013.

For determination of the potential risk of pesticide use, 30 consumers were questioned about their age, daily diet, and their body weight. The vegetables mostly grown in the area are aubergine, yard-long bean, African eggplant, and pepper. Because no fixed MRL values are available for Suriname, the pesticide residues were compared with the EU MRL's. These are the same values used by the Dutch authority to evaluate pesticide residues in crops originating from Suriname.

3.2.3 Statistical analysis

Received answers on the use of PPEs were coded, and a summary statistics was computed. A code system from 1 up to 4 was used (1, equals unknown; 2, no use; 3, sometimes; and 4, always). Because data did not fulfil the requirements of a two-way analysis of variance between the different levels of age class and educational level, a Kruskal-Wallis nonparametric test was executed to determine significant relationships among farmers' age classes and educational levels. Detailed results on the statistical analysis are provided in Appendix 4. In the case wherein data were statistically different, a multiple comparison procedure (Dunn's method) was used to isolate the group, which differs from the others.

3.2.4 Pesticide residue analysis of crops

A preliminary assessment was done on 20 crop samples purchased from 20 different farmers in the research area during September to December 2009. Crop samples (aubergine, yard-long bean, African eggplant, and pepper) were purchased. For each of the 4 types of crops, 5 different farmers were approached to sell around 1 kg of each crop sample, which resulted in a total of 20 crop samples. Samples were placed in an icebox with cooling agents and transported to Belgium. After arrival, samples were stored in the freezer (-20°C) for further analysis and subsequently analyzed on the presence of 6 different pesticides: imidacloprid, chlorothalonil, endosulfan, cypermethrin, lambda-cyhalothrin, and chlorpyrifos. These pesticides belong to different chemical classes, i.e., the neonicotins, organochlorines, pyrethroids, and organophosphates.

The second assessment to determine pesticide residue concentrations in crops was executed in the period February-March 2013. The same types of crops were purchased as from the previous of pesticides were quantified. Contrary to the first assessment, samples were extracted at the Chemistry laboratory of the Anton de Kom University of Suriname. The 20 crop samples of around 1 kg wet weight, were all chopped separately with a blender (Molinex) and for each crop sample, two subsamples (sample and parallel replicate) of 50 g each were weighed for further analysis.

The analytical procedure involved the extraction of the pesticides with a suitable solvent. Imidacloprid was extracted with an acetonitrile/water mixture (1:4), the extract was centrifugated, and subsequently filtrated through a polyvinylidene difluoride (PVDF) membrane with a pore size of 0.22 µm (Carl Roth, Karlsruke-Rheinhafen, Germany) and stored (-20°C) in vials. These vials were subsequently transported (DHL express/air cargo) to Ghent University, Belgium for quantification. Imidacloprid was separated on a platinum C18-ESP (electrospray), 3 µm column and quantified by means of high performance liquid chromatography (HPLC) (Thermo Surveyor equipped with a Surveyor LC pump plus and a Surveyor Photodiode Array (PDA) plus 5 detector from Thermo Fisher). The LOD for imidacloprid in plants was 0.5 mg/kg.

The organochlorines/pyrethroids chlorothalonil, lambda-cyhalothrin, and cypermethrin were extracted with a mixture of hexane/acetone (1:1) and dried over anhydrous sodium sulfate. The extracts were evaporated until near dryness, in a Heidolph Laborota 4000 Rotary Evaporator at 40°C, re-dissolved in solvent and transferred into vials and transported (DHL express/air cargo) to Ghent University, Belgium for Gas Chromatography Mass Spectrometry (GCMS) analysis. The limit of detection (LOD) for these compounds in plants was 0.1 mg/kg.

For organophosphate detections, ethyl acetate was used as a solvent, and an analytical method similar to the organochlorines/pyrethroids was followed. The LOD for chlorpyrifos was 0.05 mg/kg (see Table 3-5). Quantification for both organochlorine/pyrethroids, and organophosphates was done by means a 6890N gas chromatograph (Agilent Technologies) equipped with a HP-5ms column.

3.2.5 Determination of the IESTI

The international estimated intake (IESTI) is used to estimate consumer exposure based on the highest reported 97.5th percentile intake during a single day. For the calculation of the IESTI, Formula 3-2 (WHO 2013) was used. In this formula the unit weights of the edible portion were lower than that of the Large Portion (LP). This was the case for all the commodities investigated. For a pesticide residue with an established Acute Reference Dose (ARfD), the IESTI is calculated by means of:

$$IESTI = \frac{Ue \times HR \times Vvar + (LP - Ue) \times HR}{bw} \quad \textit{(Formula 3-2)}$$

In which:

LP: highest large portion provided (kg food/day) (97.5th percentile of eaters)
HR: highest residue in a composite sample of edible portion found in data from supervised trials from which the MRL was derived (mg/kg)
bw : average body weight for a population age group (kg) (default = 65 kg)
Ue : edible portion of the unit weight (kg)
Vvar: variability factor representing the ratio of the 97.5th percentile residue to the mean residue. A standard variability factor of 7 is used within the EU, when the URAC (unit weight of the raw agricultural vegetable, in kg) is between 25 g up to 250 g (Kneževič et al. 2012).

3.3 Results

3.3.1 Demographic and general information

A good response was given by all 78 G.A.P. farmers to the questions (response rate of 100%). The majority (91%) of these farmers were male. Only two farmers (2.6%) belonged to the youngest age class (below 25 years), while the biggest group (64.1%) was between 41 and 60 years old. Within that age class, the highest percentage (ca. 25%) of interviewees had followed a primary and secondary education (see Table 3-1). Only one farmer had a college degree, while 6.4% of the farmers had no education at all.

3.3.2 Field area and soil type

Most farmers (68%) grow their crops on a field with an area in the range of 0.05-1 ha with mostly clay soils or a mixture of clay with sand or shells. Only a small fraction of farmers (13%) grows their crops on sandy soils.

3.3.3 Pesticide knowledge, application and safety practices

From the survey, it was concluded that most farmers knew the names of the pesticide products they used.
For the non-authorized pesticides, however, only 23% of farmers knew the names of these products. The pesticide storage facility was visited to make an inventory of products actually used and in stock, and to observe the way pesticides were stored. From these field observations it was concluded that 58% of farmers stored their pesticides locked away from children and other unauthorized individuals.

Table 3-1: Education of farmers

Age class	No. of farmers		% of farmers		Prim[a] (%)		Sec[b] (%)		College[c] (%)		No. education (%)	
	M	F[d]	% M	% F	% M	% F	% M	% F	% M	% F	% M	% F
<25	1	1	1.3	1.3	0	0	1.3	1.3	0	0	0	0
25-40	14	1	17.9	1.3	6.4	1.3	9	0	1.3	0	1.3	0
41-60	45	5	57.7	6.4	28.2	3.8	25.6	2.6	0	0	3.8	0
>60	11	0	14.1	0	11.5	0	1.3	0	0	0	1.3	0
Total	71	7	91	9	46.1	5.1	37.2	3.9	1.3	0	6.4	0

[a] *Primary level or grades 5-12 years*
[b] *Secondary level or grades 12-19/20 years*
[c] *College 20+ years or tertiary level of education*
[d] *Male (M) and female (F)*

This survey also revealed that two farmers gave incorrect answers for the proper use of pesticides. One farmer used malathion (insecticide) as an herbicide, while another used paraquat (herbicide) as a ground disinfectant. From the survey, it is further concluded that only 51% of the farmers know that they risk health problems when they are exposed to pesticides.
The main pesticides used for ground disinfection are λ-cyhalothrin (37%), malathion (11.6%), chlorothalonil (6.4%), and captan (6.4%). For treatment of pests and diseases, chlorothalonil (42.3%), λ-cyhalothrin (30.8%), diafenthiuron (26.9%), mancozeb (21.8%), malathion (19.2%), and imidacloprid (18%) were used. For weed control, most farmers (74%) used paraquat (see Table 3-2). Less than 5% of the farmers used non-authorized pesticides. For ground disinfection, 12% of the farmers applied forbidden cocktails (mixtures of two or more pesticides/compounds), while 42.3% of farmers did not use any pesticide at all. For crop protection, 7.7% of farmers did not use any pesticides. The average working hours a day were 5.1 ± 2.2 h.

Table 3-2: Pesticides used by farmers (%) for ground disinfection and crop protection and the WHO toxicity class to which they belong (WHO 2010)

Active ingredient	Trade name	Use a.s.[a]	WHO Toxicity Class[b]	Ground Disinfection (%)	Crop Protection (%)
imidacloprid	Admayor 42.8%	I	II	1.3	1.3
pyriproxifan	Admiral 10 EC[f]	IGR	III	0	2.6
carbendazim	Backtral 500 g/l	F	U	1.3	9.1
5-o'desmethyl avermectin	Bio-emax	Bio/I		1.3	3.9
chlorothalonil	*Bravo 50%*	*F*	*U*	*6.5*	*42.9*
captan	*Captan*	*F*	*U*	*6.5*	*9.1*
cypermethrin	Cyperkill 25 EC	I	II	1.3	2.6
diazinon	Basudine 60%	I	II	5.2	1.3
chlorpyrifos	Dursban Mole Cricket Bait 0.5%	I	II	2.6	1.3
carbendazim	Entral	F	U	1.3	1.3
Carbofuran[c]	Furodan 5%	I	Ib	1.3	0
ethyl 2-(4-(6-chloro-2-benzoxazolyl)oxy) phenoxy)propanoate	Furore	H		2.6	
glyphosate	*Round-up*	*H*	*III*	*16.9*	
paraquat	*Gramoxone*	*H*	*II*	*74.4*	
γ-HCH[c]	Lindane	I	II	1.3	0
imidacloprid	*Imidox 20%*	*I*	*II*	*5.2*	*18.2*
λ-cyhalothrin	*Karate 2.5%*	*I*	*II*	*37.7*	*31.2*
malathion	*Malathion*	*I*	*III*	*11.7*	*19.5*
mancozeb	*Manzeb 50 WP[d]*	*F*	*U*	*2.6*	*22.1*
azadirachtin	Neemazal 0.3%	Bio/I	U	0	2.6
diafenthiuron	*Pegasus 500 SC[e]*	*AC*	*II*	*2.6*	*27.3*
chlorfenapyr	Pirate 24%	I, MT	II	0	3.9
fenbutatin-oxide	Torque 55%	MT	II	0	1.3
dimethoate/endosulfan	Twinox 300 g/l[c]	I/I	III	1.3	0

The pesticides used the most are presented in italics and in red

[a]H: herbicide, I: insecticide, IGR: insect growth regulator, F: fungicide, Bio: biopesticide (Insecticide), AC: acaracide, MT: miticide

[b]WHO toxicity class: highly hazardous (1b), moderately hazardous (II), slightly hazardous (III), and unlikely to present acute hazard in normal use (U)

[c]Non-authorized pesticides (Staatsbesluit Suriname no. 18, February 2005)

[d]Wettable Powder

[e]Suspension Concentrate (SC)

[f]Emulsifiable Concentrate(EC)

3.3.4 Comparison of applied dose (kg/ha) with recommended dose on the label

The dose (kg/ha) of frequently used pesticides (Table 3-3) was compared with the recommended dose on the label. The results for the pesticides λ-cyhalothrin, diafenthiuron, mancozeb, chlorothalonil, imidacloprid, and malathion show that respectively 70.6, 57.9, 93.3, 72.7, 91.7, and 72.7% of the farmers apply a dose which is equal to or lower than the recommended dose (0-100% interval) (Figure 3-1). However, it was found that between 10.5 and 18.2% of farmers apply a dose which is higher than 1000% (>10 times) the recommended dose. In two cases, a dose was found which was 1041 and 1500 times (for λ-cyhalothrin) and 690 and 1036 times (malathion) the recommended dose of the label.

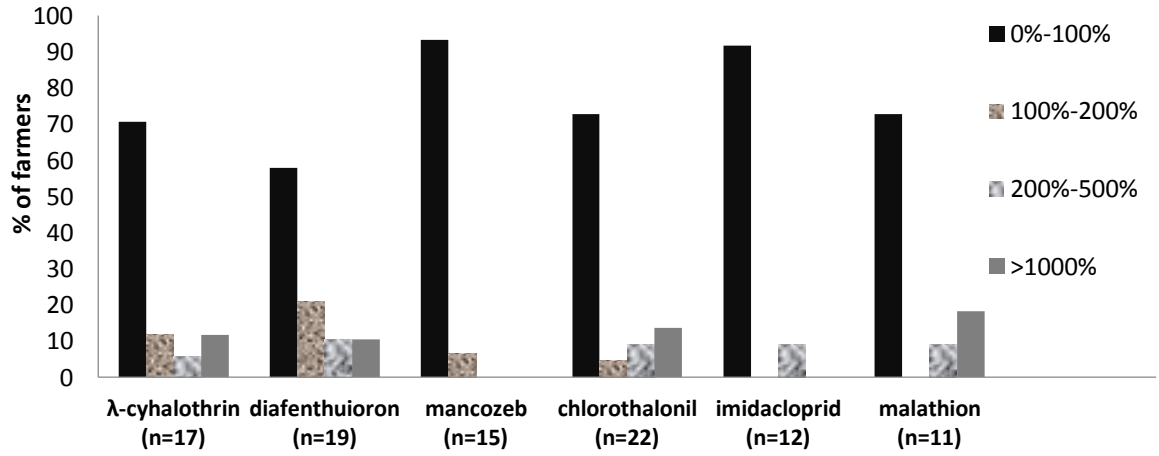

Figure 3-1: Percentage of farmers who apply a dose which is within the recommended label dose (0%-100%), between 1-2 times higer (100%-200%), 2-5 times higher (200%-500%), and higher than 10 times (>1000%) the recommended dose, with n representing the number of farmers who use a certain type of pesticide (e.g. n = 19 means that 19 farmers use diafenthiuron)

Comparison of the average applied doses with those applicable in Europe, showed that for the pesticides λ-cyhalothrin, mancozeb, chlorothalonil, imidacloprid, and malathion, respectively, 70.6, 100, 59.1, 83.3, and 72.7% of the farmers applied a dose, which was equal to, or lower than the recommended dose (0-100% interval). In addition, some pesticides were applied in too high amounts. For the pesticides chlorothalonil, malathion, and λ-cyhalothrin, respectively, 13.6, 18.2, and 5.8% of the farmers applied a dose, which was more than ten times higher (>1000%) than the recommended dose.

The average dose applied per crop treatment in the Commewijne district was calculated to be 4.15 kg active ingredient per hectare (SD=16.31, n=72 farmers).

Table 3-3: Overview of the percentage of farmers using a certain pesticide, the range of the applied pesticide dose, the average dose with standard deviation (SD), and the recommended label dose for the main pesticides used in the Commewijne area

Pesticide	% of farmers	Range of applied dose (kg/ha)	Average dose (kg/ha)	SD	Recommended label dose (kg/ha)
chlorothalonil	28	0.014-80	8.4	22	0.58-1.4
diafenthiuron	24	0.010-32	2.2	7.3	0.096-0.13
λ-cyhalothrin	22	0.00050-30	3.0	8.6	0.01-0.02
mancozeb	19	0.0096-1.4	0.24	0.36	1.7-2.2
malathion	13	0.018-114	18	39	1-2
imidacloprid	12	0.0053-2.4	0.35	0.68	0.24-0.96
captan	6	0.030-3.0	0.82	1.14	0.75

3.3.5 Safety practices

Approximately 85% of the farmers used a mouth cap, 90% wear protective clothes, 92% wear boots and only 44% uses gloves (Figure 3-2). Also 41% of the farmers make use of safety glasses during pesticide handling and application.

The total number of farmers using PPEs was the highest for farmers belonging to the age class 41-60 years. However, breakdown of this use in relative percentages of farmers per total number of farmers within each age class (Figure 3-2, picture above, right) shows comparable results for the use of boots, protective clothes and a mouth cap, while lower relative percentages were found for the use of gloves and a mouth cap (age class 41-60 years) and protective clothes (age class 25-40 years). According to Table 3-1, the highest level of education was within the age class 41-60 years. Statistical analysis, making use of a Kruskal-Wallis one-way ANOVA on ranks test, did not reveal a significant difference between the different age classes, educational levels and the use of PPEs.

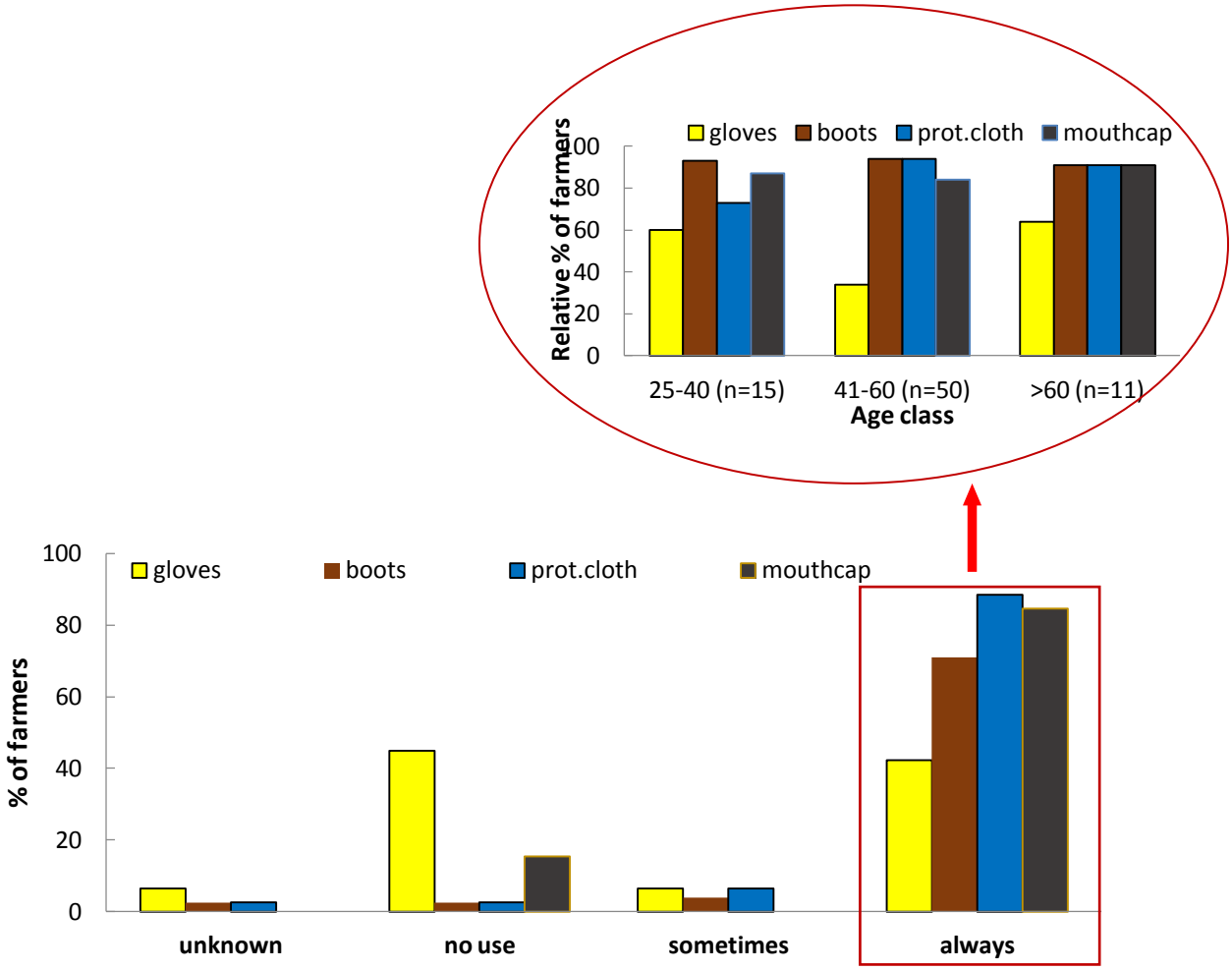

Figure 3-2: Use of PPEs (gloves, boots, protective clothes and mouth cap) by farmers in the Commewijne district in Suriname, with breakdown of the numbers found for the category "always" (Figure above, right) in relative percentages of farmers within the three main age classes (n=15 for age class 25-40 years; n=50 for age class 41-60 years and n=11 for the age class >60 years) with n, representing the total number of farmers within each age class

3.3.6 Results for residue analysis

In Table 3-4, the results of the first pesticide residue assessment (year 2009) are presented. In 40% of the 20 samples analyzed, pesticides residues were detected, but concentrations were below the EU MRL values.

Table 3-4 Average pesticide concentrations (n=2)[a] with EU-MRL values. Results are obtained from the first sampling comparison of crops in Commewijne, Suriname. (period September-December 2009)

Sample nr and crop	Pesticide	Concentration (mg/kg)	EU MRL[c] (mg/kg)
13 Aubergine[b]	λ-cyhalothrin	0.03	0.5
2 Yard Long Bean	λ-cyhalothrin	0.02	0.2
19 Yard Long Bean	λ-cyhalothrin	0.06	0.2
8...Pepper	Chlorothalonil	0.2	2
16 Pepper	Chlorothalonil	0.5	2
10 African Egg Plant	Chlorothalonil	0.01	2
15 African Egg Plant	Imidachloprid	0.02	0.5
17 African Egg Plant	Imidachloprid	0.01	0.5

[a] *n=2 means that each sample has a parallel duplicate*
[b] *Sample number 13 (aubergine), out of a total of 20 crop samples, purchased from 20 different farmers. The 20 crop samples consisted of 5 samples of Yard Long Bean, 5 samples of African Egg Plant, 5 samples of Aubergine and 5 samples of Pepper*
[c] *EU-database: http://ec.europa.eu*

In Table 3-5, the number of times is given that a pesticide was detected in a certain crop, for the second residue analysis, which was performed in 2013.

Table 3-5: Number of detections of pesticides for the second sampling comparison of crops. For each crop, 5 samples were analyzed retrieved from 5 farmers. Analysis were performed during February-March 2013 and presented with the limit of detection (LOD)

Pesticide	Imida-cloprid	Chloro-thalonil	λ-cyhalo-thrin	Endosul-fan	Cyperme-thrin	Chlorpy-rifos
LOD (mg/kg)[a]	0.5	0.1	0.1	0.1	0.1	0.05
Yard Long Bean	4	ND[b]	ND	ND	ND	ND
African Egg Plant	1	2	ND	ND	ND	ND
Aubergine	1	1	ND	ND	ND	ND
Pepper	2	2	ND	ND	ND	ND
Total	**8**	**5**	**0**	**0**	**0**	**0**

[a] *Limit of detection (LOD) expressed in mg pesticide/kg vegetable*
[b] *Not Detected*

From the 20 samples analyzed, 65% showed pesticides residues. Imidacloprid was found in 8 samples and chlorothalonil in 5 samples. The other pesticides were either not present or below the limit of detection (LOD).
Imidacloprid was found in all 4 types of crops, while chlorothalonil was not detected in yard-long beans. All residues of chlorothalonil were below the EU MRL of 2 mg/kg, while 80% of the imidacloprid residues did not comply to EU MRL values (see Table 3-6).

Table 3-6: Comparison of average pesticide concentrations obtained (n=2)[a] with EU-MRL values, in Commewijne, Suriname. Results obtained from the second sampling comparison (period February-March 2013). Values presented in red are above the EU-MRL

Sample nr and crop	Pesticide	Concentration (mg/kg)	EU MRL[c] (mg/kg)
3 Yard Long Bean[b]	Imidachloprid	1.268	0.05
1 Yard Long Bean	Imidachloprid	0.062	0.05
5 Yard Long Bean	Imidachloprid	0.838	0.05
2 Yard Long Bean	Imidachloprid	3.691	0.05
9 African Egg Plant[b]	Imidachloprid	1.908	0.5
16 Aubergine	Imidachloprid	1.779	0.5
15 Pepper	Imidachloprid	0.094	1
14 Pepper	Imidachloprid	0.115	1
9 African Egg Plant	Chlorothalonil	0.001	2
8 African Egg Plant	Chlorothalonil	0.246	2
13 Pepper	Chlorothalonil	0.0534	2
19 Aubergine	Chlorothalonil	0.00532	2
14 Pepper	Chlorothalonil	0.697	2

[a] *n=2 means that each sample has a parallel duplicate and that the pesticide concentration is based on the average of the concentrations found for each sample and its duplicate*

[b] *3Yard Long Bean means sample 3 out of a total of 20 crop samples, which were purchased from 20 different farmers. These 20 samples consisted of 5 samples of Yard Long Bean, 5 samples of African Egg Plant, 5 samples of Aubergine and 5 samples of Pepper*

[c] *EU-database: http://ec.europa.eu*

Calculation of the IESTI value is based on a worst-case scenario and therefore the highest residue value obtained for imidacloprid was used. The large consumption portion (Formula 3-2) or the 97.5th percentile of portion sizes taken by people consuming a commodity (in kg of food per day) was calculated to be 0.150, 0.455, and 0.214 for aubergine, African eggplant, and yard-long bean, respectively. The comparison of the IESTI with the EU ARfD (0.08 mg/kg/BW/day), showed that in all types of vegetables, this value is below the toxicity level and ranks between 38.1 and 74.5% of the EU ARfD (see Table 3-7).

Table 3-7: Comparison of the IESTI with the EU ARfD and the newly proposed EU ARfD (results in green) for the different samples/crops

Sample nr crop	HR[a] imida-cloprid mg/kg	IESTI	EU ARfD in mg/kg/ BW/day	IESTI% of ARfD	IESTI[b]% of new ARfD
16 Aubergine	1.779	0.0345	0.08	43.2	50.8
2 Yard Long Bean	3.691	0.0596	0.08	74.5	99.3
9 African eggplant	1.908	0.0308	0.08	38.5	54.1

[a] *Highest residue of imidacloprid in a specific crop*

[b] *Newly proposed IESTI of 0.06 mg/kg/BW/day (EFSA 2013)*

3.4 Discussion

The main objectives of the study in this chapter, were to assess data on pesticide usage, safety practices (use of PPEs), and to estimate the potential risk of pesticide usage (calculation of the IESTI). The strength of this study was the relative short time frame for the assessment of valuable and multiple outcomes such as the type and amount of pesticides used and dietary intake and the high response rate of farmers. The high response rate and the reduction of recall bias of this study have minimized the bias to which cross sectional studies are normally susceptible to. The low educational level of farmers and their inability to express themselves using the official language was a limitation in answering questions.

This survey received a response rate of 100%. In other studies (Issa et al. 2010; Lopus et al. 2010; Zyoud et al. 2010), lower response rates were found due to mailing of the survey instead of a face-to-face interview and the lack of a good registration system of the active farmers. In the present study, 64% of the farmers were in the age class of 41-60 years and results are comparable to those in Issa et al. (2010), where 59.1% of the farmers were within the age class 41-60 years. In other studies conducted in the West Bank, Palestina (Zyoud et al. 2010) and in Zimbabwe (Magauzi et al. 2011), farmers were much younger and the mean age was 38.8 and 28 years, respectively. The majority of farmers in Commewijne, Suriname had only a primary educational level (53.8%). Some pesticide labels are written in English and are difficult to read or to understand by these farmers. Similar remarks were also reported in Hurtig et al. (2003); Recena et al. (2006) and Polidoro et al. (2008).

In the research area, insecticides were used the most, followed by fungicides and herbicides. This order in usage is also observed in other countries, namely Chili (Burleigh et al. 1998), Tanzania (Ngowi et al. 2007) and the West Bank, Palestina (Zyoud et al. 2010). The applied dose was not consistent with the pesticide label information and may be linked to the low educational level of farmers, the lack of a registration system for pesticide use, and insufficient training programs on the use of pesticides (Salameh et al. 2004; Ibitayo 2006 and Abhilash and Singh 2009). A wide range of values was found for the applied dose by farmers. The average pesticide dose applied in the Commewijne district was 4.15 kg active ingredient per hectare of crops (SD=16.31, 72 farmers), which is somewhat higher than the global average dose (3.2 kg active ingredient per ha of crops) found in Schreinemachers and Tipraqsa (2012). In that study, lower amounts of pesticides applied per crop treatment were found in poor countries compared to rich countries. In addition, great differences were observed in pesticide usage per country whether or not a rich or poor country.

For the use of PPEs (mouth cap, boots, and protective clothes), good results were obtained. However, approximately 10% of the farmers do not put on protective clothes. A possible reason for this behaviour is according to Cropper (1994) that such clothes are inconvenient, and reduces productivity in the field, especially in hot tropical countries. Farmers also do not understand the health consequences related to misuse or no use of PPEs. In the present study,

51% of the farmers knew that they risk health problems when exposed to chemicals; however, in practice, their behaviour towards some types of PPEs (gloves and safety glasses) was not according to G.A.P. The same view was given in Norkaew et al. (2010) and Panuwet et al. (2012), who noted that 75% of farmers from the north-eastern region of Thailand do not make use of PPEs and are ignorant of the consequences for personal health.
In the study conducted by Matthews (2008), the attitude and behaviour related to the use of crop protection products of more than 8500 smallholders in 26 countries was evaluated for the period 2004-2006. For the use of protective clothes and boots by Surinamese farmers, comparable results were obtained for the countries Colombia (Latin America), Morocco and Cameroon (African countries). Very poor results (less than 15% of the farmers were protected by PPEs) were obtained for the Asian countries namely Bangladesh, Philippines, and Sri lanka. Several studies (Van de Lande 2001; Cataňo et al. 2008; Matthews 2008) also describe that instead of a mouth cap, a dust mask, a piece of wet cloth, or a handkerchief is mostly used, which does not give sufficient protection against pesticide vapors. A good practice is according to Fishel (2012), the use of a special type of respirator, namely for liquid pesticides belonging to WHO toxicity class I. An example of that is the National Institute for Occupational Safety and Health (NIOSH) approved respirator equipped with an organic vapor (OV) cartridge. This type of PPE was however not observed during the survey, possibly because these types of PPEs are not readily available in Suriname and are too expensive for the average farmer.

Several farmers export fresh products to e.g. the Netherlands. Those who apply a non-authorized pesticide and a dose, which is higher than the recommended one, are at a higher risk that their vegetables are rejected at the border, or pose a threat to consumer's health. A small number of farmers (less than 5%) applied non-authorized pesticides, possibly because these pesticides were old stocks and very effective. This value was expected to be much higher, because only 23% of farmers knew the names of these illegal pesticides. According to some extension officers, some non-authorized pesticides are smuggled into Suriname from neighbouring countries such as Guyana. According to Van Sauers-Muller and Ester (2006), endosulfan, an organochlorine pesticide, was still imported in Suriname in 2004, while it is not authorized in the Netherlands for more than 15 years. Other studies (Van de Lande 2001; Snelder et al. 2008) also mention the use of highly hazardous and moderately hazardous pesticides, such as carbofuran (WHO class Ib) and λ-cyhalothrin (WHO class II). In the study of Ryan Galt (2010), an assessment was made of non-authorized pesticides on fresh vegetables imported from 21 countries into the USA during 1996-2006. Results for Guatemala show the highest rates of adverse residues compared to countries like Spain, Jamaica, and China. In these countries, the organophosphate methamidophos is frequently used while it is within the list of adverse residues. Methamidophos detections are also mentioned in a review (Quintero et al. 2008) on the occurrence of organophosphate pesticides in 6 different vegetables from Venezuela.

Although, not in the list of pesticides used by farmers in the present study, the organophosphate methamidophos was also detected in a concentration higher than the MRL in celery originated from Suriname. Besides the detection of methamidophos, also 52 detections

were done for imidacloprid and 18 for chloorthalonil by the NWVA (Netherlands Food and Consumer Product Safety Authority) of which 15% were above the MRL (NVWA 2013). Chlorothalonil and imidacloprid were detected in the present study (second assessment conducted in 2013), but only the results for imidacloprid were above the EU MRL for 30% of the samples. Imidacloprid is usually not included in food monitoring programs of most countries; therefore, little information is available on residue data (Cox 2001). Similar results (0.01-0.3 mg/kg imidacloprid) were found in the study of Fernandez-Alba et al. (2000) in all 45 samples of peppers, tomatoes, and cucumbers, while in the study of Daraghmeh et al. (2007), imidacloprid was detected in all 32 samples of apples, eggplants, and potatoes, with concentrations ranging from 0.24 up to 0.41 mg/kg.

Pesticide residues obtained during the first sampling comparison of crops in 2009, were all below the EU-MRL values. The second pesticide residue assessment was done in 2013 and was greatly improved compared to the first, by means of the adequate storage of samples and reducing the time between sampling and analysis. The main reason for that was that in 2009, no facilities and possibilities were present in Suriname, to prepare and extract the samples. Another aspect of importance is, that the farmers were informed about the visit and its purpose, namely to determine the amount of pesticide(s) present in crops. This could have resulted in 2009 in a bias, because vegetables intended for personal use by farmers, could have been given to the person involved in the sampling. The reason for that is because farmers usually have two kind of fields, one for personal use (less or no pesticides applied) and one for the market or export (pesticides are frequently applied).

The ARfD of 0.08 mg/kg/BW/day was used for comparison with the IESTI to assess the food safety risk of consumer exposure. Experts propose to lower this ARfD value to 0.06 mg/kg/BW/day, awaiting the results of ongoing research to provide more data on the effects of imidacloprid on the developing human nervous system, in particular the brain (EFSA 2013). If lowered, the IESTI for imidacloprid in combination with Yard Long beans becomes almost 100% indicating a possible risk.

3.5 Conclusion

It can be concluded from this study that farmers in the Commewijne area apply a dose, which is mostly equal or lower than the recommended label/EU dose. A small percentage (less than 5%) of the farmers uses non-authorized pesticides and their knowledge about these pesticides is poor. Good results were obtained for the use of PPEs, namely the wearing of protective clothes and shoes but not for the use of gloves or safety glasses. No statistical significant differences were obtained between different age classes, educational levels, and the use of PPEs. The short-term risk assessment of imidacloprid did not show any risk as the IESTI values were all below 100% of the ARfD.

Farmers need to be aware of existing legislation and preferably should follow adequate educational programs to implement sustainable agricultural practices.
Monitoring tools such as an analytical laboratory in Suriname, sufficient meteorological stations and legislative control measures (e.g. border control for illegal import of prohibited pesticides) are necessary to ensure food safety in Suriname. In addition, food-monitoring studies must be executed on a regular basis. The Surinamese government can implement action plans such as those given within the EU directive on sustainable use of pesticides (EU Directive 2009).

Chapter 4 Assessment of the quality of different environmental compartments in the research area (Commewijne)

Abstract
In this chapter the quality of surface water and sediment, the microbial quality of irrigation water and the vegetation diversity are assessed. Information can be used to review the potential of constructed wetlands to reduce or remove contaminants from agricultural runoff and to validate data received on pesticide usage during the G.A.P.-survey (Chapter 3). Surface water samples contained the fungicide chlorothalonil and the insecticides, lambda-cyhalothrin and α-endosulfan. The insecticides lambda-cyhalothrin and imidacloprid were also found in the sediment. In several of the water samples from the drainage ditches in the farmland residues of lambda-cyhalothrin and imidacloprid were found, while chlorothalonil was not detected. This may indicate a rapid dissipation from the water. The general water quality of the ditches in the farmland was good, with the exception of one location, for which values higher than the norm were found for Total Dissolved Solids (TDS) and the electrical conductivity (EC_{cond}). Results showed the presence of total and fecal coliforms, and *Escherichia coli* in numbers above the WHO guideline, indicating a poor microbial quality. Plants frequently detected in the main drainage ditches were inventorised. The use of surface water for irrigation purposes especially during dry seasons, poses a health risk to users of this water and consumers of vegetables, not only because of the pesticides detected, but also because of the microbial quality was impaired.

4.1 Introduction

At the start of this research, no data were available for Suriname on which type and in what concentration pesticides are present in different environmental compartments (e.g. surface water, plants and sediment); neither about the microbial quality of irrigation water and the types of (wetland) plants present in different water bodies. It was, therefore, difficult to take decisions on which pesticides, which concentration range and which plants to use in constructed wetland studies. There are several possibilities for water bodies to become contaminated with pathogens (Figure 4-1). As described in Chapter 1, farmers in the research area do not have a sewer system and untreated wastewater from e.g. households reach surface water systems mostly by means of tubes placed by them. Most farmers do not have a fence, allowing the intrusion of animals into the agricultural field with the possibility of fecal contamination of adjacent water bodies (Gerba and Choi 2006). In the research area, these were mostly domestic animals. During the dry season, ditch water is used for irrigation purposes. Irrigation performed with water of poor microbiological quality can potentially spread bacteria, viruses and parasites to crops.

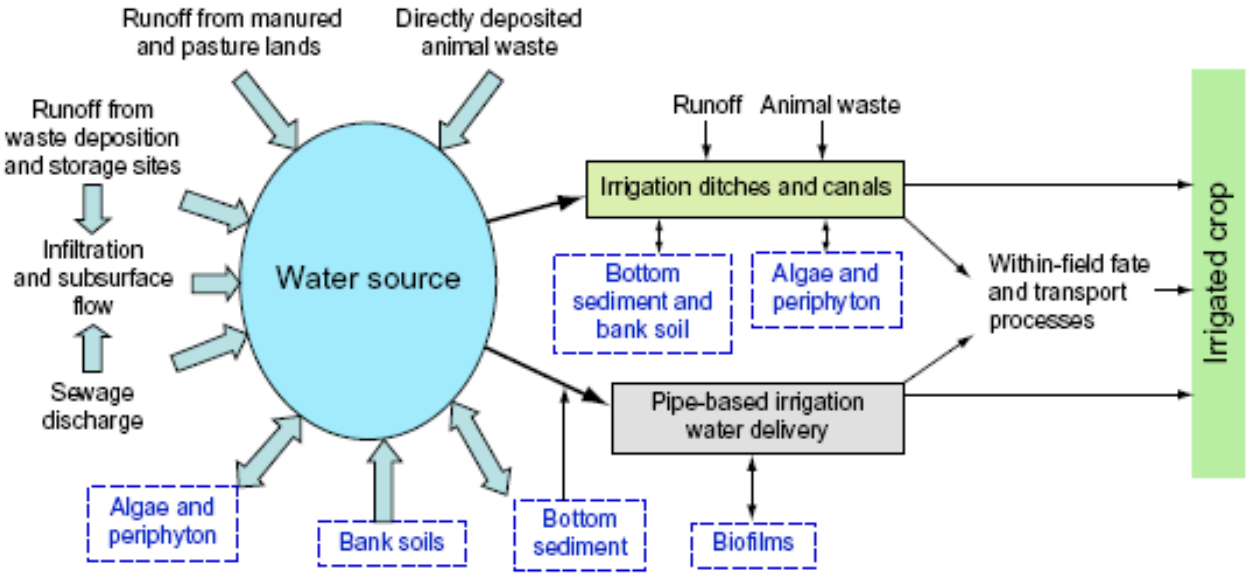

Figure 4-1: View of processes and reservoirs affecting the microbial quality of irrigation waters (Pachepsky et al. 2011)

The aim of this chapter is to assess the types and amounts of pesticides present in surface water and sediment and to determine the microbial quality of irrigation water in the Commewijne district. In addition, water samples from drainage ditches present on the farmland were analyzed on 3 of the frequently used pesticides. The information can be used to assess the possibility of constructed wetlands to reduce or remove these contaminants from agricultural runoff (Decamp and Warren 2000; US EPA 1995) and to validate data received on pesticide use during the G.A.P.-survey. The vegetation diversity was inventarised in the two main drainage ditches in the Alkmaar area in Commewijne. This allows choosing the type of plants to be used in future wetland experiments. The rationale was to choose those plants that can sustain their selves (frequently occurring) in an environment with pesticides e.g. a drainage ditch receiving agricultural runoff water.

4.2 Material and methods

4.2.1 Identification of study location ("areas of concern") and sampling sites

The study was executed in district Commewijne, Suriname. The district has a population of 31,420 and an area of 2,353 km² (Census statistieken 2012). Interviews with agricultural extension officers, local farmers and a hydrologist (A. Amatali from the WLA ("Water Loopkundige Afdeling" of the Ministry of Public Works) helped in identifying the locations to sample the surface water (small creeks, main and small drainage ditches present at the farmland) and sediment. Selection of the most vulnerable sites to pesticide contamination was further based on hydrology. Figure 4-2 presents the main waterways (Suriname River, Commewijne River and the Cottica River) and their connectivity with smaller waterways (creeks, swamps).

For surface water sampling, W2 (Figure 4-2) was chosen as a reference point upstream from the two main agricultural regions, namely Alkmaar and Tamanredjo. Choosing this reference point will give more insight in pesticide concentrations in surface water, not influenced by the research area. Within the Alkmaar region, sampling points W3 up to W6 were selected, because they were located in 1 of the 4 main ditches in the Alkmaar area (see Table 4-1, ditch D4). This ditch receives surface water from a total area of about 240 hectare (Abdoel 2011). Coordinates were measured by means of a GPS system (Brand: Garmin). The other sampling locations were selected in a similar manner and in small creeks (e.g. Orleane creek, Commetewane creek and Mindriniti creek) which receive runoff from smaller agricultural areas such as Slootwijk, Sinabo and Canawatibo (the last two cannot be viewed on the hydrological chart). Sediment sampling was done at the same locations as for water sampling. For water and sediment sampling, the main ditches from the Alkmaar area were also selected because they frequently receive runoff from agricultural fields and because of a more dense population of farmers compared to other areas such as Nieuw Amsterdam and Meerzorg.

Sampling of surface water took place in three periods: 1) May-June 2009 (long rainy season), 2) May to June 2011 (long rainy season) and 3) August 2011. From May to June 2011, water from the ditches located near the farmland was analyzed on 3 types of pesticides, while in August 2011, water samples of 6 main drainage ditches in the research area (Alkmaar region) was analyzed on the presence of imidacloprid. The other pesticides from the first period were not analyzed. Sediment sampling was only done in period three (August 2011) and both imidacloprid and the pesticides from period 1 were analyzed. The period May up to June was chosen and based on information retrieved from the Ministry of Agriculture in Commewijne and from the GAP survey. It was found to be a period in which most agricultural activities took place and pesticides were frequently applied. In addition, it was expected that rainfall would also facilitate transport of pesticides from the field to these surface water sampling locations. For assessment of the concentration of pesticides in sediment from surface water, August 2011 was chosen because the surface water level would be low (this would benefit sediment sampling) and pesticide concentrations higher compared to the rainy seasons. The different pesticides analyzed are presented in section 4.2.2.

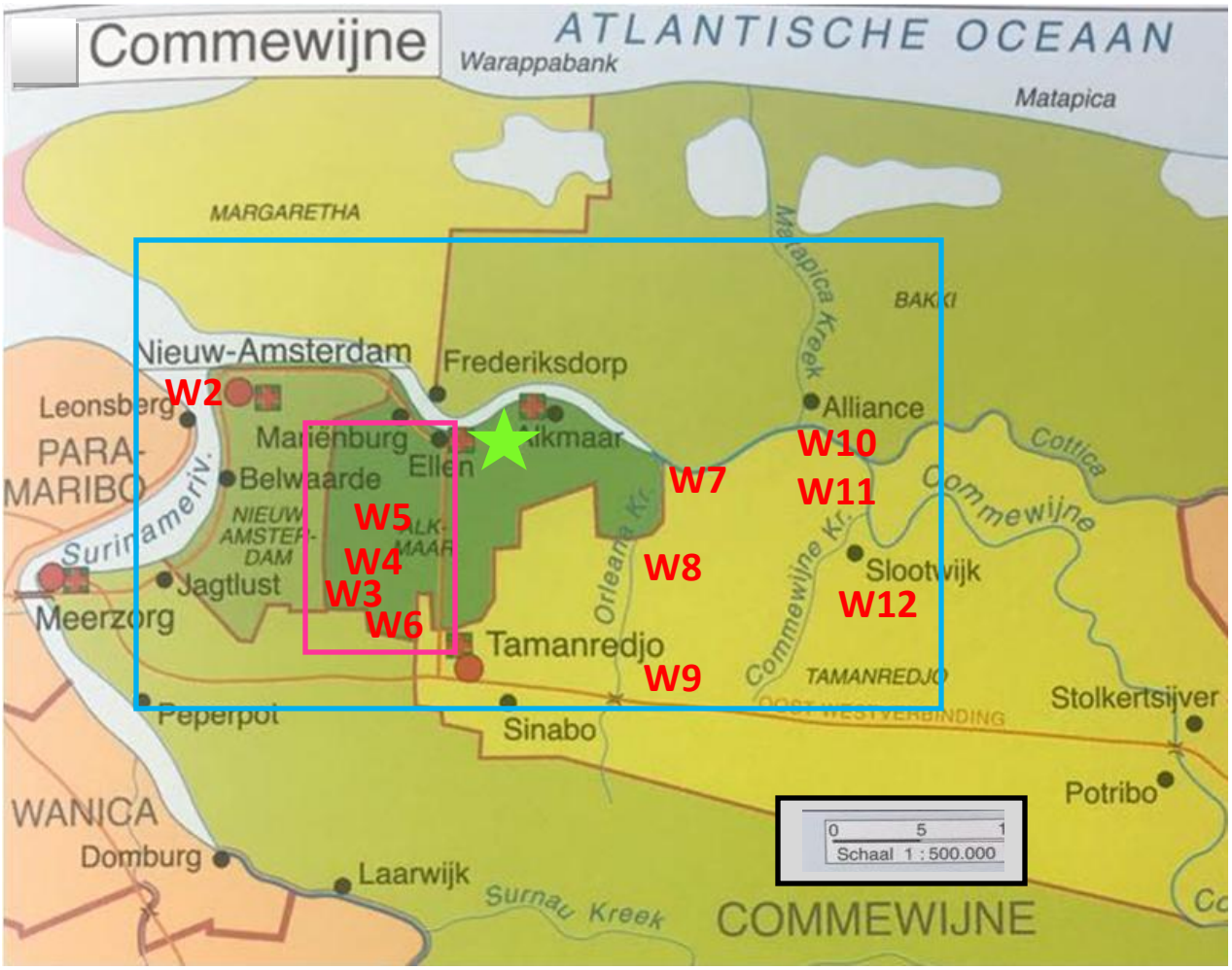

Figure 4-2 : Chart of district Commewijne, with the main research area presented by the blue rectangle and the locations used for sampling of ditch water/ditch sediment presented by the smaller purple rectangle. Surface water samples are numbered W2 up to W12. The green star represents the located were the field wetland described in chapter 7 is located (Atlas Suriname Wereldland 2008)

Table 4-1: Description of surface water and sediment sampling locations

Sample	Location	Coordinates ^{0}N/^{0}W
W2	Marienburgpier	0576270 ^{o}N/054.76914 ^{o}W
W3	Pt. Tilakdhariroad	0583330 ^{o}N/055.01867 ^{o}W
W4	Pt. Tilakdhariroad	0583839 ^{o}N/055.02107 ^{o}W
W5	Across Mohan store	0583911 ^{o}N/055.02101 ^{o}W
W6	Pt. Tilakdhariroad	0582915 ^{o}N/055.01895 ^{o}W
W7	Orleanebridge	0577483 ^{o}N/054.97172 ^{o}W
W8	Orleanestart	0582065 ^{o}N/055.02238 ^{o}W
W9	Orleanemid	0577835 ^{o}N/054.97140 ^{o}W
W10	Commetewanecreek	0576946 ^{o}N/054.89846 ^{o}W
W12	Mindrineticreek	0578110 ^{o}N/054.89012 ^{o}W
D1[a]	Drainwallerwegleft	0582220 ^{o}N/055.62501 ^{o}W
D3	Drainwallerwegright	0582224 ^{o}N/055.02503 ^{o}W
D4	Drainalkmaarright	0583926 ^{o}N/055.02378 ^{o}W
D5	Drainzorgvlietright	0583931 ^{o}N/055.02385 ^{o}W
D6	Drainalkmaarleft	0583923 ^{o}N/055.02376 ^{o}W
D7	DrainZorgvlietleft	0583928 ^{o}N/055.02383 ^{o}W

[a]*D1, D3 up to D7 are the main drainage ditches of Alkmaar*

In the May-June 2011, water from drainage ditches present on the farmland, receiving runoff after irrigation or rainfall, were also sampled in a similar way as for surface water (see section 4.2.2). Thirty sampling points within the Alkmaar area (Commewijne) were selected to determine the ditch water quality. Analysis was done on pesticide residues of three of the most frequently used pesticides (chlorothalonil, imidacloprid and lambda-cyhalothrin) and the general water quality parameters (Temperature (T), Dissolved Oxygen (DO), acidity (pH), Total Dissolved Solids (TDS) and conductivity (EC_{cond}). Compared to surface water testing (see Table 4-1 for sampling locations) a different approach was followed (e.g. different pesticides were analyzed) and results are therefore presented separately. Within a distance of 8 km of the main road, 22 sampling points were selected. For the side roads: Wallerweg and Tje Tjeweg, these were 6 and 2, respectively. The strategy was to take water samples from the drainage ditches of farmers, immediately after farmers' application of pesticides, followed by irrigation/rainfall. This was necessary to have more insight in the actual amount of the pesticides present in these ditches/runoff.

For the microbial quality assessment of irrigation water, locations were chosen which were vulnerable to contamination with e.g. faeces and where water was used for irrigation purposes. The microbial parameters, total coliforms, fecal coliforms and *Escherichia coli* were used as indicators for possible contamination of water with faeces. To investigate this, 17 locations were sampled, in the period of September through December 2009 of which 10 belonged to phreatic groundwater and 7 to surface water. Within Commewijne (the Alkmaar and Tamanredjo regions), the following locations were chosen: Tamansarie, Tamanredjo, De Hulp, Canawatibo, Alkmaar, Orleanecreek, San Souci, Mariëndaal, Welbedacht and Mijn Geluk.

For the study on vegetation diversity, the ditches draining most of the agricultural fields and which were almost fully vegetated were selected. Plants in these ditches were frequently exposed to chemicals such as pesticides, present in the runoff coming from the agricultural land and therefore they might have developed mitigation strategies for their removal/reduction.

4.2.2 Method of water and sediment sampling and analysis

For water sampling, an uncapped 1-L Nalgene bottle was held in the water with the mouth pointing in the direction of the flow. As the water deepened, the bottle was moved up and down to collect a sample representative of the water column. This was repeated after 10 minutes for 3 samples and all samples were subsequently mixed, resulting in a composite sample for one location. Sediment was collected using a tube soil corer, 2 cm in diameter and 25 cm long. At each location, 4 cores were collected and combined. Samples were kept on ice immediately after sampling, frozen and stored at -20 °C. Samples were sent by means of DHL express to the laboratory for Crop Protection Chemistry, UGent for further analysis.

The surface water samples and sediment samples were analyzed on the presence of six different pesticides representing different chemical classes. Chlorothalonil and endosulfan belong to the chemical class of organochlorines, cypermethrin and lambda-cyhalothrin to the pyrethroids, chlorpyrifos to the organophosphates and imidacloprid to the neonicotins). Sampling was done with the following exceptions: a) in period 1 (2009) imidacloprid was not analyzed; b) in period 2 (May-June 2011), only 3 of the most frequently used pesticides (chlorothalonil, imidacloprid and lambda-cyhalothrin) were analyzed and c) in period 3 (August 2011) only imidacloprid was analyzed in water from six main drainage ditches, while for sediment analysis, all pesticides from period 1 were analyzed, including imidacloprid.

The analytical procedures involved the extraction of samples (water and sediment) with a suitable solvent. For the organochlorines/pyrethroids: α-endosulfan, lambda-cyhalothrin, chlorothalonil and cypermethrin, a mixture of hexane/acetone (1:1) was used for the extraction, followed by drying over anhydrous sodium sulphate, evaporation until dryness, and dissolution in hexane. For the organophosphates, ethyl acetate was used as a solvent, and a method similar as for the organochlorines/pyrethroids was followed.

For quality control purposes, a blank reagent was incorporated, simultaneously with freshly prepared standard solutions, and sample extracts within each set of tests performed with the GCMS or HPLC. The blank reagent consisted of the same matrix (solvent) as standard solutions and sample extracts, however, without pesticides. The sensitivity of the analytical method was evaluated by the determination of the limit of detection (LOD) by means of the signal-to-noise (S/N) ratio criteria, namely LOD equaling 3 S/N (US EPA 2000), Table 4-2).

The quantification of organochlorines, pyrethroids, and organophosphates was done by means of gas chromatography - mass spectrometry (GC-MS). A 6890N gas chromatograph (Agilent Technologies) equipped with an HP-5ms column was used. For the analysis of neonicotinoid

imidacloprid, a method described by Baskaran et al. (1999) was used, involving an extraction with 40 ml of a mixture of acetonitrile + water (80:20 Volume/Volume) followed by centrifugation and filtration of the supernatant through a PVDF membrane, described above. Imidacloprid was quantified by means of HPLC (Thermo Surveyor) equipped with a surveyor LC pump plus a Surveyor PDA plus 5 detector (Thermofisher) and a platinum C18ESP 3 µm column.

Table 4-2: Detection limits for different pesticides in water and sediment

Pesticide	LOD in water (µg/l)	LOD in sediment (µg/kg or mg/kg)
α-endosulfan	0.02 µg/l	0.02 mg/kg
Chlorpyrifos	0.02 µg/l	0.02 mg/kg
Lambda-cyhalothrin	0.001 µg/l	0.01 µg/kg
Chlorothalonil	0.1 µg/l	0.02 mg/kg
Imidacloprid	0.5 µg/l	0.5 mg/kg

The quantification of organochlorines, pyrethroids, and organophosphates was done by means of gas chromatography - mass spectrometry (GC-MS). A 6890N gas chromatograph (Agilent Technologies) equipped with an HP-5ms column was used. For the analysis of neonicotinoid imidacloprid, a method described by Baskaran et al. (1999) was used, involving an extraction with 40 ml of a mixture of acetonitrile + water (80:20 Volume/Volume) followed by centrifugation and filtration of the supernatant through a PVDF membrane, described above. Imidacloprid was quantified by means of HPLC (Thermo Surveyor) equipped with a surveyor LC pump plus a Surveyor PDA plus 5 detector (Thermofisher) and a platinum C18ESP 3 µm column.

4.2.3. Method of irrigation water sampling and assessment of microbial quality

Samples were collected aseptically in 500 ml sterile plastic (polypropylene) bottles and were transferred in an ice box to the laboratory. The microbial analysis (Most Propable Number (MPN) counts) were executed by “het Centraal Laboratorium, afdeling Bacteriologie, van het Bureau voor de Openbare Gezondheidszorg” Suriname. The US EPA accepted Method 8001 (MPN method) was used. Total coliforms were confirmed by means of BGB (Brilliant Green Bile) broth, fecal coliforms with EC medium broth and *E. coli* with EC/MUG (4-methylumbelliferyl-β-D-glucuronide) medium broth and the use of a UV lamp (365 nm) to samples containing *E. coli* will fluoresce. Results obtained were compared with the criteria from the World Health Organization (WHO 1989).

4.2.4. Method of vegetation diversity assessment

Plant coverage can be estimated by determining the relative coverage (abundance) by different plant species in plots or blocks (Austin 2007). This assessment was done for two main ditches present on the left and the right side of the Pt. Tilakdhari road in Alkmaar, Commewijne (located within the smaller purple rectangular in Figure 4-2) and was executed in August 2011. The approximately 8 km road was divided in 76 blocks: 38 to the left and 38 to

the right. The different plants were identified by Mrs. Angela Grant from the National Herbarium of Suriname at the Anton de Kom University of Suriname. Determination up to the species level could not be conducted for all observed plants, because no expert was available for the identification of grasses in Suriname. The abundance per block was determined by observation (counts), comparing the appearance of the relevant species with the total number of different species. A count equals the presence of a plant species in a block, leaving the possibility for more counts of that plant species over the different blocks. Beside the abundance, the coverage for the plant species that were mostly present in the ditch was calculated by means of Formula 4-1. The sum of the maximum abundance over the 75 blocks was set at 7500%, taking into consideration that the maximum abundance per block is 100%.

Coverage: $\left[\frac{sum\ of\ abundance\ of\ 1\ plant\ specie\ over\ 75\ blocks\ (\%)}{sum\ of maximum\ abundance\ for\ the\ 75\ blocks\ (\%)}\right]$*100% *(Formula 4-1)*

4.3. Results and discussion

4.3.1 Pesticide residues in surface water

The residues found in water samples analyzed in 2009 are shown in Table 4-3.

Table 4-3: Amount of pesticides detected in surface water samples from Commewijne and comparison of these values with international guidelines. Values higher than the guideline are presented in red

Sample	Coordinates	Chlorothalonil(µg/l)	Λ-cyhalothrin (µg/l)	α-endosulfan (µg/l)	Imidacloprid (µg/l)
W3	0583330 °N 055.01867 °W	147	12[a]	ND[b]	NT[e]
W4	0583839 °N 055.02107 °W	86	21	11.9	NT
W5	0583911 °N 055.02101 °W	88	29	ND	NT
D6	0583923 °N 055.02376 °W	ND	ND	ND	0.64
D7	0583928 °N 055.02383 °W	ND	ND	ND	1.83
NORM		**0.18[a]**	**0.00047[c]**	**2[d]**	**0.23[a]**

[a] *CCME (Canadian Council of Ministers of the Environment) (1994; 2007; 2010)*
[b] *Not detected*
[c] *KRW (European 'Kaderrichtlijn Water' (2012))*
[d] *Footprint-International Union of Pure and Applied Chemistry-Pesticide Properties Dababase (IUPAC-PPBD) (2015)*
[e] *Not analyzed because no method was available to quantify imidacloprid in water in 2009*

The results obtained for the testing of imidacloprid in water from main drainage ditches in 2011 is also given. Results for 2009 show that 3 of the 16 water samples contained the fungicide chlorothalonil and the insecticides lambda-cyhalothrin and α-endosulfan, while imidacloprid wat not analyzed because of analytical restrictions.

The residues were above the norm set by the Canadian Council of Ministers of the Environment (CCME 1994; 2007; 2010) for surface water and may pose a health threat to the environment (humans, animals and aquatic life). Humans can be exposed by means of e.g. consumption of contaminated water and crops. During very dry periods water from these contaminated locations is used for irrigation purposes.

The water samples from the main drainage ditches, D1, D3 up to D7 were analyzed in 2011 on the presence of imidacloprid. Two water samples contained a residue of imidacloprid, which were both above the norm (Table 4-3).

According to Mohr et al. (2012), a NOEC (No Observed Effect Concentration) of 0.6 µg/l imidacloprid was derived from results of a pond mesocosm study, which was pulsed once with 10.7 µg/l imidacloprid. The measured imidacloprid concentrations were above this NOEC. According to Fossen (2006), concentrations up to 36 µg/l were linked to acute surface water exposure. In Jemec et al. (2007), however, much higher acute surface water exposure concentrations (up to 320 µg/l) are provided. The norm for lambda-cyhalothrin could not be found in the CCME database, but for other pyrethroids the norm for the protection of aquatic life was more stringent (e.g. deltamethrin 0.0004 µg/l and permethrin 0.004 µg/). According to the Water Framework Directive of the European Parliament, the Maximum Acceptable Concentration-Environmental Quality Standard (MAC-EQS) for surface water is set at 0.00047 µg/l (KRW 2012).

From the survey on good agricultural practices, the organochlorine pesticide endosulfan was found to be used by 1 of the 78 farmers (see Chapter 3). In the study of Dalvie et al. (2003), endosulfan was detected in 48% of surface water samples (of a total of 185) in the Hex River valley, present in the farming region of the Western Cape, South Africa, with concentrations ranging between 0.02 µg/l and 2.22 µg/l. These results appeared to peak in mid-points and were influenced by dilution in points downstream or after they confluence with a tributary. This effect could not be observed in the present study because endosulfan was detected in only one point (W4). This sampling point was located in the main drainage ditch of the Alkmaar region. Because of its properties (a half-life time in soil of 120 days and sorption coefficient (K_{oc}) of 17.52 L/g) endosulfan is persistent (Quinete et al. 2013). Considering these properties, the low water solubility and the low flow in the ditch, endosulfan was not measured in other points within this ditch. The localization in one point may also indicate the outcome of a point source contamination. For endosulfan, which is banned in the EU and toxic to aquatic organisms, mammals, birds, honeybees and earthworms, different toxicological endpoints are presented in the Footprint IUPAC-PPBD database (2015). The acute 96 hour LC_{50} (mg/l) to fish (*Oncorhynchus mykiss*) is 0.002 mg/l, while the acute 48 hour EC_{50} (mg/l) to aquatic invertebrates (*Daphnia*

magna) is much higher (0.44 mg/l or 440 µg/l). The detected concentration of 11.9 µg/l in the current study poses a health risk to aquatic organisms, especially fish.

During the present study in Commewijne, not all pesticides used in the research area were analyzed. Therefore, there is a possibility that some pesticides were present, but could not be analyzed due to analytical restrictions. The residue testing of water and sediment was conducted only once. A more frequent sampling strategy would give a better representation of the pesticides present in the different sampling points, especially when considering factors such as the time of pesticide applications and growing season. It would also be interesting to test water and sediment from one location simultaneously e.g., high lambda-cyhalothrin concentrations in water might also indicate its presence in the sediment phase.

4.3.1.1 Pesticide residues in the sediment from surface water

Lambda-cyhalothrin was detected at a concentration of 0.050 µg/g dry weight (DW) sediment in only 1 of the 16 sediment samples, namely in W5, across Mohan store. The concentration was much higher than the acute toxicity threshold towards *H. azteca* (5.6 ng/g DW sediment) (Amweg et al. 2005). It was concluded that this pesticide was frequently used. In addition, it was also expected to be detected more frequently in the sediment phase, especially because of its persistence nature in the soil/sediment phase. Other pesticides like chlorothalonil, endosulfan, cypermethrin and chlorpyrifos were either not present or were below their limit of detection. This seemed also the case for lambda-cyhalothrin. Other possibilities of this pesticide not being detected, was that it was not applied during the period of the sediment residue testing or the information provided by the farmers about this pesticide during the G.A.P. survey was not reliable.

The neonicotinoid imidacloprid was detected in all sediment samples (see Table 4-4). High concentrations were found in sampling points W2, W3 and W9. Locations W2 (reference point for the Alkmaar and Tamanredjo regions) and W9 are areas were fishing activities take place and W3 is a drainage ditch from the Alkmaar area. The concentration of imidacloprid found in point W2 indicates that imidacloprid was not related to agricultural activities in the Alkmaar and Tamanredjo regions, but can be the result of agricultural activities of the resort Nieuw Amsterdam (Figure 4-2), where farmers also cultivate land.

Table 4-4: Imidacloprid concentrations (µg/g DW) detected in sediment samples of Commewijne. The sampling locations and moisture content of the sediment samples are also presented.

Sample	Description	Moisture content (%)	µg/g DW sediment
W2	Marienburgpier	35.2	16.2
W3	Pt. Tilakdhariroad	41.1	7.8
W4	Pt. Tilakdhariroad	33.2	1.26
W5	Across Mohan store	36.8	2.85
W7	Orleanebridge	22.6	2.35
W8	Orleanestart	57.2	1.04
W9	Orleanemid	37.0	43.1
W10	Commetewanecreek	19.5	1.10
W12	Mindrineticreek	33.5	0.65
D1[a]	Drainwallerwegleft	48.7	1.66
D3	Drainwallerwegright	43.6	1.57
D4	Drainalkmaarright	42.1	1.49
D5	Drainzorgvlietright	38.5	6.0
D6	Drainalkmaarleft	47.3	1.73
D7	Drainzorgvlietleft	53.2	2.75

Imidacloprid is authorized in EU Member States, where a variety of application techniques such as foliar sprays, soil drenches, dipping solutions, irrigation, drip irrigations, rodlets (also known as plant sticks) and stem applications, namely brush and trunk injections, are applied (EFSA 2015). The half-life of imidacloprid in soils in the absence of light (comparable to the sediment environment) is 229 days. Depending on the concentration, imidacloprid is able to disrupt nicotinic acetylcholine receptors in the insect central nervous system (Fossen 2006). Sub lethal effects were found on sediment dwellers (*H. incongruens*) exposed to imidacloprid residues ranging between 25 µg/kg and 100 µg/kg in sediment from a rice plot. Growth rate inhibition was higher (16% more inhibition) in sediment samples taken 5 days post application than in those taken 8 h after application. It is related to the slow translocation of this pesticide from the water phase to the sediment (Daam et al. 2013). All imidacloprid residues found in sediment (Table 4-4) were much higher than the above-mentioned range (e.g. the lowest concentration measured was 650 times higher than the lowest value of 25 µg/kg) and might pose a risk to organisms present in the sediment.

4.3.1.2 Ditch water quality assessment

The ditch water quality assessment refers to the general water quality assessment and the residue analysis done for the pesticides chlorothalonil, imidacloprid and lambda-cyhalothrin in 30 small drainage ditches near the farmland in Alkmaar, Commewijne. The results are presented in section 4.3.1.2.

General water quality

The range of values obtained for the general water quality parameters: dissolved oxygen (DO), temperature (T), acidity (pH), total dissolved solids (TDS), and electrical conductivity (EC) are presented in Table 4-5.

Table 4-5: Results for general water quality parameters of small ditches (range presented for 30 samples) near farmers agricultural land in Alkmaar, Commewijne and comparison with international guidelines of Belgium (Vlarem II) and the Food and Agriculture Organization of the United Nations (FAO)

Sample	DO (mg/l)	T (^{0}C)	pH	TDS (mg/l)	EC_{cond} (µS/cm)
Range	5.30-11.53	27.1-35.2	5.8-9.2	25.2-910	36.1-1302
NORM	**6**[a]	**25-28**[a]	**6-8.5**[b]	**0-2000**[b]	**0-3000**[b]

[a] *VLAREM II*
[b] *FAO (1985)*

Values for TDS and the EC_{cond} were found to be higher than their respective norms, but this was observed for one measuring point only. Depending on the tide and opening of the drainage sluice, salt water (from the nearby Commewijne River) may flow land inwards and reach the drainage ditch location.

Pesticide residues in water from ditches at the farmland

For the 30 ditch water samples analyzed, the following results were obtained: no residues were found for chlorothalonil; lambda-cyhalothrin (range 0.71-3.8 µg/l) was found in 6 samples and imidacloprid (range 0.25-3.9 µg/l) in 8 samples. The measured values for lambda-cyhalothrin and imidacloprid were much higher than the 0.00047 µg/l norm of the KRW (2015) and the 0.23 µg/l set by the CCME (2010). These values give an estimate of which pesticide concentrations can be expected in ditch water from the Alkmaar areas and can be used for wetland feasibility studies to mitigate pesticide pollution originating from agricultural runoff. Results must also be validated by means of field runoff experiments using the recommended application rates, and testing of the water phase of drainage ditches immediately after rainfall or a simulated irrigation. The three selected pesticides belonged to the frequently used pesticides. It was also expected to detect chlorothalonil in the ditch water. It was, however, not the case. This might indicate that chlorothalonil is rapidly removed from the water phase or it is a bias in the G.A.P. survey. The first seems more obvious, because chlorothalonil is volatile and it rapidly dissipates from water with half-life times of 0.1 days (PPDB 2016).

4.3.2 Microbial quality

The results for the microbiological analysis are divided into results for phreatic groundwater (GW) and surface water (SW). They are presented in Table 4-6 and Table 4-7, respectively.

Table 4-6: Results for microbiological analysis of phreatic groundwater (September, November and December 2009) and the international guideline of the WHO (values in red are above this norm)

GW Sample[a]	TC[b] Sept.	TC Nov.	TC Dec.	FC[c] Sept.	FC Nov.	FC Dec.	E. coli[d] Sept.	E. coli Nov.	E. coli Dec.
					MPN/100 ml				
GW1	700	6.9	900	400	6.9	700	0	0	400
GW2	200	1700	1400	200	1700	1100	200	200	400
GW3	0	28000	30000	0	28000	50000	0	11000	50000
GW4	7000	12	3300	7000	12	2200	1700	0	0
GW5	1700	400	7000	1700	400	3000	200	400	1300
GW6	2200	5000	2200	1700	2200	1700	400	400	200
GW7	400	2200	900	400	1300	900	200	200	0
GW8	400	Dry[e]	Dry	400	Dry	Dry	0	Dry	Dry
GW9	1300	3400	11000	1300	1100	11000	200	400	800
GW10	0	9.2	Dry	0	6.9	Dry	0	0	Dry
NORM					**1000 MPN/100 ml**				

[a] *GW 1-GW5: Alkmaar; GW 6: Welbedacht; GW 7: De Hulp; GW 8: Mariëndaal; GW 9: Tamanredjo; GW 10: Tamansari*
[b] *Total Coliforms*
[c] *Fecal Coliforms*
[d] *Escherichia coli*
[e] *The sampling Location was dry and water could not be sampled*

The parameters analyzed were total coliforms, fecal coliforms and *Escherichia coli*. Samples were collected on September 4, November 10 and December 9, 2009. On all locations, detections were done for the parameters indicating fecal contamination.

4.3.2.1 Phreatic groundwater

Results presented in Table 4-6 show that for September 2009, total and fecal coliforms were detected in 80% and *E. coli* in 60% of the water samples. From the detections, 50% of fecal coliforms were higher than the norm of 1000 MPN per 100 ml set by the World Health Organisation (WHO). For *Escherichia coli,* this value was 17%. Results obtained for November and December 2009 show that total and fecal coliforms were detected in all samples. From those detections, 56% (November) and 75% (December) of fecal coliforms were above the norm. In November and December, respectively, 67% and 75% of *Escherichia coli* was detected. From these detections, respectively 17% and 33%, were above the norm.

4.3.2.2 Surface water

Results for the microbiological analysis of surface water (SW) are presented in Table 4-7. The same parameters were analyzed as for phreatic groundwater. Measurements were done on September 9, November 12 and December 9, 2009. Results show that both total and fecal coliforms were found in all samples.

Table 4-7: Results for microbiological analysis of surface water (September, November and December, 2009) and the international guideline of the WHO. Values higher than this guideline are presented in red

SW Sample[a]	TC[b] Sept.	TC Nov.	TC Dec.	FC[c] Sept.	FC Nov.	FC Dec.	E. coli[d] Sept.	E. coli Nov.	E. coli Dec.
					MPN/100 ml				
SW1	400	2300	200	400	2300	0	400	800	0
2SW	400	5000	900	200	900	900	200	600	200
SW3	7000	2600	400	7000	1700	400	7000	1700	200
SW4	1400	1400	90	1100	800	30000	700	0	600
SW5	1400	> 160000	1700	1400	> 160000	1700	400	7000	900
SW6	1700	Dry[e]	Dry	1700	Dry	Dry	1400	Dry	Dry
SW7	2300	Dry	Dry	2300	Dry	Dry	0	Dry	Dry
Norm					**1000 MPN/100 ml**				

[a] *SW 1: Canawatibo; SW 2: Orleanekreek; SW 3: Welbedacht; SW 4: De Hulp; SW 5: Mijn Geluk; SW 6: San Souci; SW 7: Tamansari*
[b] *Total Coliforms*
[c] *Fecal Coliforms*
[d] *Escherichia coli*
[e] *The sampling location was dry and water could not be sampled*

Results for September 2009 show that 71% of the detections were above the norm of 1000 MPN per 100 ml. For *Escherichia coli,* this value was 86%, with 33% crossing the norm, while for November 2009, 100% (total coliforms) and 60% (fecal coliforms) of detections were above the norm. For *Escherichia coli,* 80% of the samples were tested positive of which 50% were above the norm.
Results for December 2009 show that the total coliforms were detected in all and fecal coliforms in 80% of the samples, with respectively 40% and 50% above the norm. For *Escherichia coli* all samples were tested positive, but with values below the norm.
The total coliforms represent the *Escherichia-Aerobacter* group and are often used to determine process efficiency of e.g. chlorination. They are the least accurate in identifying fecal pollution, because failures have often been reported to indicate health risk from bacterial pathogens (WHO 2001). It is used as a general indicator of potential contamination with pathogenic organisms, however, many coliform bacteria live in the soil, and these organisms may be the source of those that appear in water, especially in surface water. The presence of total coliforms may or may not indicate fecal contamination (UNEP/WHO 1996). Fecal coliforms are more specific because they refer to the coliforms that live in the intestinal track of humans and many other animals. The clear indication of fecal contamination is, however, confirmed by the presence of *E. coli*.

According to Pachepsky et al. (2011), there is a lack of data on the microbiological quality of irrigation water worldwide. Several processes can affect the microbial quality of irrigation water such as different types of runoff, deposition of animal waste, discharge from leaky sewer lines and infiltration (Figure 4-1). The different microbial reservoirs are algae and periphyton, bank

soils, bottom sediment and biofilms. Exposure of water to the external environment (in the case of surface water) will lead to more contamination than when water is confined by soil layers (ground water). It addition, it may result in disease outbreak. Fecally contaminated irrigation water is often the source of pathogen contamination of fresh, ready-to-eat fruits and vegetables and may raise the number of food borne disease outbreaks (Bichai et al. 2012; Betts 2014; Hidayatullah 2014; Uyttendaele et al. 2015). Introduction of enteric pathogens from irrigation water is associated with either the source/type of water or the irrigation method. Surface water and wastewater are mostly of a lesser microbial quality compared to ground and rainwater, while the method of irrigation plays an important role in the mode of contamination and transfer of bacteria, viruses, or protozoa to produce. Several studies (Hamilton et al. 2006; Song et al. 2006; Cevallos-Cevallos et al. 2012) show that subsurface or drip irrigation lowers the risk of transfer of pathogens to growing plants compared to furrow and sprinkler irrigation. It minimizes the exposure of the irrigated water to the produce, and it lowers the risk of splashing of contaminated soil on vegetables. Based on field observations, most farmers in the Alkmaar and Tamanredjo area use the sprinkler method and a small number performs irrigation by hand (data not shown). The drip irrigation method is, however, not used in the study area.
As presented in Chapter 2, constructed wetlands are able to remove pathogens from various types of wastewater. A review on the removal of human pathogens and fecal indicators in different types of CW is presented in Wu et al. (2016). However, the use of a single stage CW is usually insufficient to reach environmental standards. In Chapter 2, it has been suggested to use a combined system of wetlands (hybrid system), such as VF combined with HF CW (Garcia et al. 2013) or a combination of VF-HF-VF CW (Tunçsiper et al. 2012) with the aim to increase pathogen removal.

4.3.3 Vegetation diversity

The number of counts done in all 75 blocks was 365. The most common plant species were Poaceae *Phragmites australis* (counts: 54 times, coverage: 21%), Araceae *Montrichardia arborescence* (counts: 51 times, coverage: 18%), Onagraceae *Ludwigia stolonifera* (counts: 39 times, coverage: 6%), Convolvulaceae *Ipomoea aquatica* (counts: 24 times, coverage: 6%), two types of grass species (*Gramine sp*. and *Cyperaceae sp*.) (counts: 14 times, coverage: 3%), Nymphaeaceae *nymphaea sp.* (counts: 12 times, coverage: 4%), Poaceae *Echinochloa polystachya* (counts: 6 times, coverage: 3%), Lemnaceae *Spirodela polarize* (counts: 15 times, coverage: 2% and Apiaceae *Hydrocotyle umbellata* (counts: 11 times, coverage: 1%).
The complete list of plant abundance per species per block is presented in Appendix 2, Table b. This study was a preliminary assessment to identify the type of plants in the main ditches of the Alkmaar area and their frequency of occurrence. The plants (*E. mutata, N.amazonum, P.australis and E. polystachya)* belong to the most counted species in the research area. They were therefore, taken up in the mesocosm studies (Chapter 5 and 6). Other influencing factors such as ditch water pH, TDS, DO, water depth, velocity were not measured. Wu et al. (2015) suggest that these water quality parameters have a linkage with the type of vegetation. In that study, analysis suggests that in ditches in Northern China, most plants are usually located in aquatic environments with low TDS concentrations, while nutrients (TN and TP) are significant

factors for plant growth. Also small-sized drainage ditches (< 5 m width) with shallow water depth and slow flow velocity (≤ 0.1 m/s) receiving primary agricultural drainage water, frequently keep more plant species and a higher diversity than large-sized drainage ditches. The frequency and cover of emergent plants, including *P. australis*, *T. angustifolia*, *S. triqueter*, and *S. trifolia,* increases in smaller sized ditches (depth < 70 cm), whereas submerged plants generally reduces in bed cover and frequency of appearances. Submerged plants tended to distribute in deeper water to obtain sufficient space for rooting and the growth of shoots (Squires and van der Valk 1992; Wu et al. 2015). The highest abundance in the present study was observed for *P. australis.* Several studies (Agudelo et al. 2010; Elsaesser et al. 2011; Maillard et al. 2011; Page et al. 2011; Imfeld et al. 2013) describe the mitigation capacity of this plant towards pollution involving a wide range of pesticides. They are investigated in different constructed wetland systems such as partially or fully vegetated. These wetlands were used for the treatment of respectively: the insecticide (chlorpyrifos); a mixture of herbicides (diuron, simazine and atrazine); a mixture of 5 pesticides (herbicides and insecticides); a mixture of 10 fungicides and 1 herbicide, and the herbicides glyphosate and AMPA. Other examples are the use of *Ludwigia* species for the mitigation of runoff containing chlorpyrifos (Moore et al. 2002), *Ipomoea aquatica* in studies involving nutrients (Lin et al. 2002; Chairuangsri et al. 2014) and heavy metal removal (Rai et al. 2009). More details on the type of CW, operational conditions, removal efficiencies and mechanisms are presented in Appendix 1, Table a.

4.4 Conclusions

The results of this study show that all investigated compartments (surface water, sediment and ditch water) were contaminated with pesticides. The main locations with pesticide detections were the drainage ditches located at the farmland, receiving agricultural runoff. The pesticides detected in surface water were the fungicide chlorothalonil and the insecticides α-endosulfan, lambda-cyhalothrin and imidacloprid. It should be noted that α-endosulfan is banned in Suriname and should not be used anymore. All detections where higher than their respective water norms and, therefore, may pose a health threat to humans and the ecosystem. Detection of pesticides in the reference point W2 indicated other sources of pollution than the local runoff from the Alkmaar and Tamanredjo regions. The microbiological quality of irrigation water was poor for both phreatic groundwater and surface water, but of lesser quality for surface water. It might therefore be harmful when used for e.g. irrigation purposes. Plants detected during the vegetation diversity assessment such as *Phragmites australis*, *Ludwigia species* and *Ipomoea aquatica*, are used in different CW studies involving pesticides, nutrients and heavy metal removal. This study was limited to the long rainy season and the start of the long dry season (August) and to the quantification of only a few of the most frequently used pesticides. Simultaneous analysis of water and sediment within one sampling point may provide a better insight in pesticide occurrence. Measurements done in other seasons and analysis of other types of pesticides will give a better understanding of the water, sediment and microbial quality.

Chapter 5 Mitigation of pesticides pollution by wetland plants: feasibility study for the treatment of agricultural runoff in Suriname (South America)

This chapter has been compiled from:
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ABSTRACT
In agricultural areas, pesticides can enter waterbodies by means of agricultural runoff. CWs are able to remove several pollutants including pesticides. Few studies exist from South America and therefore information is urgently needed on mitigation of pesticide pollution by native plants in water ecosystems. To this aim, mesocosm experiment using polypropylene tubs was set up. Two types of plants (*Nymphaea amazonum* and *Eleocharis mutata*) were planted in these tubs. The mesocosms were exposed to a low (10 µg/l) and high (30 µg/l) dose of lambda-cyhalothrin, while for imidacloprid, a low (60 µg/l), a high (180 µg/l) and an extra high (1000 µg/l) dose was applied. Batch experiments were performed for two weeks each. Up to 100% of lambda-cyhalothrin and imidacloprid dissipated in the water phase, although complete removal of imidacloprid in *E. mutata* mesocosms, was limited to batch 1 at time 216 h. Statistical two-way ANOVA analysis ($\alpha = 0.05$) showed that the removal of lambda-cyhalothrin was independent of the dose applied and the type of plant, while for imidacloprid, removal was dependent on the dose applied and the type of plant for the highest dose of 1000 µg/l. At the end of the last batch, analysis of plants and sediment showed that 48.5% of the applied amount of lambda-cyhalothrin was detected in the sediment and 0.4% in the upper parts (shoots and leaves) of *N. amazonum* plants, while the amount in roots was below the limit of detection. For *Eleocharis mutata* mesocosms, 44.6% of lambda-cyhalothrin was detected in sediment and 0.5% in roots. For *N. amazonum* mesocosms, 78.9% of the applied amount of imidacloprid was retained in plants (shoots and leaves and roots) and 17.3% in sediment, while for *Eleocharis mutata* mesocosms only 0.5% of imidacloprid was detected in plants (shoots and leaves and roots) and 15.4% in sediment. The different amounts of pesticides detected in different parts of the wetland mesocosms suggest that different processes such as (bio)degradation, photolytic degradation, adsorption, accumulation in plants and sedimentation might take place, which are e.g. related to pesticide properties, environmental conditions (such as pH, temperature, presence of sunlight) and the type of plants. The half-life time (DT_{50}) of lambda-cyhalothrin in the water-phase of both types of mesocosms was on average 1 day and between 1-6 days for imidacloprid, with median values of around 1 day and 6 days respectively. The results obtained, provide input data for the design and construction of a field scale wetland.

5.1 Introduction

The use and efficiency of constructed wetlands (CWs) for the removal of pesticides from polluted waste streams such as agricultural runoff, has been demonstrated in several studies (Budd et al. 2009; Elsaesser et al. 2011; Moore et al. 2013; Vallée et al. 2015A: 2015B; Vymazal and Březinová 2015; Maillard et al. 2016). CWs have been used successfully in removal/retention of pesticides and other constituents in agricultural runoff and can prevent and reduce their amounts in surface water. Results of mesocosm studies can be used in designing such systems on larger (e.g. field) scale. First order disappearance rates can be expressed as chemical half-lives (DT_{50}). One of the first steps in the design is estimation of these values. The half-life depends on the chemical nature of the compound and varies with wetlands characteristics, such as vegetative cover, vegetation type, climatological conditions, and other factors (Focus 2006). Literature values for chemical half-lives can be unreliable for a CW design, because they are not obtained from wetlands studies (WRP 1994). Therefore one objective of this study, was to determine the persistence (DT_{50}) of pesticides in the water phase of a CW. The main processes involving the removal of pesticides were described in chapter 2 and are mainly dependent on pesticide properties like DT_{50}, solubility, K_{ow} (octanol-water partition coefficient), vapour pressure and K_d (sorption coefficient). In vegetated wetland systems, uptake of pesticides by plants is dependent on different factors such as pesticide chemistry, plant lipid content, the hydraulic retention time and the use of mono-cultures of multiple plant types (Moore et al. 2009B; Stehle et al. 2011; Moore et al. 2013).

This study is one of the first studies conducted in Suriname and South America and comprises the Commewijne area. No data is available in the literature on the removal of the pesticides lambda-cyahlothrin (a pyrethroid insecticide) and imidacloprid (a neonicitinoid insecticide) in wetland systems planted with *Eleocharis mutata* and *Nymphaea amazonum*. The major properties of both pesticides are presented in Table 5-1. Imidacloprid (application rate between 0.24-0.6 kg/ha) has a high water solubility, a low vapour pressure, and a low octanol-water partitioning coefficient and is therefore considered to have a high runoff potential into surface waters (Fernάndez-Bayo et al. 2007; CCME 2007). Imidacloprid belongs to the neonicotinoid class of synthetic organic insecticides and is widely used to control both piercing and sucking insect pests around the world (Gupta et al. 2016). In contrast to imidacloprid, lambda-cyhalothrin (application rate between 0.01-0.02 kg/ha) has a very low water solubility, is volatile and has a high octanol-water partitioning coefficient. Because of these properties, lambda-cyhalothrin binds strongly to sediment or substrates containing high organic matter and has short environmental half-lives in water (Hand et al. 2001; Bouldin et al. 2006; Budd et al. 2009).
Lambda-cyhalothrin is a broad spectrum insecticide that is used to control a wide range of insects and mites (FAO 2015). It is used in agriculture and for public health protection (treatment against malaria vectors and mosquitos). Several articles (Bouldin et al. 2005; Bouldin et al. 2006; Budd et al. 2009; Budd et al. 2011) investigated the removal of lambda-cyhalothrin

in CW systems, while little information is found for imidacloprid in these systems. The main aim of this study is to investigate the removal/retention of these frequently used insecticides, by vegetated CWs mesocosms, planted with different plant monocultures. Exposure of mesocosms to different concentrations of pesticides and pesticide accumulation in sediment and plants was investigatedx

5.2 Materials and methods

5.2.1 Experimental design

Mesocosm experiments were performed at the Anton de Kom University of Suriname from June to September 2011. A total of 10 poly propylene tubs were used in this experiment [60 cm (L)x30 cm (W)x18 cm (H)]. Tubs were filled with a mixture of sandy-loam sediment (9 cm high), which originated from a drainage ditch in the research area, and potting soil (ratio 2:1). The sandy loam soil was first analyzed on the presence of pesticides and tested negative. The potting soil was purchased from the main Agrocenter in the research area and was applied to promote the growth of wetland plants (USDA 1998). Five mesocosms were planted with *Nymphaea amazonum*, 5 with *Eleocharis mutata* plants with 1 of each type of mesocosm used as a "control" (Figure 5-1). No pesticides were added to the controls, which were used to compare the growth of plants with and without pesticides. Plants were chosen because of their abundance in the research area and mature plants were harvested from a pond in the research area where no previous agricultural activities (use of pesticides) had taken place. Tapwater was added gradually until 7 cm above sediment to resemble ditch water levels in the field. The mesocosms were placed in a room (with a roof on top to exclude rainfall) of which the sidewalls were from wire mesh, enabling sunlight to reach them. For lambda-cyhalothrin, 4 batches were performed of 2 weeks each, with target concentrations of 10 (batches 1 and 2) and 30 µg/l (batches 3 and 4). For imidacloprid, 5 batches were performed with target concentrations of respectively 60 (batches 1 and 2), 180 (batches 3 and 4) and 1000 µg/l (batch 5). Because of storage problems with samples at time 336 h, batches 4 and 5 (imidacloprid mesocosms)) were executed for 9 days instead of 14 days. Batches 2 and 4 are a repetition of respectively batches 1 and 3. Batch 1 was executed in weeks 1 and 2 (a total of 2 weeks) and batch 2 in weeks 3 and 4, batch 3 in weeks 5 and 6, followed by a similar order for batches 4 and 5.

Batch 1, involving lamda-cyhalothrin was performed with *N. amazonum* mesocosms 1 and 2 (replicate of mesocosm 1) and *E. mutata* mesocosms 5 and 6 (replicate of mesocosm 5) exposed to a dose of 10 µg/l and executed during 2 weeks (week 1 and 2). Batch 2 was a repetition of batch 1 and executed 2 weeks later than batch 1, by exposing the same mesocoms (1, 2, 5 and 6) to the same dose (10 µg/l lambda-cyhalothrin) as was done in batch 1. The same methodology was followed for consecutive batches 3 (executed 2 weeks later than batch 2) and 4 (executed 2 weeks later than batch 3), with the difference that a higher dose of 30 µg/l was applied (see Figure 5-1, right).

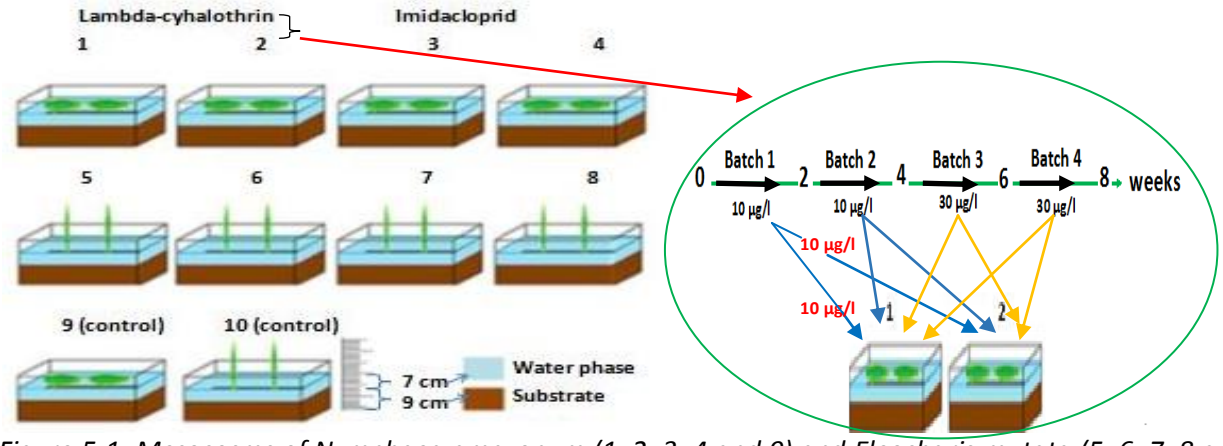

Figure 5-1: Mesocosms of Nymphaea amazonum (1, 2, 3, 4 and 9) and Eleocharis mutata (5, 6, 7, 8 and 10), see figure left. Note 1: Mesocosms 1, 2, 5 and 6 were exposed to lambda-cyhalothrin, and mesocosms 3, 4, 7 and 8 to imidacloprid. Note 2: mesocosm 2 is a replicate of 1, 4 a replicate of 3, 6 a replicate of 5 and 8 a replicate of 7. The figure to the right, gives a detailed explanation of how N. amazonum mesocosms (1 and 2) are exposed to lambda-cyhalothrin during the different batches over the total time-span of the experiment. The E. mutata mesocosms (5 and 6) were given the same treatment

General water quality measurements (pH, temperature, total dissolved solids (TDS)) were performed biweekly by means of hand-held field meters (Brand Extech). The field meter consisted of an electrode, which was placed directly in the water of the mesocosms, followed by reading of the scale. Evapotranspiration was adjusted by the addition of tap water, which was approximately 0.34 litres per day. The temperature range in the mesocosms was 26.1-28.3 ^{o}C.

The pesticide concentration was determined for the water phase at time 0, 4, 24, 48, 96, 216 and 336 h and in shoots and leaves (parts exposed to the water column), roots and sediment at the end of the experiment.

A two-way ANOVA (Sigma plot version 12) was used to determine significant differences (α=0.05) in the pesticide concentrations/removal in water within one specific type of mesocosm/plant and between the different mesocosms or plants in the different batches (see Appendix 4, statistical outputs for chapter 5). ANOVA was applied because all data fulfilled the assumptions for performing an ANOVA test; 1) data was normally distributed and 2) data fulfilled the assumption for homoscedasticity or equal variance. In the case of significant differences, which indicates that at least one group differs from the other groups, the Multiple Comparison Procedure (Fisher's Least Significant Difference Method) is used to specify, which of the groups differs significantly.

5.2.2 Pesticide applications

Plants were given a 5-weeks equilibrium period (Kadlec and Wallace 2009) to adapt to the environmental conditions in the mesocosms. Each applied pesticide dose was made by mixing the appropriate amount of technical grade pesticide with 1 litre of water from the mesocosms. This was done by gentle stirring in a beaker, followed by addition of the pesticide-water mixture to the mesocosm followed by mixing gently with a glass rod. The target concentrations of lambda-cyhalothrin (10 µg/l) and imidacloprid (60 µg/l) were based on recommended field applications of lambda-cyhalothrin at 0.05 kg a.i./ha (Karate 2.5 EC (Emulsifiable Concentrate) purchased at Agrimex N.V.) and imidacloprid at 0.3 kg a.i./ha (Imidox 20% SL (soluble concentrate) purchased at H.J. De Vries Agro). The obtained mixture was immediately applied to the water layer in the mesocosms to simulate a 10 mm precipitation event from a 0.05 ha (typical size of field area) with 50% of rainfall ending up in runoff, and containing 1% of the applied amount of pesticide. The physico-chemical properties and fate characteristics of imidacloprid and lambda-cyhalothrin are presented in Table 5-1.

Table 5-1: Physico-chemical properties and fate characteristics of imidacloprid and lambda-cyhalothrin (Footprint IUPAC-PPDB database 2015)

Physico-chemical property/ Fate characteristic	**Imidacloprid**	**Lambda-cyhalothrin**
Water solubility (mg/l, 20 °C)	610	0.005
log K_{ow}	0.57	6.9
Water-Sediment DT_{50} (days)	129	12
Aqueous photolysis DT50 (days) at pH 7	0.2	40
Aerobic sediment, field DT_{50} (days)	174	26.9
Henry´s Constant (Pa m^3 mol^{-1})	$1.7x10^{-10}$	$2.00x10^{-2}$
Vapour pressure at 20 °C (mPa)	$4x10^{-7}$	0.0002
Freunlich sorption coefficient K_f	2.23	2144

5.2.3 Pesticide analysis

Lambda-cyhalothrin was analyzed in water, plants and sediment according to a procedure described by Bennett et al. (2000). All plants were removed from the mesocosms and different plant species were analyzed seperately. To determine the growth of plants, plants were dried with tissue paper and wet biomass was determined in a similar manner as before the start of the experiment. The biomass (wet weight) of all *N. amazonum* and *E. mutata* plants was at the start of the experiment 233.43 against 125.00 grams and at the end of the experiment 228.22 against 408.97 grams respectively. At the end of the experiment, the plants were harvested and gently washed to remove excessive sand especially near the roots. After drying with paper tissue, the mass was determined gravimetrically by means of an analytical balance (Mettler). It was observed that larger leaves of *N. amazonum* would fall off, and that new ones would develop.

After this step, plants were divided in shoots and leaves, and roots, while for sediment, the top 3 cm layer was removed from each mesocosm and well mixed, followed by a gravimetrical determination of the total mass and the moisture content (Moore et al. 2000). For analysis, around two times 10 g (shoots and leaves, roots) and two times 25 g (sediment) were further extracted with a suitable solvent.
For plants, the sonication method was used and for sediment a 6 hour Soxhlet extraction, with both methods involving the use of ethyl acetate as solvent. Water samples were extracted by means of a liquid-liquid extraction with ethyl acetate as solvent. The obtained extract was dried over sodium sulphate and evaporated until near dryness and re-dissolved in hexane, prior to injection into the GC (Gas Chromatograph).

Quantification was done by means of a GC-MS (Gas Chromatography Mass Spectrometry) 6890N (Agilent technologies) equipped with a HP-5ms column. The oven temperature program was as follows: 70 °C held for 2 min to 150 °C at a rate of 25 °C/min, held for 5.20 min to 200 °C at a rate of 3 °C/min, held for 21.87 min; and then to 280 °C at 8 °C/min, held for 7 min. The injector temperature was 280 °C. The carrier gas helium was set to a constant flow of 55 ml/min. The limit of detection (LOD) for lambda-cyhalothrin in water, sediment and plants was 0.001 µg/l, 0.01 µg/kg and 0.01 µg/kg respectively. Mean recoveries for lambda-cyhalothrin in water (liquid-liquid extraction), sediment (Soxhlet procedure) and shoots and leaves (sonication method) were 96.7 ± 0.4% (n=3), 87.2 ± 15.9% (n=3) and 90.0 ± 7.1% (n=3) respectively.

For the residue analysis of imidacloprid, water samples were filtered through a syringe filter with a PVDF (Poly Vinylidene Difluoride) membrane (pore size of 0.22 µm, Carl Roth, Karlsruke-Rheinhafen, Germany). Sediment and plant samples were analyzed by a method described by Baskaran et al. (1999). This method involved an extraction with a mixture of acetonitrile and water (80 + 20 by volume; 40 ml) followed by centrifugation and filtration of the supernatant through a PVDF membrane.

Imidacloprid was quantified by means of HPLC (High Performance Liquid Chromatography) (Thermo Surveyor) equipped with a surveyor LC pump plus, a Surveyor PDA plus 5 detector (Thermofisher) and a platinum C18ESP 3 µm column. The LOD for imidacloprid in sediment and plants was 0.5 mg/kg and in water, 0.5 µg/l. The mean recoveries (average of 3 measurements which were conducted simultaneously, n=3) for imidacloprid in water, sediment and shoots and leaves were 87.0 ± 4.4%, 92.7 ± 26.6% and 91.3 ± 8.1 respectively.

5.2.4 Calculation of the removal efficiency

The removal efficiency of pesticides from the water phase is calculated according to Formula 5-1.

$$Removal\ efficiency = \frac{C_0 - C_x}{C_0}\ x\ 100\% \qquad \textit{(Formula 5-1)}$$

In which:

C_0: concentration of pesticide at time 0 hours (µg/l)
C_x: concentration of pesticide at time x hours (µg/l)

5.2.5 Calculation of DT_{50} in the water phase

The dissipation of pesticides from the water phase of wetland mesocosms is calculated by means of Formula 5-2. This is based on a first order reaction rate model (Kadlec and Wallace 2009). In this model, k equals the reaction rate constant or removal coefficient and is calculated by means of a linear regression using Formula 5-3:

$$DT_{50} = \frac{ln2}{k} \qquad \textit{(Formula 5-2)}$$

$$ln[A] = -kt + ln\,[A]_0 \qquad \textit{(Formula 5-3)}$$

In which:

[A]: concentration of lambda-cyhalothrin/imidacloprid at time (t) = x h (µg/l)
$[A]_0$: concentration of lambda-cyhalothrin/imidacloprid at time (t) = 0 h (µg/l)
k: removal coefficient or the slope obtained from linear regression (h^{-1}).
See Appendix 3 (Figures a up to t) for additional information on the regression analysis.

The distribution of DT_{50} values was evaluated by means of a box plot. Outliers were identified by means of the inter quartile range (IQR), which is the difference between the 3rd quartile and the 1st quartile. For each dataset, border values were found by means of respectively: 3rd quartile + 1.5 IQR (upper limit) and 1st quartile - 1.5 IQR. (lower limit). All values out of this range were noted as outliers. The output is presented in Appendix 6, Tables f-2 and f3).

Statistical analysis (t-test) was used to find significant differences between the DT_{50} values of pesticides in the different mesocosms. A two-way ANOVA (Sigma plot version 12) was used to see whether or not significant differences (α=0.05) exist in the mean DT_{50} values among the different levels of 'type of plant', and the different levels of 'target concentrations'. In the case of significant differences, which indicates that at least one group differs from the other groups, the Multiple Comparison Procedure (Tukey test) is used to specify, which of the groups differs significantly

5.3 Results

5.3.1 Water quality parameters

The measured values in Table 5-2 show that the pH, salinity (S), conductivity (EC_{cond}) and Total Dissolved Solids (TDS) slightly increased in all mesocosms (including the controls) in the period of two months. The measurements were done at temperatures between 26.1-28.3 ^{o}C.

Table 5-2: Measured values (n=16[a]) for water quality parameters in mesocosms with pesticides and without pesticides (controls) at the start and at the end of the experiment

Mesocosms	pH	Salinity (ppt)	EC_{cond} (mS)	TDS (g/l)
1 up to 8	4.46[b]/6.18[c]	1.14/1.56	2.30/3.13	1.61/2.18
9 and 10 (controls)	4.38[c]/5.33[d]	1.11/1.56	2.23/3.13	1.56/2.16

[a] *Values represents the range of 16 measurements*
[b] *The first value represent the average (n=8; values for 8 mesocosms) at the start of the experiment*
[c] *The second value represents the average (n=8) at the end of the experiment*
[c] *The first value represent the average (n=2; values for 2 mesocosms) at the start of the experiment*
[d] *The second value represents the average (n=2) at the end of the experiment*

5.3.2 Dissipation of lambda-cyhalothrin from the water phase

Results for the dissipation of lambda-cyhalothrin from the water phase in the different mesocosms are presented in Figure 5-2.

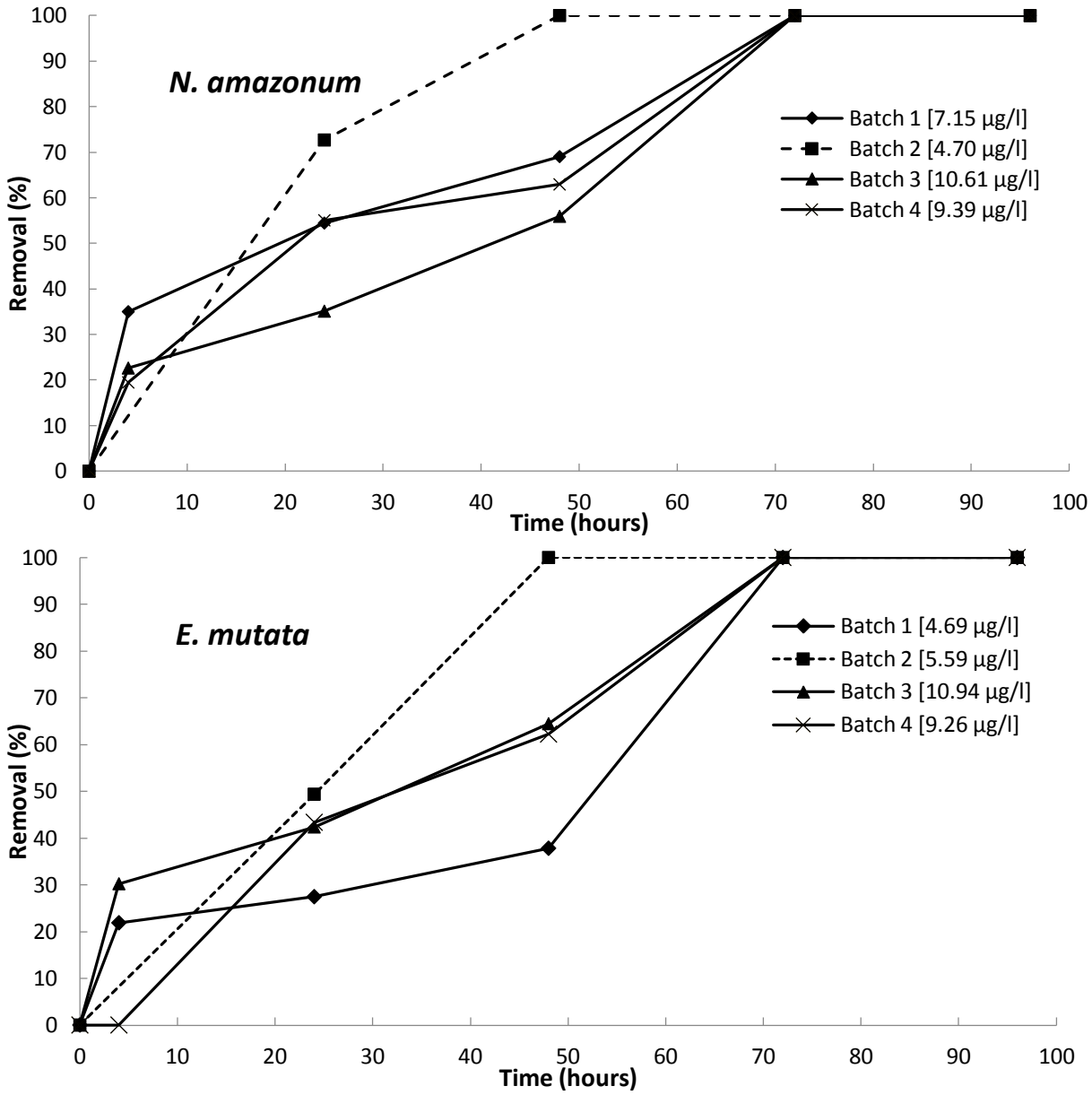

Figure 5-2: Removal (%) of lambda-cyhalothrin from the water phase of Nymphaea amazonum and Eleocharis mutata mesocosms.The Nymphaea amazonum mesocosm were exposed to pesticide target concentrations of 10 µg/l (batches 1 and 2) and 30 µg/l (batches 3 and 4) for a duration of 2 weeks. The actual starting concentrations are between parenthesis in the legend. Removal (%) at each time point, was based on the average of 2 values

Results for batch 1 show that at time 72 h, concentrations of lambda-cyhalothrin in the water phase of all *N. amazonum* mesocosms were below the limit of detection (0.001 µg/l), and 100% dissipation was reached.

For batch 2, dissipation of the pesticides was already reached at time 48 h. For both batches 3 and 4, a 100% removal was reached at time 72 h, however for batch 4 (repetition of batch 3), a

higher removal was observed at time 24 h (55.0%) and 48 h (62.9%) compared to batch 3 (35.1% at time 24 h and 55.9% at time 48 h respectively).
For the *E. mutata* mesocosms, results for batch 1 show that at time 72 h, concentrations of lambda-cyhalothrin in the water phase of all mesocosms (5 and 6) were below the limit of detection (0.001 µg/l), and a 100% dissipation was reached. Results for batch 2 show that a 100% dissipation is reached at 48 h instead of 72 h. When batch 4 is compared with batch 3, this trend is not observed and comparable results (higher removal efficiencies) are found for the time interval 24-48 h. Similar to batch 1, a 100% dissipation is reached at time 72 h.
Statistical results obtained for a two way ANOVA test, with the concentration as dependable variable and factors time and batches (different target concentrations), showed that for *Nymphaea amazonum* mesocosms significant differences exists between batches 1 and 2 (10 µg/l target concentration) compared to batch 3 (30 µg/l target concentration). Significant differences also exists between batches 3 and 4, while for the *Eleocharis mutata* mesocosms these differences were found between batches 2 and 3. For lambda-cyhalothrin a 100% removal is obtained from the water phase, between 48-72 h, which is independent of the dose applied (10 and 30 µg/l) and type of plant (*N. amazonum* and *E.mutata*).

5.3.3 $DT_{50\ wetland}$ of lambda-cyhalothrin

The DT_{50} of lambda-cyhalothrin (see Table 5-3) was calculated by making use of Formula 5-2 and 5-3.

Table 5-3: Average[a] half-life time values, the median and acceptable DT_{50} range for lambda-cyhalothrin in the water-sediment phase of wetland mesocosms. Values in red are outliers

lambda-cyhalothrin (target concentration)	***N. amazonum* mesocosms**			***E. mutata* mesocosms**		
	k $(h)^{-1}$	**R^2**	**DT_{50} (h)**	**k $(h)^{-1}$**	**R^2**	**DT_{50} (h)**
Batch 1 (10 µg/l)	0.042	0.9067	16.50	0.019	0.9149	36.48
	0.018	0.8730	38.51	0.014	0.9536	N.D.[b]
Batch 2 (10 µg/l)	0.031	0.9912	22.36	0.034	0.9778	20.39
	0.066	0.9993	10.50	0.036	0.9906	19.25
Batch 3 (30 µg/l)	0.028	0.8704	24.76	0.027	0.8314	25.67
	0.030	0.8687	23.10	0.031	0.9661	22.36
Batch 4 (30 µg/l)	0.032	0.9934	21.66	0.022	0.9692	18.24
	0.035	0.9775	19.80	0.038	0.9886	18.24
DT_{50} ± SD (h), n=4, Batches 1 and 2	22.0 ± 12.0			25.4 ± 9.6		
DT_{50} ± SD (h), n=4, Batches 3 and 4	22.3 ± 2.1			24.4 ± 5.6		
Average DT_{50} (h) ± SD (h)	**22.2 ± 7.5**			**23.0 ± 6.5**		
Median (h)	**22.1**			**20.4**		
Acceptable[c] DT_{50} range (h)	**12.00-30.35**			**10.84-31.92**		

[a]For N. amazonum mesocosms (Batches 1 and 2), the average was based on 4 values obtained for mesocosms exposed to 10 µg/l lambda-cyhalothirn, while the overal average was based on respectively 8 and 7 values for respectively N. amazonum and E. mutata
[b]Not determined
[c]DT_{50} range without the outliers

The average DT_{50} of lambda-cyhalothrin was 23.0 h for *Nymphaea amazonum* mesocosms and 22.2 h for *Eleocharis mutata* mesocosms, with R^2 values ranging between 0.8687-0.9993 and 0.8314-0.9906 respectively (88% of all R^2 values were higher than 0.9).

The average DT_{50} does not differ much from the median values for both types of mesocosms, which indicates a normal distribution, which was also confirmed by statistical tests (Appendix 4, statistical outputs for chapter 5).

Comparison of the DT_{50} values shows that between the 2 types of mesocosms, the differences are not statistically significant ($p = 0.583$), which is expected because the range for the acceptable half-life times of lambda-cyhalothrin in both types of mesocosms is equal and between 0.5-1.3 days.

5.3.4 Dissipation of imidacloprid from the water phase

Results obtained for the removal of imidacloprid from the water phase in different mesocosms are presented in Figure 5-3.
The highest removal efficiency (100%) was observed for the *Eleocharis mutata* mesocosms during batch 1 (target concentration 60 µg/l) at time 336 h. For the consecutive batches, with the exception of batch 5 (highest target concentration), lower removal efficiencies were obtained. Statistical two-way ANOVA analysis revealed that contrary to the lambda-cyhalothrin experiments, the different target concentrations played a significant role in the removal of imidacloprid from the water phase of the mesocosms. The removal was dependent ($p=0.048$ at the $\alpha= 0.05$ level) of the type of plant for the highest applied concentrations (1000 µg/l). Results showed that for the highest target concentration (1000 µg/l) at the time interval 24 h-72 h, higher removal efficiencies were obtained for the Nymphaea amazonum mesocosms compared to the Eleocharis mutata mesocosms.

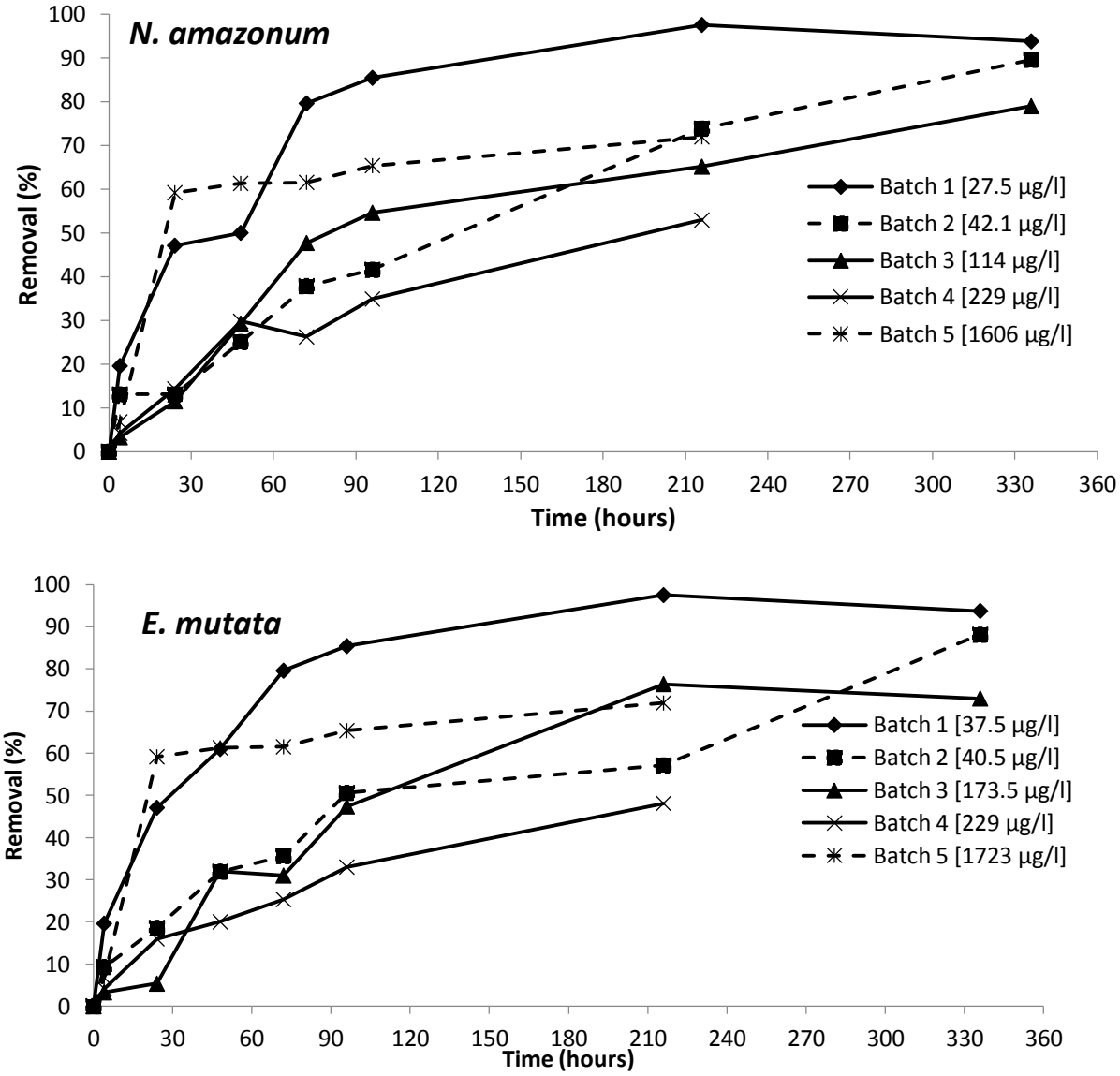

Figure 5-3: Removal (%) of imidacloprid from the water phase of Nymphaea amazonum and Eleocharis mutata mesocosms. The Nymphaea mesocosm no. 3 and its replicate, no. 4, (see Figure 5-1) were exposed simultaneously to the same pesticide target concentrations of 60 µg/l (batches 1 and 2), 180 µg/l (batches 3 and 4) and 1000 µg/l (batch 5) for a duration of two weeks for each batch (batch 5 was conducted for 9 days). The actual starting concentrations are between parenthesis in the legend. Removal (%) at each time interval, was based on the average of 2 values (mesocosm 3 and 4), while for Eleocharis mutata that was based on the results obtained for mesocosms 7 and 8

5.3.5 $DT_{50\ wetland}$ of imidacloprid

The results obtained for the half-life of imidacloprid in the water phase of the two types of wetland mesocosms are presented in Table 5-4.

Table 5-4: Average[a] half-life time values, the median and acceptable[c] DT_{50} range for imidacloprid in the water phase of wetland mesocosms. Values in red are outliers

Imidacloprid (target concentration)	***N. amazonum* mesocosms (**			***E. mutata* mesocosms**		
	k $(h)^{-1}$	**R^2**	**DT_{50} (h)**	**k $(h)^{-1}$**	**R^2**	**DT_{50} (h)**
Batch 1 (60 µg/l)	0.013	0.9836	53.3	0.013	0.9348	53.3
	0.028	0.9092	24.7	0.026	0.831	26.7
Batch 2 (60 µg/l)	0.005	0.8978	139	0.009	0.9583	77.0
	0.010	0.9115	69.3	0.007	0.9206	99.0
Batch 3 (180 µg/l)	0.005	0.8803	139	0.005	0.7016	139
	0.005	0.8727	139	0.005	0.9478	139
Batch 4 (180 µg/l)	0.003	0.848	231	0.003	0.9362	231
	0.004	0.9574	173	0.003	0.9207	231
Batch 5[d] (1000 µg/l)	0.005	0.5466	139	0.005	0.6238	139
	0.005	0.614	139	0.006	0.6493	116
DT_{50} ± SD (h), n=4, Batches 1 and 2	71.5 ± 48.4			64.0 ± 31.1		
DT_{50} ± SD (h), n=4, Batches 3 and 4	170.4 ± 43.6			184.8 ± 53.4		
DT_{50}[b], n=2, Batch 5	139			127		
Average[a] DT_{50} (h) ± SD (h)	**125.0 ± 60.4**			**125.1 ± 67.4**		
Median (h)	**139**			**127.5**		
Acceptable[c] DT_{50} range (h)	**8.3-217**			**0-224**		

[a] *Average was based on 10 values or the values obtained for all batches*
[b] *Batch 5 was conducted once and the average half-life time values were based on 2 values. No standard deviation (SD) was therefore calculated*
[c] *DT_{50} range without the outliers*

Lower DT_{50} values are observed in mesocosms exposed to lower target concentrations (batches 1 and 2) compared with those exposed to higher target concentrations (e.g. batches 3 and 4). Significant differences were found in imidacloprid mean half-life values for mesocosms exposed to dose 1 (60 µg/l) compared to mesocosms exposed to dose 2 (180 µg/l) and dose 3 (1000 µg/l), with p-values of respectively 0.004 and 0.016 (see Appendix 4, statistical outputs for chapter 5). In *Nymphaea amazonum* mesocosms, the DT_{50} varied from 24.8-139 h (1-6 days) for the 60 µg/l, 139-231 h (6-9 days) for the 180 µg/l and it was 139 h (6 days) for the 1000 µg/l, while for the *Eleocharis mutata* mesocosms this was 1-4 days, 6-9 days and 5-6 days respectively. The correlation coefficient, R^2, varied from 0.831 up to 0.984 for the batches 1 and 2 (regression coefficient R^2 values were higher than 0.9, 75% of the time), while for batches 3 and 4, R^2 values varied from 0.702-0.957 with values higher than 0.9, 50% of the time. Low correlation coefficients (R^2 values between 0.547-0.649) were obtained for DT_{50} values of batch 5 (dose 1000 µg/l). It is expected that with exclusion of the outliers, the regression coefficient (R^2) will increase.

To evaluate the distribution of the half-life time values for both pesticides in the different types of mesocosms, box plots (Figure 5-4). were constructed with data provided in Appendix 6 (Tables f-2 and f-4).

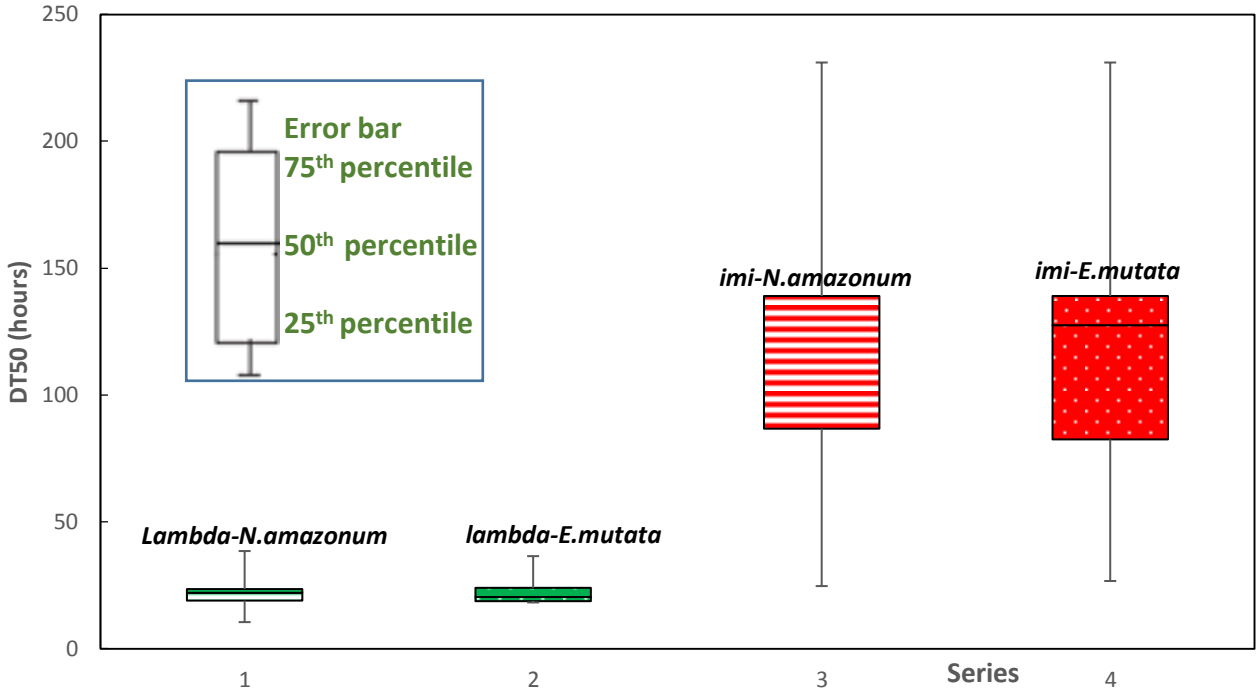

Figure 5-4: Boxplots showing the distribution of half-life time values obtained during a total of 4 and 5 batches (Table 5.3 and Table 5.4) for respectively, lambda-cyhalothrin and imidacloprid in the waterphase of N. amazonum and E. mutata mesocosms. The results are given over 4 series e.g. series 1, distribution of average half-life time values of lambda-cyhalothrin in the waterphase of N. amazonum mesocosms; series 3, distribution of average half-life time values of imidacloprid in the waterphase of N. amazonum mesocosms, etcetera. Note: for the imi-N.amazonum distribution, the median is equal to the 3th quartile or 75th percentile value

Comparable results between the average half-life time values of lambda-cyhalothrin and imidacloprid in both types of mesocosms were found. Comparison of the median values however showed small differences in distribution of the DT_{50} values. The average half-life times of lambda-cyhalothrin (22.2 h (n=8) against 23.0 h (n=7) for respectively *N. amazonum* and *E. mutata* mesocosms) are slightly higher (positive skewness) than the median values of 22.1 h and 20.4 h. The positive skewness can be clearly seen from the much larger whisker above of the box, which is much larger for the *E. mutata* mesocosms, because of the greater difference. The smaller negative whisker (series 2) is caused by the much smaller difference of 0.505 between the 1st quartile value and the lowest or minimum DT_{50} value, compared to that of series 1 (lambda-*N. amazonum*), for which this difference was 8.475 (see Appendix 6, Table f-2). The median half-life time for imidacloprid in the water phase of *Nymphaea amazonum* mesocosms was 139 h (6 days) and 127.5 h (5 days) for *Eleocharis mutata* mesocosms, while equal average values were found of 125 h for both types of mesocosms. Statistical analysis however did not show significant differences between the DT_{50} values of lambda-cyhalothrin ($p = 0.583$) obtained for *N. amazonum* and *E. mutata* mesocosms and the same was observed for the DT_{50} values of imidacloprid ($p = 0.987$). The median half-life times calculated for lambda-cyhalothrin and imidacloprid, are in agreement with the time which is needed for 50% removal

from the water phase (Figure 5-2 and 5-3), with the exception of the results for imidacloprid (batch 5). The half-life time, which can be estimated from Figure 5-3, is much lower (around 1 day) for both types of mesocosms compared to the calculated DT_{50} which was around 5-6 days. This 5-6 days period was based on linear regression analysis, for which a low correlation coefficient was found and which might explain the difference in the half-life time values.

5.3.6 Pesticides detected in sediment, plant material and roots

The amount of pesticides detected in the different compartments of the wetland mesocosms is presented in Table 5-5. For *Nymphaea amazonum* mesocosms, 48.5% (1.65 mg) of the applied dose of lambda-cyhalothrin was detected in the sediment, 0.4% in shoots and leaves, while the amount in roots was below the limit of detection (0.01 µg/kg). For *Eleocharis mutata* mesocosms, 44.6% of lambda-cyhalothrin was detected in sediment and 0.5% in roots. Contrary to the *Nymphaea amazonum* mesocosms, the amount in shoots and leaves was below the limit of detection. Results showed that the main pathway of lambda-cyhalothrin in the mesocosms was adherence to sediment particles. Comparing the two types of mesocosms and especially looking at the total amount of pesticide (µg/g), more lambda-cyhalothrin was found in the *Nymphaea amazonum* plants (shoots and leaves), while the amount of lambda-cyhalothrin in sediment for both plant mesocosms was approximately the same.

Table 5-5: Amount of pesticides detected in different parts (sediment, shoots and leaves, and roots) of wetland mesocosms. The results expressed in percentages are presented in green

Total mass added (mg)		**λ-cyhalothrin 1.65**			**Imidacloprid 28.1**		
Mesocosm	**Medium**	**µg**	**% of total**	**µg/g**	**µg**	**% of total**	**µg/g DW[b]**
N.amazonum	Sediment	797.2	**48.5**	0.28	4866	**17.3**	1.7
	Plant upper part (shoots and leaves)	6.68	**0.4**	2.02	18083	**64.3**	2085
	Roots	ND[a]			4092	**14.6**	
	Total in plant	6.68	**0.4**	2.02	22175	**78.9**	2085
E. mutata	**Sediment**	733.3	**44.6**	0.27	4324	**15.4**	1.6
	Plant upper part (shoots and leaves)	ND			131		
	Roots	8.60	**0.50**	0.51	17.1	**0.5**	13.5
	Total in plant	8.60	**0.50**	0.51	147	**0.5**	13.5

[a] *Not Detected*
[b] *Dry Weight*

For *Nymphaea amazonum* mesocosms, 78.9% (28.1 mg) of the applied amount of imidacloprid was retained in plants (shoots and leaves, and roots) and 17.3% in sediment, while for *Eleocharis mutata* mesocosms, only 0.5% was detected in shoots and leaves, and roots and 15.4% in sediment.

The amount of imidacloprid detected in the plants of *Nymphaea amazonum* mesocosms was over 3320 times higher than the amount of lambda-cyhalothrin in the same type of mesocosms. This indicates a higher accumulation capacity towards imidacloprid. On the other hand, a higher dose of imidacloprid (17 times more) was added and available for the shoots and leaves, growing in the mesocosms. Compared to lambda-cyhalothrin, imidacloprid is also more water-soluble. Although a very high concentration of imidacloprid (1000 µg/l) was used in the last batch (no. 5), no negative effects were observed for the plants in the control mesocosms.

5.4 Discussion

The metabolic fate of pesticides is dependent on several factors such as abiotic environmental conditions (pH, moisture, sediment and temperature), microbial community, plant species, pesticide characteristics and biological and chemical reactions (Van Eerd et al. 2003; Yuan et al. 2007). The increased values for the EC_{cond}, S and TDS mentioned in section 5.3.1, may be caused by evapotranspiration. The measurements were conducted before adjusting the water level. The pH increase in constructed wetlands can be the result of algal photosynthesis or photosynthesis by submerged macrophytes (Vymazal 2007; Moore et al. 2013; Brogan et el. 2014).

The fate of lamda-cyhalothrin over the different batch experiments especially in repeated batches (2 and 4) was characterized by high dissipation from the water phase, especially during the time interval 0-48 hours. This can be caused by an enhanced biodegradation due to the repeated exposure with lambda-cyhalothrin. It is possible that repetitive exposure to the same pesticides over time may have caused induction and adaptation of microbes, leading to establishment of organisms capable to rapidly degrade the pesticides in the wetland (Kadlec and Wallace 2009; O'Geen et al. 2010). Occurrence of enhanced biodegradation is mentioned in other wetland studies (Weaver et al. 2004; Rose et al. 2006; Budd et al. 2011). This enhanced biodegradation must however be confirmed by means of e.g. respiration tests. Especially because lambda-cyhalothrin, a hydrophobic pesticide with a high log K_{ow} is rapidly transferred to the sediment phase of the wetland, were it might not be readily available to microorganisms. The ability of microorganisms to degrade pesticides is also influenced by the presence of dense vegetation in mesocosms. Dense vegetation can diminish pesticide toxicity towards aquatic species and microorganisms (Brogan and Relyea 2013). In that study, the toxicity of malathion was mitigated through the plants effects on the water quality (increasing pH), causing rapid hydrolysis and detoxification in water. In Moore et al. (2013) the effect of plant density is given as a possible reason for the lower amount of lambda-cyhalothrin found in sediment. The higher the plant density, the less lambda-cyhalothrin is found in sediment. In current study, plant density is expected to increase, because of plants growing in the mesocosms with time. However, when the biomass (wet weight) of all *N. amazonum* and *E. mutata* plants are compared at the start and at the end of the experiment, only the biomass of *E. mutata* plants was increased significantly, while a small decrease in biomass is observed for *N. amazonum* plants (section 5.2.3), because large leaves would die-off. Because this was also observed in the

control, it is not likely caused by the addition of pesticide. The somewhat lower amounts of lambda-cyhalothrin found in sediment of the *E. mutata* mesocosms, might be caused by the higher amount of biomass.

The results for the accumulation of lambda-cyhalothrin were 0.4% in *Nymphaea amazonum* (shoots and leaves) and 0.5% in *Eleocharis mutata* mesocosms (uptake by roots alone). In Bouldin et al. (2006), a much higher percentage (72.1%) of lambda-cyhalothrin was adsorbed to the roots of *Juncus effusus* within 8 h of exposure. After 8 days, 25.4% was translocated to the upper plant tissue, while for *Ludwigia peploides* 85.5% partitioned in the root biomass. Also 0.634 µg lambda-cyhalothrin/g of DW of *Juncus effusus* and 1.913 µg/g DW of *Ludwigia peploides* was found, while in present study the amount of lambda-cyhalothrin was 2.02 µg/g DW for *Nymphaea amazonum* and 0.51 µg/g DW for *E. mutata*. The experiment in Bouldin et al. (2006) was in contrary to the current study (56 days) a short-term exposure of 8 days. The occurrence of pesticides in different parts of the plant indicates different mechanisms of metabolism, which are still unknown (Crowly et al. 1997; Mc Kinlay and Kasperek 1999).

The DT_{50} determines the pesticide persistence in the environment. A value higher than 100 days refers to a persistent pesticide (Poissant et al. 2008). In this experiment, the DT_{50} of lambda-cyhalothrin in the water phase was on average 1 day for both types of mesocosms. This is in agreement with the results obtained by Hand et al. (2001), who conducted degradation and sorption studies with aquatic plants using 1 g wet weight of plant and a pesticide dose of 5 µg/l. In that study, half-life times of other pyrethroids were noted with low DT_{50} values (< 1 day) ranging from 1-4 h for deltamethrin, less than 6 h for permethrin, 10 h for esfenvalerate and less than 24 h for cypermethrin. Higher DT_{50} values (12 days) were found in the IUPAC-PPDB Footprint database for water-sediment systems, which might be related to lower temperatures and different pH values. According to Moore et al. (2013), relatively high temperatures (at or above 20 ^{0}C) and slightly acidic environments as in present study, will result in higher degradation rates and much lower DT_{50} values. According to Starner (1999) a high variability in pesticide half-lives for one specific pesticide is related to the influence of parameters such as microbial populations and water chemistry (e.g. use of filtered versus unfiltered water, the presence of dissolved organic carbon (DOC) and the presence of metal ions as catalysts).

The main pathway for lambda-cyhalothrin (a pyrethroid) in this study was through sedimentation of particles containing the pesticide and was observed for both *N. amazonum* and *E. mutata* mesocosms. This was also the case in Hand et al. (2001), were 70% of the applied lambda-cyhalothrin was found in the sediment phase of a sediment-water system, 1 day after pesticide application. In Moore et al. (2009B), the ability to remediate the pyrethroid permethrin by four macrophytes was evaluated in mesocosms amended with a target concentration of 5 µg/l and a pesticide retention time (PRT) of 4 h. The accumulation in plants after 12 h, ranged from 9.09 ± 2.08 µg up to 746 ± 214 µg for cis-permethrin and 4.14 ± 0.69 µg up to 398 ± 140 µg for *trans*-permethrin. The sorption to the sediment (10 g DW) ranged from 200 ± 4.52 µg up to 249 ± 53.6 µg for cis-permethrin and from 60.8 ± 5.4 µg up to 81.4 ± 29.4 µg for *trans*-permethrin. Only for cis-permethrin statistically significant differences (α = 0.05) in

plant-uptake were observed, while in current study uptake was independent of the dose applied and the type of plant. Similar results were reported in Moore et al. (2008) for a V-shaped vegetated ditch exposed to 20 µg/l permethrin, in which after an 8 hours experiment, 50% of both cis- and trans-permethrin masses were detected in sediment.

Less than 50% from the amount of lambda-cyhalothrin added could be quantified in sediment and plants. It might be possible that part of the lambda-cyhalothrin was (bio) transformed into other compounds (CO_2, H_2O, metabolites) or evaporated out of the mesocosms. According to the PPDB (2016), 3-phenoxybenzoic acid, and (Z)-3-(2-chloro-3,3,3-trifluoro-propenyl)-2,2-dimethylcyclo-propane carboxylic acid, are the relevant metabolites formed in soil. From PPDB (2016) and Table 5.1, it can be derived that lambda-cyhalothrin is volatile, but stable towards aqueous photolysis at pH 7. Another possibility is adherence of pesticides to the tubs. Although data in the literature could not be found on the amounts lost through this pathway, this might be possible (based on the log K_{ow} value en the hydrofobicity of this pesticide).

For the water-soluble imidacloprid, no comparable data was found for similar mesocosm studies. According to Starner and Goh (2012), no peer-reviewed literature on imidacloprid detections in agricultural areas of the United States is available. Contrary to lambda-cyhalothrin, lower removal efficiencies were obtained for repeated batches 2 and 4, which may be related to the high aqueous concentrations, which inhibited microbiological actions (Fogarty and Tuovinen 1991). For the highest target concentration (1000 µg/l) however, removal efficiencies increased during the start of the experiment up till time 224 h, with a higher removal compared to batches 2, 3 and 4. The increase in removal efficiency might indicate a possible enhanced biodegradation towards this pesticide or toxicity reduction caused by macrophytes (Brogan and Relyea 2013; Moore et al. 2013). The high amount of imidacloprid was dosed after a period of 8 weeks, during which period, the mesocosms were already exposed to increasing imidacloprid concentrations (batches 1 up to 4). Compared to batch1 however starting from time 72 h, a higher removal was observed for batch 1 (lowest target concentration). Statistical tests also show significant differences between the DT_{50} of batch 5 and batch 1, indicating that for Batch 5, the half-life is significantly higher. The latter contradict the previous statement of an enhanced biodegradation, which would be characterised by lower DT_{50} values for batch 5.

The uptake of imidacloprid by plants and sediment was also different compared to that for lambda-cyhalothrin. For *Nymphaea amazonum* mesocosms, 78.9% of the added amount of imidacloprid was present in plants and 17.3% in sediment. For *Eleocharis mutata,* only 0.53% was found in plants and 15.38% sediment, which indicates comparable amounts in sediment.

Because imidacloprid is non-volatile (PPDB 2016), the major pathway for this pesticide, in the *Nymphaea amazonum* mesocosms was uptake by plants and sediment particles (96.2% in total), while for the *Eleocharis mutata* mesocosms this value was only 15.9%. Losses of imidacloprid could be caused by photolysis or the fact that some amounts of pesticides remained in the water phase at the end of batch 5, or adsorption to the tubs. Imidacloprid's

adsorption however will be a minor process, compared to that of lambda-cyhalothrin, because of the low log K_{oc} and the good solubility of imidacloprid in water and the fact that this was not observed for the *N. amazonum* mesocosms (around 96% was traced back in plants and sediment) of which tubs consisted of the same type of material. According to the PPDB (2016), imidacloprid is very sensitive towards photolysis with an aqueous half-life at pH 7 of 0.2 days. The difference in the total amount of imidacloprid recovered between the two types of mesocosms, might be related to photolytic decay, degradation processes or formation of metabolites. Both types of mesocosms were not placed in direct sunlight, and it might therefore be possible that from the angle were the *E. mutata* mesocosms were placed, these mesocosms received more sunlight, resulting in a higher photolytic decay of imidacloprid. Because a comparable removal of imidacloprid was obtained from the water phase at the end of batch 5, the amount present in the water phase cannot be used to explain the difference in the amounts quantified and further research is therefore necessary to investigate the uptake by plants during more frequent time intervals and to measure the amount of metabolites formed. The effect of sunlight might be excluded by shielding the mesocosms from sunlight.

Although the amount of imidacloprid per gram of plant DW differs significantly for both plant species, the highest amount was present in the plant upper part (shoots and leaves). According to Sur and Stork (2003), translocation experiments to determine the mobility and distribution of imidacloprid in plants shows that there is a good acropetal (upwards) translocation to shoots and leaves and a poor basipetal (downwards) translocation to roots. This was also revealed in Wilson et al. (2000) and Alsayeda et al. (2008). In the study of Romeh (2010), imidacloprid was accumulated by broadleaf plantain plant (*Plantago major L*) present in water solution. The maximum levels were 15.7 µg/g of DW (in roots) and 37.21 µg/g of DW (in leaves). In present study, 2085 µg/g DW was found in *Nymphaea amazonum* plants, which indicates a very high accumulation potential towards imidacloprid.

The high water solubility and low sorption coefficient (K_{oc}) indicates that imidacloprid has a low tendency to adsorp to sediment particles (CDPR 2006). However, that can change and depends on several factors such as sediment type, pH, use of organic fertilizers and presence or absence of a ground cover. In nature, a ground cover is normally not present, while in the mesocosm used in present study this can be seen as plastic, and leaching is therefore excluded. In the study of Fernández-Bayo et al. (2007), sorption of imidacloprid was not directly linked to the sediment organic content, but was dependent on the type of sediment (high clay and organic carbon content) and soil-amendments (fertilizers, compost, shells) used.

The half-life time of imidacloprid in water was calculated to be around 5 days, which is higher than the 2 days found for the mean half-life in paddy water in the study of Phong et al. 2009. The photolytic half-life time is less than 3 h (CCME 2007). Longer half-life times found in present study (compared to the 2 days) are possible, because mesocosms were not placed in full sunlight. According to US EPA (2008), the photolytic half-life time ranges between 0-2 days (water) and 39 days (sediments). In the current study different (higher) half-life times were found for the different batches e.g. the higher the target concentration the higher the value for

DT_{50} (see Table 5-4), with the exception of batch 5 (very high target concentration of 1000 µg/l). Similar results were obtained by Kanrar et al. (2006) for the DT_{50} in paddy field water with values ranging from 1.6 days (application rate: 45 g a.i./ha), 1.9 days (60 g a.i./ha) and 2.8 days (90 g a.i./ha). According to Table 5-1 the water-sediment DT_{50} of 129 days is much higher than those found in current study. No further information was found under which conditions these experiments were conducted. In Moore et al. (2013) half-life times are related to water temperature and pH. Given the temperature (≥ 20) and slightly acidic-alkaline conditions (pH 5.7-8.7) in that study, it was estimated that the aqueous half-life of the insecticide diazinon is closer to 12 days instead of the 138 days mentioned in US EPA (2006). With higher temperatures and slightly acidic conditions in current study, this might also be the case. Recent research on imidacloprid degradation (Gupta et al. 2016) focuses on the degradation capacities by a new bacterial strain (*Pseudomonas sp.* RPT 52) isolated from an agricultural field. This strain is able to degrade 46.5% of imidacloprid (0.5 mM, exposure 40 h) in a sterile minimal medium according to first order kinetics. The strain uses imidacloprid as a carbon and energy source.

5.5 Conclusions

Repeated exposure of mesocosms with the same pesticide, showed higher removal efficiencies for pesticides from the water phase of most of the mesocosms and the possible influence of plant densities, which needs to be further investigated. The removal of lambda-cyhalothrin was independent of the dose applied and the plant type, while for imidacloprid, dissipation was dependent on the dose applied and the type of plant (for the highest target concentration). Lambda-cyhalothrin's dissipation was much faster from the water phase of both types of mesocosms compared to imidacloprid. A higher accumulation of the pesticides lambda-cyhalothrin and imidacloprid by *Nymphaea amazonum* plants over the *Eleocharis mutata* plants was observed and different plant uptake mechanisms took place.

Quantification of the amount of pesticides in harvested plants and in sediment from wetland mesocosms, showed that less than 50% of lambda-cyhalothrin is recovered. Imidacloprid was almost comletely recovered in *Nymphaea amazonum* mesocosms, while only a small amount (around 16%) was traced back in plants and sediment of *Eleocharis mutata* mesocosms. These differences which are possibly related to photolytic decay, (bio) degradation, sorption to the walls of the tubs and formation of metabolites must be further investigated.

Chapter 6 Fate of chlorothalonil, lambda-cyhalothrin and imidacloprid in wetland mesocosms

Abstract
Mesocosms used in wetland science allow for low cost experiments under controlled conditions. In the present study, the ability of wetlands planted with *Phragmites australis* to remediate pesticides (chlorothalonil, lambda-cyhalothrin and imidacloprid) in agricultural runoff in Suriname was investigated by comparing pesticide removal in vegetated and non-vegetated mesocosms. Mesocosms were exposed to chlorothalonil and lambda-cyhalothrin for a period of 2 weeks and to imidacloprid for a period of 10 days during 5 consecutive batches. Complete chlorothalonil dissipation from the water phase in batch 1 was observed after 97 h, while in batch 2 this was already the case after 48 h. With the exception of batch 4, chlorothalonil concentrations were below the limit of detection after 334 h. For both vegetated and non-vegetated mesocosms, similar results were obtained for lambda-cyhalothrin. However, batches 3 and 4 showed a decrease in lambda-cyhalothrin's removal rate, which was more visible in batch 4. Imidacloprid's dissipation increased gradually for all batches up to 100% in batch 5 for both types of mesocosms up to the end of the experiment. Statistical analysis showed no significant differences between the removal efficiency of the vegetated and non-vegetated mesocosms, for all three pesticides. The amount of pesticides detected in *P. australis*, was the highest for lambda-cyhalothrin, followed by imidacloprid and chlorothalonil. Results of plant-uptake were compared with the previous conducted mesocosm experiment to decide which plant to select, for future experiments. Calculated half-life times for pesticides in water were 0.66, 3.9 and 2.4 days for the vegetated mesocosms and 0.83, 2.9 and 2.8 days for the non-vegetated mesocosms, respectively for chlorothalonil, lambda-cyhalothrin and imidacloprid. Lambda-cyhalothrin was characterised by the highest DT_{50} value which was used as input data to calculate the surface area of a field scale constructed wetland capable to remove the three pesticides.

6.1 Introduction

Aquatic ecosystems can be impaired by agricultural pollutants such as pesticides and sediments and cause injury to humans and aquatic organisms. Impairment of macro-invertebrate community structures and ecosystem functions by the presence of pesticides in agricultural streams is reported in the Footprint IUPAC-PPBD database 2015. Chemicals such as atrazine and alachlor have been banned by several governments based on their risk assessments (Matamoros and Rodríguez 2016). CWs provide an effective and economical way to improve the quality of wastewater streams such as agricultural runoff, through biological, physical and chemical means.
Wetlands are used worldwide to purify effluent from agricultural and urban runoff (Moore et al. 2013; Tournebize et al. 2013; Vallée et al. 2015B; Vymazal and Březinová 2016A). Wetlands

are a recommended practice to solve the problem of agricultural diffuse pollution (Budd et al. 2011; Locke et al. 2011; Tournebize et al. 2013: Vallée et al.; 2015B). Their design is a function of the properties of the wastewater, the pollutant loading rate, the available surface and the climatic conditions of the wetland area (Kadlec and Wallace 2009; Hijosa-Valsero et al. 2010). Mesocosm studies are a simple approach to retrieve input data for the design of a field scale wetland. Mesocosms are small ecosystem experiments under replicated and controlled conditions and at low costs (Ahn and Mitsch 2002). They provide key parameters to design wetlands for a specific target pollutant, related to the water flow (Q), the chemical half-life time (DT_{50}) and the hydraulic retention time (HRT) (WRP 1994). Another important aspect is the selection of the appropriate type of vegetation needed for mitigation of pesticide associated runoff (Cooper et al. 2004; Bennett et al. 2005; Moore et al. 2011; Moore et al. 2013; Vallée et al. 2014). Plants provide the structural surface area occupied by the microbial flora that attack dissolved pollutants; they provide physical filtration for reduction of particulate matter, shade water surface and prevent algal re-growth and reduce temperature fluctuations (Beketov and Liess 2008; Kadlec and Wallace 2009). The vegetation used in the present study was *P. australis*, a perennial and flood-tolerant grass with an extensive rhizome system which usually penetrates to depths of about 0.6–1.0 m. Stems of these plants are rigid with hollow internodes with the range in shoot height from less than 0.5 m to 4–5 m (Vymazal 2013A). In terms of continents, *P. australis* is the most frequently used species in wetland systems in Europe, Asia and Central/South America (Vymazal 2013B). Vymazal (2013C) reviewed 643 SFCWs used for the treatment of several types of waste water in 43 countries, with a recorded plant species total of 150. From that study it was concluded that *Phragmites australis* was among the 5 most frequently used macrophytes genera (*Typha, Scirpus (Schoenoplectus), Phragmites, Juncus* and *Eleocharis*). However, no data is found on the ability of *P. australis* to mitigate runoff containing the pesticides chlorothalonil, lambda-cyhalothrin and imidacloprid in CW systems. These 3 pesticides belonged to the most frequently used pesticides in Commewijne, Suriname.
The first objective of present study is to investigate the dissipation and retention of the fungicide chlorothalonil and the two insecticides lambda-cyhalothin and imidacloprid in wetland mesocosms with and without the presence of *P. australis*. The second objective is to obtain input data for the design of a field scale constructed wetland capable to remove the three pesticides.

6.2 Materials and Methods

In this study, 3 frequently used pesticides (chlorothalonil, an organochlorine fungicide, lambda-cyhalothrin, a pyrethroid insecticide and imidacloprid, a neonicotinoid insecticide) were evaluated in wetland mesocosms planted with *P. australis.* The decision to make use of this plant was not only based on its frequent and worldwide use, but on the fact that results from previous experiments (e.g. chapter 5) were not yet available. The properties of the pesticides are presented in Table 6-1.

Table 6-1: Physico- chemical properties, environmental fate characteristics (Footprint IUPAC PPDB 2015) and field application rates of chlorothalonil, lambda-cyhalothrin and imidacloprid

Physico-chemical property/ Fate characteristics	**Chlorothalonil**	**Interpretation[h]**	**Cyhalothrin Lambda--**	**Interpretation[h]**	**Imidacloprid**	**Interpretation**
Water solubility (mg/l, 20 °C)[a]	0.81	L	0.005	L	610	H
log K_{ow}[b]	2.94	M	6.9	H	0.57	L
Water-Sediment DT_{50} (days)[c]	0.1	F	12	F	129	S
Aerobic sediment, field DT_{50} (days)[d]	44	M	25	L	174	H
Henry´s Constant (Pa m^3 mol^{-1})[e]	$2.5x10^{-2}$	L	$2.0x10^{-2}$	L	$1.7x10^{-10}$	L
Vapour pressure at 20 °C (mPa)[f]	0.076	L	0.0002	L	$4x10^{-7}$	L
Freunlich sorption coefficient K_f[g]	534	L	2144	N	2.23	M
Recommended field application rate (kg/ha)	**0.58-1.4**		**0.01-0.02**		**0.24-0.96**	

[a]< 50 mg/l: low; > 500 mg/l: high
[b]< 2.7: low bioaccumulation; 2.7-3: moderate; > 3.0 : high
[c]< 30 = fast; 100-365: slow degredation
[d]< 30: low or non-persistent; 30-100: moderately persistent; 100-365: high or persistent
[e]< 0.1: low or non-volatile; [f] < $1x10^{-4}$: non-volatile or low; > $1x10^{-4}$: high or volatile
[g]gives an interpratation on pesticide mobility, with
[h]Interpretations based on the Footprint (2015) website: low (L); moderate (M); high (H); fast (F); slow (S) and non-mobile (N)

6.2.1 Experimental design

The mesocosm experiments were performed at Ghent University, Belgium, in a greenhouse with an average temperature of 20.±.2 ^{0}C (n=59) and the relative humidity of 40.2.±.4.5 (n=59) in the period October 19, 2010 up to December 23, 2010 (66 days). The mesocosms were designed according to the SFCW systems. The factor rainfall and flow were excluded. For the study of chlorothalonil and lambda-cyhalothrin, 10 polypropylene tubs (0.56 m [L] x 0.38 m [W] x 0.41 m [D]) were used. For each pesticide, 2 vegetated and 2 non-vegetated tubs were used, as well as 2 controls (vegetated with no addition of pesticides) to distinguish plants evolution in mesocosms with and without exposure to pesticides. Each tub was filled with 15 cm soil-mix substrate; consisting of a volume ratio of 2:1 of respectively pot soil and soil (sandy loam). The soil had the following characteristics; pH (7.17); EC_{con} (120.6 µS/cm); OM (3.5%); OC (0.9%); Sand (56.5%); Loam (34.9%); Clay (8.7%) and C.E.C. (7.95). The vegetated ones were planted with 4 *Phragmites australis* plants. The tubs were gradually (1 cm/day) filled with water up to a level of 10 cm above soil (Figure 6-1) and equal to a free water volume of 21.3 liters. For each

pesticide a batch of 2 weeks was performed. A batch experiment consisted of the addition of pesticide stock solution to the water phase of wetland mesocosms to reach the desired target concentration, followed by sampling and analysing this water phase, temporally for a period of two weeks. After an adaptation period of 5 weeks, water from the mesocosms was amended with the pesticide stock solution to reach a target pesticide concentration (typically found in runoff) of 500 µg/l (see section 6.2.2). After gently stirring the obtained water mixture, composite samples were prepared by taking water (by means of a water bucket) from 4 different locations in the mesocosms and combining these 4 samples to obtain a representative water sample. Pesticide concentrations in water were measured at time 0 h, 2 h, 4 h, 22 h, 47 h, 72 h, 97 h and 334 h and were measured in shoots and leaves (plant upper part exposed to the water colum) and roots at the end of the experiment or after execution of all 5 batches. Prior to that, all plants were removed from the mesocoms and were divided in 2 parts; shoots and leaves and roots (Bennett et al. 2000).

For the study of imidacloprid, polypropylene tubs (0.77 m [L] x 0.15 m [W] x 0.16 m [H]) in a different shape were used (Figure 6-1). Because of lab space restrictions and time to manage the sampling, 6 smaller tubs were used in this trial which was set up 10 days later than the trial with lambda cyhalothrin and chlorothalonil (Figure 6-2). Two tubs were planted with *P. australis* (4 plants/tub), 2 tubs operated without the presence of plants, while 2 tubs functioned as a control. Each tub was filled with a mixture of 10 litres (equal to 5 kg) universal pot soil and 5 litres (equal to 7.2 kg) of soil (sandy loam). The potting soil was applied to promote the growth of wetland plants (USDA 1998). The tubs were gradually (1 cm/day) filled with water up to a level of 5 cm above soil (equal to a free water volume of 5.8 liters). After a waiting period of 5 weeks, water from the mesocosms was amended with imidacloprid stock solution to reach a target pesticide concentration (typically found in runoff) of 250 µg/l (see section 6.2.2). After this step, water was sampled in a similar manner as described for lambda-cyahlothrin and chlorothalonil at time 0 h, 2 h, 4 h, 6 h, 22 h, 76 h, 145 h and 240 h. This was equal to a batch exposure of 10 days, and chosen because the minimum interval between two recommended imidacloprid applications in the field was equal to 7 days. As was the case for lambda-cyahlothrin and chlorothalonil, a batch consisted of addition of pesticide stock solution, to both vegetated and non-vegetated mesocosms at time 0 hours, followed by a 10-day exposure, during which time sampling and analysis of water took place. This, to determine the dissipation/retention of the pesticide during this time interval. Evapotranspiration in each mesocosm was adjusted by the addition of tap water, which was approximately 0.24 litres per day. Batch 1 started at day 1, by making use of mesocosms 11, 12, 13 and 14 and ended on day 10. The second batch (no. 2) started at day 11 and ended on day 20, by addition of the same amount of pesticide to the same mesocosms (11, 12, 13 and 14) used in batch 1 for a time-span of 10 days. The other batches (3, 4 and 5) were executed according to a similar method e.g. batch 3 was executed from day 21-day 30, batch 4 from day 31 day 40 and batch 5 from day 41 up to day 50. After 5 batches (50 days), plant samples in each tub were harvested, and analyzed in a similar way as for the 2 other pesticides.

6.2.2 Pesticide applications

Plants were given a 5-week adaptation period, prior to pesticide application. The amount of pesticides dosed was based on field application rates (1.25 kg/ha chlorothalonil, 0.25 kg/ha lambda-cyhalothrin and 0.48 kg/ha imidacloprid).

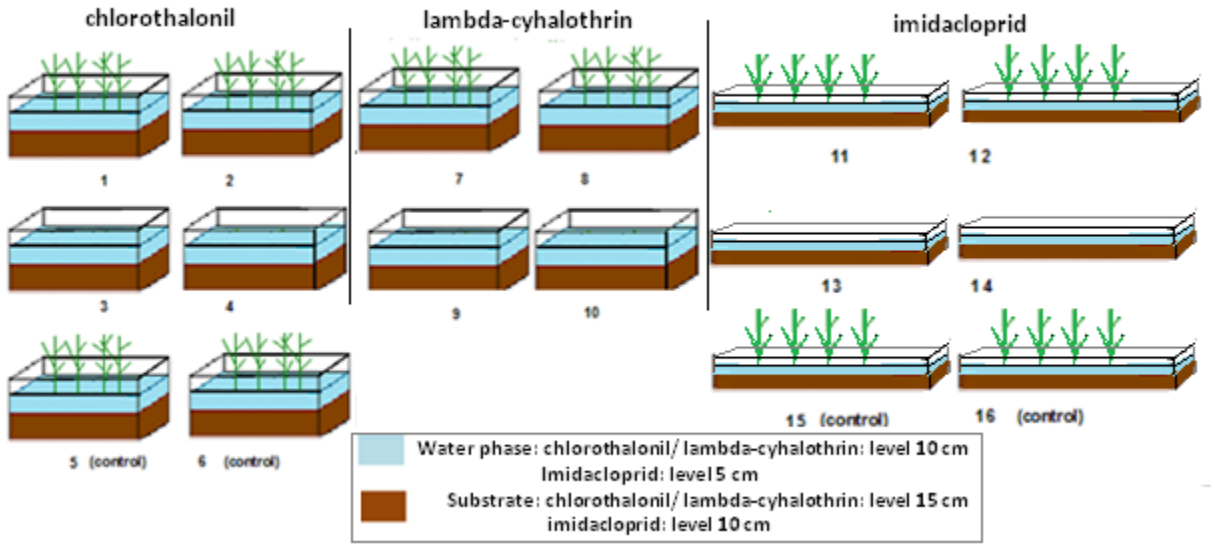

Figure 6-1: Experimental set-up of mesocosms: vegetated mesocosms (P. australis) ([1+2]; [7+8]; [11+12]) and non-vegetated mesocosms ([3+4]; [9+10]; [13+14]). Mesocoms 1, 2, 3, 4 were exposed to chlorothalonil; mesocosms 7, 8, 9, 10 were exposed to lambda-cyhalothrin; mesocosms 11, 12, 13, 14 were exposed to imidachloprid and mesocosms 5, 6, 15, 16 were the controls with no pesticides. Mesocosm 2 is a parallel replicate of 1, mesocosm 4 of 3, mesocosm 6 of 5, mesocosm 8 of 7, mesocosm 10 of 9, mesocosm 12 of 11, mesocosm 14 of 13 and mesocosm 16 of 15

For this calculation, a typical field size of 500 m^2 was chosen. This takes into account the assumption of a runoff of 1% for chlorothalonil and imidacloprid and 5% (worst-case scenario) for lambda-cyhalothrin (Moore et al. 2001) and considers a rainfall event of 10 mm for which 50% runs off. All pesticides were purchased from Dr Ehrenstorfer GmbH, Germany.

Each applied dose was made by mixing the appropriate amount (10 ml for chlorothalonil and lambda-cyhalothrin and 1.5 ml for imidacloprid) of pesticide stock solution (1000 mg/l) with two litres of water from the mesocosms. This was done by gentle stirring in a beaker and subsequent addition to the mesocosm. The total volume of water was 20 litres for the chlorothalonil and lambda-cyhalothrin amended mesocosms and 12 litres for the imidacloprid amended mesocosms. After addition of pesticides, the water in the mesocosms was mixed by gently stirring with a glass rod. This stirring was repeated each time, before sampling of the water phase.

Figure 6-2: Image of the experimental set-up, with smaller mesocosms (brown) used for the imidacloprid experiments

6.2.3 Statistical analysis

Statistical analysis (Sigma plot version 12) was used and two main tests were performed. If the data fulfilled the assumptions for ANOVA (a normal distribution and equal variance), a 'Two Way ANOVA' was carried out to compare pesticide removal efficiencies with the factors vegetation/no vegetation and the factor time. Because the data did not fulfil the assumptions for ANOVA, a Kruskal-Wallis One Way Anova on Ranks was applied to investigate significant differences in removal efficiency among vegetated and non-vegetated batches and between different time intervals. In the case of significant differences a pair wise, multiple comparison procedure (Tukey or Dunn's method) was used. The output is presented in appendix 4 (statistical outputs for chapter 6).

6.2.4 Pesticide analysis

Lambda-cyhalothrin and chlorothalonil were analyzed in water and plants according to a procedure described by Bennett et al. (2000). The amounts in sediment could not be quantified, because of problems with the storage of these samples. All plants were harvested from the mesocoms, and were dried with tissuepaper and wet biomass was determined by measuring the weight with an analytical balance (Brand: Mettler). After this step, plants were divided in shoots and leaves, and roots, and their masses and moisture content were determined. For both, plant upper part (shoots and leaves) and roots, the sonication method was applied, which is an extraction with ethyl acetate. Samples were prewetted with 1 mL of ultrapure water prior

to the addition of ethyl acetate. The mixture was agitated with a sonicator (Transonic T700 (brand: Elma)) for 1 min in pulse mode using an 80% duty cycle. Following sonication, the mixture was centrifuged 2000-2500 rpm) and cleaned up by means of a mixture of silica gel and sodium sulphate (placed in a column) and making use of different eluents ((a) 5 mL of hexane and b) 10 mL of 10% acetone in hexane). After cleanup the collected extract was evaporated up to near dryness and redissolved in 1 ml hexane. Quantification was done with a GCMS 6890N (Agilent technologies) equipped with a HP5 ms column. The limit of detection (LOD) for lambda-cyhalothrin was 0.001 µg/l water and 0.01 µg/kg plant, while for chlorothalonil this was 0.1 µg/l water and 0.02 mg/kg plant.

Water samples containing imidacloprid were filtered through a syringe filter with a PVDF membrane and a pore size of 0.22 µm (Carl Roth, Karlsruke-Rheinhafen, Germany) Plant samples were analyzed by a method described by Baskaran et al. (1999), involving an extraction with a mixture of acetonitrile and water (80:20 by volume; 40 ml) followed by centrifugation and filtration of the supernatant through a PVDF membrane. Imidacloprid was quantified by means of HPLC (Thermo Surveyor) equipped with a surveyor LC pump plus, a platinum C18ESP 3 µm column and a Surveyor PDA plus 5 detector (Thermofisher). The LOD for imidacloprid in water and plants was respectively 0.5 µg/l and 0.5 mg/kg.

6.2.5 Pesticide dissipation

6.2.5.1 Calculation of the removal efficiency

The removal efficiency of pesticides from the water phase was calculated according to Formula 5-1, section 5.2.4.

6.2.5.2 Calculation of DT_{50} in the water phase

The DT_{50} for pesticides in the water phase of wetland mesocosms was calculated by means of Formula 5-2 and 5-3 presented in section 5.2.5. The distribution of the DT_{50} values was evaluated by means of a box plot. Outliers were identified by means of the inter quartile range (IQR), which is the difference between the 3th quartile and the 1th quartile. For each dataset, border values were found by means of respectively: 3th quartile+1.5 IQR (upper limit) and 1th quartile-1.5 IQR (Lower limit). Results for the calculation of the IQR are presented in appendix 6 (Tables f-7 up to f-9). All values out of this range are noted as outliers.

6.2.5.3 Design of a field scale constructed wetland

WRP (1994) provides information for a simplistic design of a field scale wetland. The simplification directs to the single (A → B) system approach based on a first order reaction and because it is used for an initial feasibility study.
The equation for the chemical half-life (DT_{50}) of pesticides (Formula 5-2) was rearranged as follows:

$k = \frac{-1 ln \frac{Cx}{C0}}{DT50}$ *(Formula 6-1)*

In which:

Cx: concentration at time x hours
C_0: concentration at time 0 hours
k: removal coefficient

The Hydraulic Retention Time (HRT) was calculated by an arrangement of Formula 6-1.

$HRT = \frac{ln \frac{C}{C0}}{-1.k}$ *(Formula 6-2)*

The wetland area was calculated by means of Formula 6-3.

$Wetland\ Area = \frac{Flow * HRT}{Depth}$ *(Formula 6-3)*

6.2.5.4 Measurement of the flow

To estimate the flow needed for the calculation of the wetland area, a field experiment was conducted by supplying a 10 mm simulated rainfall to a small agricultural field (typical area 500 m^2), for 20 minutes by means of a 1.5 inch water pump (brand: Bageo) and a 50 cm length plastic pipe. The pipe, which was connected to the hose and had a narrow opening (few mm) to evenly distribute the water over the field. Water from the field was received in an adjacent drainage ditch (size 14m (L) x 1.4m (W) x 0.5m (H)).
After the simulated rainfall, the flow in the ditch was measured with a velocity meter (Brand: Geopacks) by means of placing the velocity rotor in the flowing ditch water. Velocity counts were measured. The flow was estimated to be 38.8 m^3/day by making use of the dimensions of the ditch near the outlet and a conversion provided in the meter's user manual. This experiment was repeated twice.

6.3 Results

6.3.1 Dissipation of pesticides from the water phase

6.3.1.1 Fate of chlorothalonil

Results for the dissipation of chlorothalonil from the water phase in vegetated and non-vegetated mesocosms are presented in Figure 6-3. At time 97 h, the concentration was below the limit of detection (0.1 µg/l) in batch 1. In batch 2 this was already the case at time 72 h for vegetated mesocosms, however for consecutive batches 3, 4 and 5, removal was less than

100% and varied between 93.1% (batch 3) and 99.4% (batch 5). At time 334 h (end of the experiment), chlorothalonil was below the limit of detection for all batches, with the exception of batch 4. Here the average concentration obtained for the 2 parallel replicates (see Figure 6-3) decreased from 229.0 µg/l to 2.0 µg/l and from 140.0 µg/l to 16.4 µg/l for respectively vegetated and non-vegetated mesocosms, which corresponds to a reduction of 99.1 and 88.3% respectively during the whole experiment.

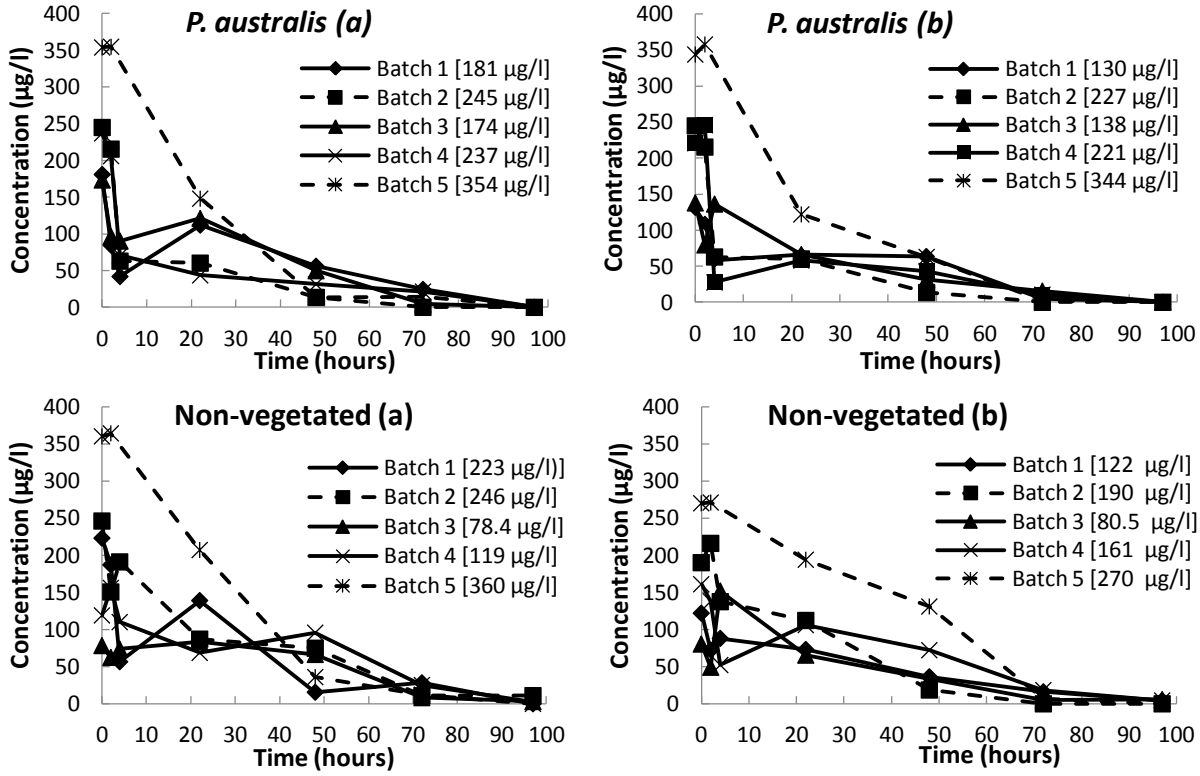

Figure 6-3: Aqueous concentration profile of chlorothalonil obtained for two parallel replicate mesocosms (noted as a and b) over 5 different consecutieve batches involving vegetated (P. australis) and non-vegetated mesocosms. Mesocosms were exposed to a chlorothalonil target concentration of 500 µg/l. The actual starting concentrations are between parenthesis in the legend. For a better presentation of results, time 334 h is removed. At that time the concentrations of chlorothalonil in the vegetated and non-vegetated mesocosms (except batch 4) were below the LOD

At time 0 h, a much higher concentration is measured in batch 5 compared to batch 1 up to batch 4, at time 0 h and during the remainder of the experiment, for both vegetated and non-vegetated mesocosms. In batch 5, concentrations were higher in vegetated than in non-vegetated mesocosms and higher than in previous batches (1-4). A Kruskall-Wallis 'One Way Anova on Ranks' followed by a multiple comparison procedure (Tukey test) showed no statistically significant differences (a result 'Do Not Test' was obtained for all comparisons) between the median values obtained for the removal efficiency among vegetated and non-vegetated batches, indicating that vegetation did not play a significant role in pesticide removal.

6.3.1.2 Fate of lambda-cyhalothrin

Results for the dissipation of lambda-cyhalothrin from the water phase are presented in Figure 6-4. At the end of the experiment (334 h) concentrations were below the limit of detection (0.001 µg/l) for all batches, with the exception of batch 3 (16.8 µg/l and 15.5 µg/l) and batch 4 (30.9 µg/l and 50.1 µg/l) for respectively vegetated and non-vegetated mesocosms. This complies with a reduction in the concentration of respectively 43.8% and 64.2% (batch 3) and 0% (batch 4). In batch 4, at all time-intervals, concentrations were higher than the initial concentration measured at time 0 h. Similar to results for chlorothalonil, high lambda-cyhalothrin concentrations (average values) were measured in batch 5 (167.0 and 233.0 µg/l) compared to previous batches (concentrations between 9.4-53.4 µg/l) at time 0 h for both vegetated and non-vegetated mesocosms. It is possible that pesticide desorption or remobilization occurred, because pesticides were applied to the same mesocosms during all batches.

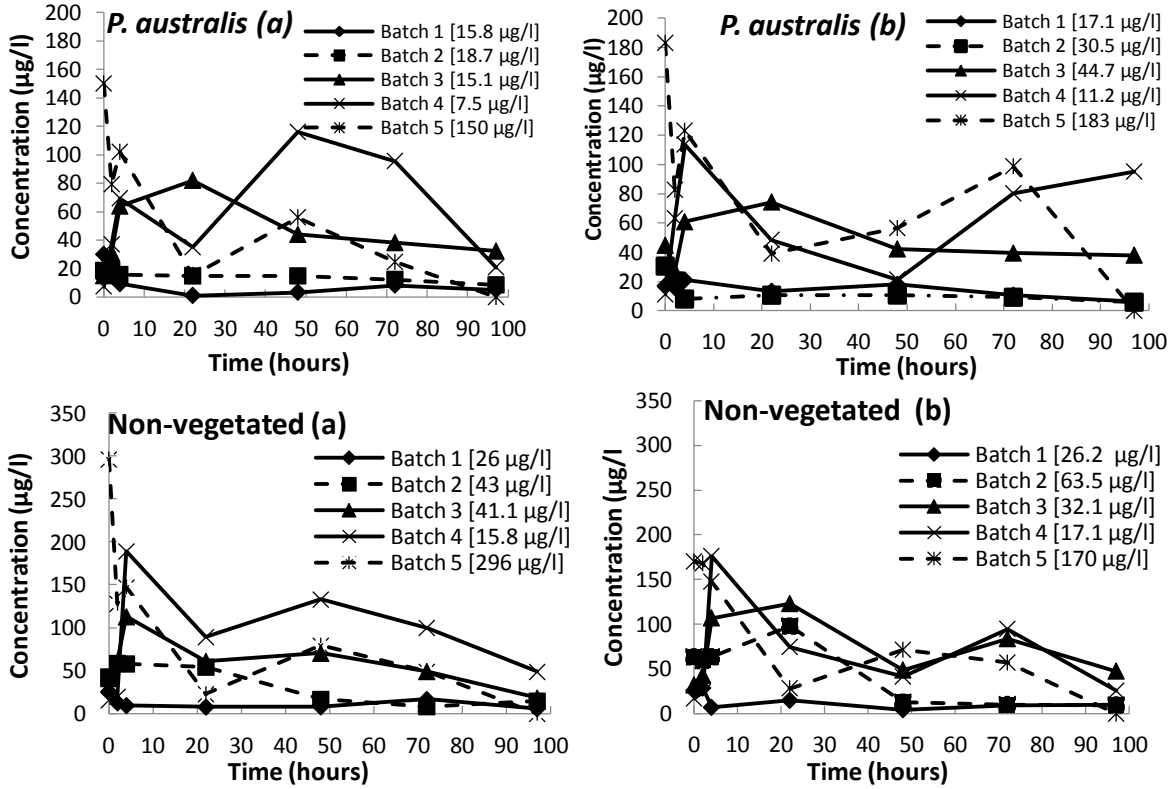

Figure 6-4: Aqueous concentration profile of lambda-cyhalothrin obtained for two parallel replicate mesocosms (noted as a and b). Vegetated (P. australis) and non-vegetated mesocosms were exposed to a lambda-cyhalothrin target concentration of 500 µg/l during five consecutive batches The actual starting concentrations are between parenthesis in the legend. For a better presentation of results, time 334 h is removed. At that time the concentrations of lambda-cyhalothrin in the vegetated mesocosms (batches 2 and 5) and non-vegetated mesocosms (batches 1, 2 and 5) were below the LOD

Statistical analyses showed that data for lambda-cyhalothrin was not normally distributed. A Kruskall-Wallis 'One Way Anova on Ranks' followed by a multiple comparison procedure (Tukey test) showed no statistically significant differences between the median removal efficiencies among vegetated and non-vegetated batches.

6.3.1.3 Fate of imidacloprid

Results for the dissipation of imidacloprid from the water phase are presented in Figure 6-5. Imidacloprid concentrations decreased from concentrations varying between 151.9 µg/l and 301.2 µg/l at time 0 h to concentrations between 7.1 µg/l and 71.3 µg/l at 240 h. This complies with an average reduction of 94.0% (batch 1), 97.1% (batch 2), 86.7 (batch 3), 96.7% (batch 4) and 100% (batch 5) for the vegetated mesocosms, while for non-vegetated mesocosms the reduction was respectively 92.2% (batch 1), 97.1% (batch 2), 75.5% (batch 3), 97.8% (batch 4) and 100% (batch 5).

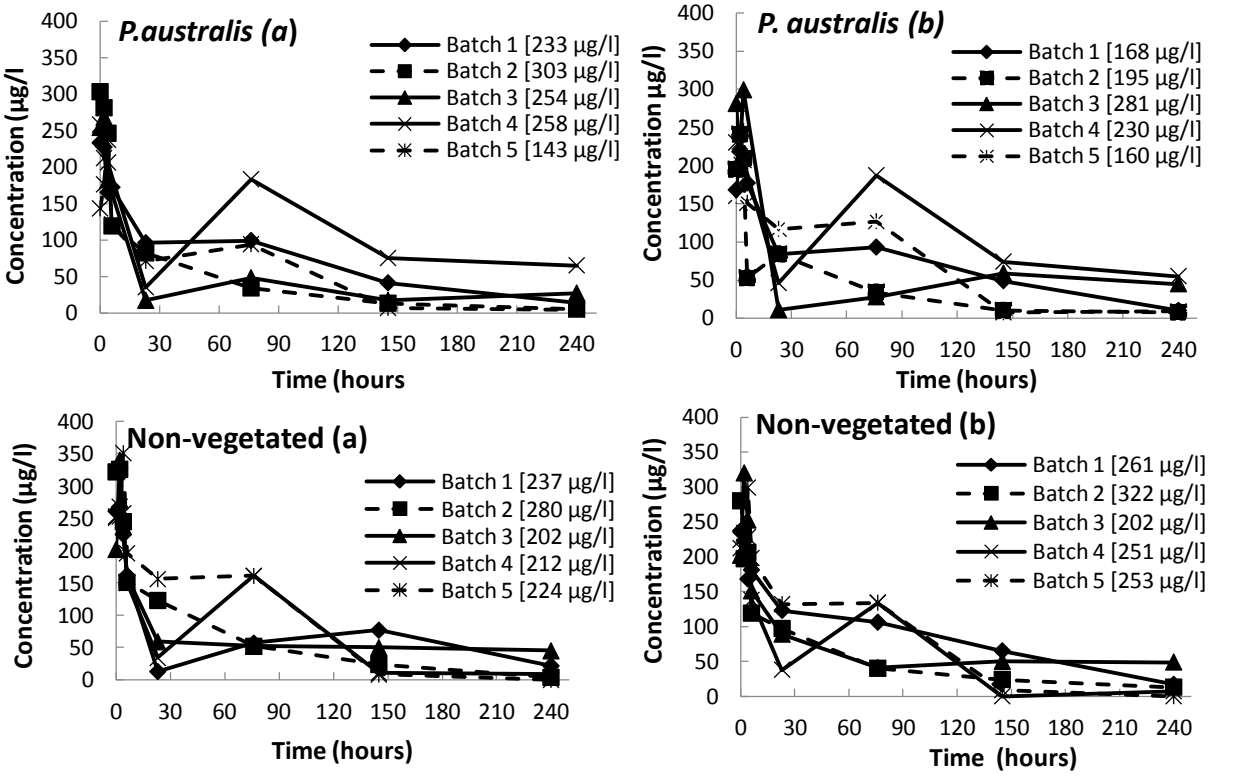

Figure 6-5: Aqueous concentration profile of imidacloprid obtained for two parallel replicate mesocosms (noted as a and b). Vegetated (P. australis) and non-vegetated mesocosms were exposed to a imidacloprid target concentration of 250 µg/l during 5 consecutive batches The actual starting concentrations are between parenthesis in the legend

Statistical analysis showed that data for imidacloprid was not normally distributed. A Kruskall-Wallis 'One Way Anova on Ranks' followed by a multiple comparison procedure (Dunn's method) showed no statistically significant differences (a result 'Do Not Test' was obtained for

all comparisons, indicating no significant differences) between the median removal efficiencies among vegetated and non-vegetated batches.

6.3.2 Pesticide analysis of shoots and leaves and roots

The amount of the pesticides detected in plants of *Phragmites australis* is presented in Table 6-2. From the total amount of chlorothalonil and lambda-cyhalothrin added (50.0 mg) to the mesocosms a total of respectively 103.1 µg (0.20%) and 879.3 µg (1.76%) was detected in the *Phragmites australis* plants. For imidacloprid this was 333.2 µg or 1.8%. The amount of pesticide in µg/g DW was the highest for lambda-cyhalothrin which was followed by imidacloprid.

Table 6-2: Amount of pesticides detected in shoots and leaves and roots (n=2)[a] in wetland mesocosms planted with P. australis

Pesticide	**Chlorothalonil**			**Lambda-cyhalothrin**			**Imidacloprid**		
Total mass added (mg)	**50.0**			**50.0**			**18.8**		
Medium	**µg**	**% of total**	**µg/g DW**	**µg**	**% of total**	**µg/g DW**	**µg**	**% of total**	**µg/g DW**
Shoots and leaves	97.0	**0.19**		319.2	**0.64**		204.8	**1.1**	
Roots	6.2	**0.01**	0.94	560.1	**1.12**	11.4	128.4	**0.7**	1.6
Total in plants	103.1	**0.20**		879.3	**1.76**		333.2	**1.8**	

[a] *Values are the average of residue concentrations obtained for two samples*

6.3.3 Calculation of the DT50 for chlorothalonil, lambda-cyhalothrin and imidacloprid

The results obtained for the half-life of chlorothalonil in vegetated mesocosms was on average 15.9 h (n=5) for vegetated and 20.0 h (n=5) for non-vegetated mesocosms. The R^2 values ranged between 0.8166 and 0.9336, indicating a much better correlation with the first order kinetics than obtained for lambda-cyhalothrin (see Table 6-3). The half-life of lambda-cyhalothrin was on average 94.2 h for the vegetated and 70.1 h for the non-vegetated mesocosms, with R^2 values ranging between 0.5564 and 0.7119. Results obtained for batch 4 were seen as outliers (because of the major difference with other DT_{50} values and very low R_2 values). They were excluded from the average half-life calculations.
The half-lives in batches 2 and 5 were almost half of the values obtained in batch 1. The half-lives of imidacloprid (see Table 6-3) in vegetated mesocosms were on average 57.3 h for vegetated and 68.1 h for non-vegetated mesocosms.

Table 6-3: Overview of reaction rate constants (k) and correlation coefficients (R^2) derived from linear regressions with the calculated average[a] half-life time values for pesticides in the water phase of vegetated (Avveg) and non-vegetated (Avnonveg) mesocosms. Outliers for the half-life time are presented in red

Mesocosm	**Chlorothalonil**			**Lambda-cyhalothrin**			**Imidacloprid**		
	k $(h)^{-1}$	**R^2**	**DT_{50} (h)**						
B1veg	0.0335	0.8468	20.7	0.0114	0.6842	61.1	0.0110	0.9414	63.0
B1nonveg	0.0322	0.8643	21.6	0.0102	0.6833	68.0	0.0060	0.9764	115.5
B2veg	0.0744	0.9107	9.3	0.0084	0.6833	68.0	0.0060	0.9764	115.5
B2nonveg	0.048	0.9228	14.4	0.0218	0.6833	31.9	0.0157	0.9376	44.2
B3veg	0.0371	0.9336	18.7	0.0037	0.5839	189.9	0.0126	0.7575	55.7
B3nonveg	0.0400	0.8166	17.3	0.0047	0.5613	149.1	0.0132	0.8823	55.7
B4veg	0.0340	0.8966	20.4	0.0010	0.0208	693.2	0.0126	0.8561	55.2
B4nonveg	0.0204	0.8667	34.0	0.0016	0.0709	433.2	0.0181	0.9477	38.3
B5veg	0.0678	0.8921	10.2	0.016	0.5564	43.3	0.0098	0.7134	70.7
B5nonveg	0.0547	0.8897	12.8	0.022	0.7119	31.5	0.0080	0.7376	86.6
Avveg[a]	0.0494	0.896	**15.9**	0.0099	0.6270	**94.2**	0.0125	0.8319	**57.3**
±	±	±	**±**	±	±	**±**	±	±	**±**
SD (h)	0.0200	0.0319	**5.6**	0.0052	0.0665	**65.9**	0.0026	0.0944	**10.7**
Avnonveg	0.0391	0.872	**20 .1 .0**	0.0147	0.6600	**70.1**	0.0122	0.8963	**68.1**
±	±	±	**±**	±	±	**±**	±	±	**±**
SD (h)	0.0134	0.0389	**8.5**	0.0086	0.0671	**55.4**	0.0051	0.0051	**32.4**
Medianveg[b] (h)		**18.7**			**64.6**			**63.0**	
Median nonveg (h)		**17.3**			**50.0**			**55.7**	
DT_{50} range veg (h)	**-5.1-35.7/[0-35.7][b]**			**1.8-161.2**			**33.2-93.2**		
DT_{50} range Nonveg (h)	**7.2-28.8**			**-52.9-173.0/[0-173][b]**			**1.8-129**		

[a]*The average half-life was calculated without the values obtained in batch 4 (excluded (very low R^2))*
[b]*Values between parenthesis represent the DT_{50} range with omission of the negative value*

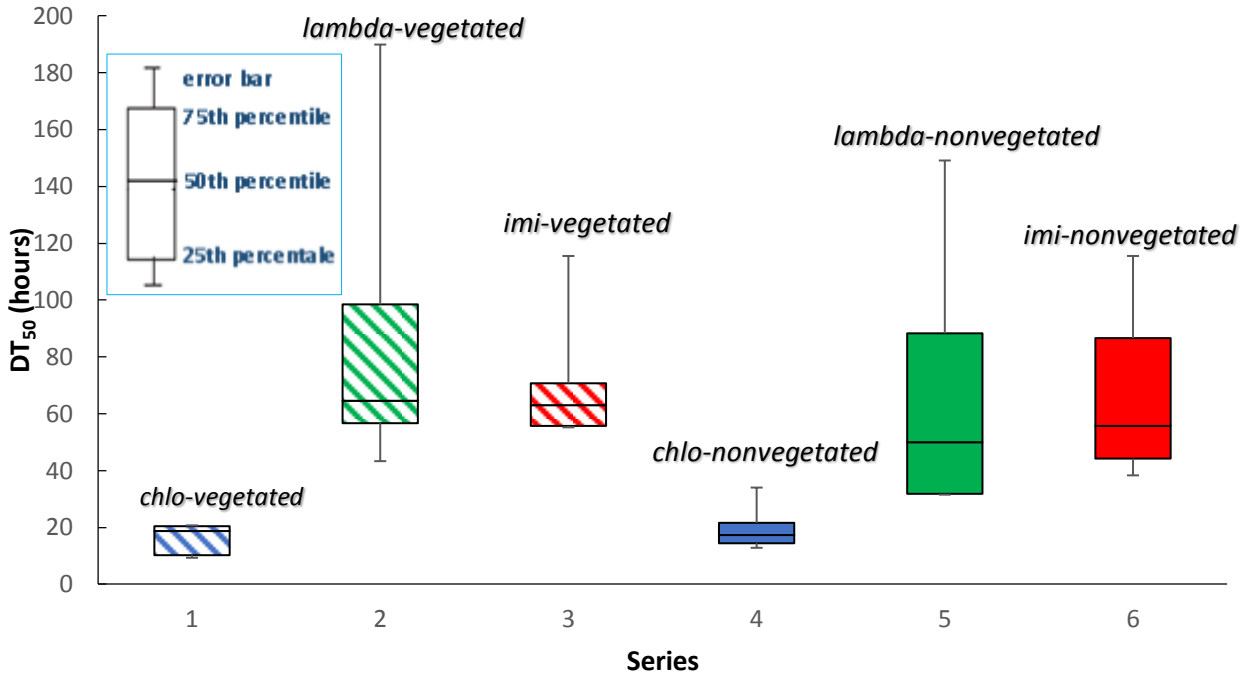

Figure 6-6: Boxplots showing the distribution of half-life time values of chlorothalonil (chlo), lambda-cyhalothrin (lambda) and imidacloprid (imi) in the water phase of vegetated and non-vegetated mesocosms obtained during a total of 5 batches (Table 6-3). The results are given over 6 series e.g. series 1 is the distribution of average half-life time values of chlorothalonil in vegetated mesocosms; series 6 is the distribution of average half-time values of imidacloprid in non-vegetated mesocosms

Comparison of the median values showed small differences in distribution of the DT_{50} values for chlorothalonil, while much higher differences were found for lambda-cyhalothrin and imidacloprid, with the highest for lambda-cyhalothrin (see Table 6-3 and Figure 6-6). The average half-life time values of chlorothalonil (15.9 h and 20.0 h in respectively vegetated and non-vegetated mesocosms) are slightly lower (negative skewness: median value 18.7 h) and higher (positive skewness: median value 17.3). The results obtained for the distribution and the differences in positive and negative error bars for each pesticide clearly indicate that the data for the half-life time values of the 3 pesticides in the water phase of vegetated and non-vegetated mesocosms is not normally distributed. This was confirmed by performing the two-way ANOVA tests (see appendix 4, statistical outputs for chapter 6). It is therefore appropriate to use median values for the half-life instead of the average values.

6.3.4 Basic design considerations for a field scale constructed wetland

The results from the mesocosm experiments showed median half-life time values of 18.7 h for chlorothanlonil, 64.6 h for lambda-cyhalothrin and 63.0 h for imidacloprid in mesocosms planted with *Phragmites australis* and average k-values of 0.0494 h^{-1}, 0.0099 h^{-1} and 0.0125 h^{-1} respectively. For design considerations, the highest average DT_{50} value of lambda-cyhalothrin

was used to calculate the wetland area dimensions. This complies with a k-value of 0.0099 h^{-1}. To achieve an overall pesticide reduction of 90%, the ratio $\frac{C}{C0}$ becomes $\frac{10}{100}$ or 0.1 (see Formula 6-1). The HRT calculated by means of Formula 6-2 becomes 233 h or 9.7 days. Field measurements were conducted to calculate the average flow, which was 38.8 m^3/day for a typical small agricultural area of 500 m^2 in the research area. Assuming a depth of 1 m, the wetland area (Formula 6-3) was calculated to have an area of 375 m^2. Taking into consideration a width of 5 m, the length becomes 75 m. The proposed dimension for a field scale surface flow wetland for treatment of agricultural runoff consisting of chlorothalonil, lambda-cyhalothrin and imidacloprid becomes 75 m (length) x 5 m (width) x 1 m (depth). Soil porosity values were not available for the research area and a value of 1 was used, which is according to Kadlec and Knight (1996) a good approximation. Information was retrieved from farmers and the local engineer in the research area that a depth of 1 m is sufficient to store a free water volume of at least 0.5 m under the present hydrological conditions. Another aspect and advantage is that the soil of the planned wetland in that area is clay, which is impermeable and is known to function as a natural liner preventing contaminated water to leach.

6.3.5 Selection of vegetation

In the previous chapter, the total amount of lambda-cyhalothrin and imidacloprid in *Nymphaea amazonum* plants was respectively 2.02 and 2085 µg/g DW and for *Eleocharis mutata,* this was 0.5 and 13.5 µg/g DW respectively. Compared to these plants, more lambda-cyhalothrin was detected in present study in *P. australis* (11.4 µg/g DW), while the highest amount of imidacloprid was found in *Nymphaea amazonum* plants. The amount of lambda-cyhalothrin found in plants (*N. amazonum*) was comparable with the findings in Bouldin et al. (2005) for *Ludwigia peploides*, while for imidacloprid no results of plant uptake were found in the literature. From results obtained in present observation, it is reasonable to perform mesocosms studies with a mixture of both plants (*P. australis* and *N. amazonum*). Also for the overall wetland performance, multiple plant types are preferred above a monoculture. For instance, in case of high loads, monocultures can die off, and pollutant's removal becomes impaired (Kadlec and Wallace 2009).

6.4 Discussion

6.4.1. Chlorothalonil

Short half-lives of chlorothalonil in water indicate a rapid dissipation. Chlorothalonil removal (100%) was more evident in the repeated batch 2 (at time 72 h) for the vegetated mesocosms compared to batch 1 (at time 97 h). In consecutive batches 3 and 4 however chlorothalonil was not below the limit of detection at time 72 h. Removal decreased to 93.1% to increase again to 99.4% (batch 5). Increased removal rates may be possible due to enhanced biodegradation rates, when mesocosms are repeatedly exposed to the same pesticide (Singh et al. 2003; Budd et al. 2011). However, this trend was not observed for the non-vegetated mesocosm, and

therefore indicates the positive influence of wetland plants on pesticide removal (Vymazal and Březinová 2015). According to Table 6-1, volatilization of chlorothalonil from the mesocosms might also have occurred. Concentrations in batch 5 were higher in vegetated than in non-vegetated mesocosms and also higher than those in batches 1 up to 4 for both vegetated and non-vegetated mesocosms. This may explain the influence of a more dense vegetation present in the end of the experiment or a possible desorption or remobilization (Passeport et al. 2013; Vallee et al. 2014) of pesticides from the sediment. According to Moore et al. (2013), more dense vegetation inhibits the sedimentation of pesticides and therefore more pesticides are present in other compartments (e.g. water, plants). Statistically, however, no significant differences in removal rates were found between vegetated and non-vegetated mesocosms.

In the study of Sherrard et al. (2004), chlorothalonil was dosed (326 µg/l) in vegetated mesocosms planted with *Scirpus cyperius*. The concentrations in water at time 45 h were already below the limit of detection (0.1 µg/l), while in present study this occurred at time 97 h for batch 1 and 5 (vegetated mesocosms) and at 334 h (for all other batches, with the exception of batch 4). The chlorothalonil removal rate constant presented in Sherrard et al. (2004) was 0.263 h^{-1}, while in present study this was much lower for the vegetated mesocosms (0.0494 h^{-1}). In both studies, data followed a first order reaction rate model. The differences between both experiments were different target concentrations, vegetation, sediment, different hardness of water and exposure time. No data was given on the temperature and relative humidity. According to US EPA (2012), degradation rates have shown to be dependent on the application rates; the higher the application rate, the slower the degradation rates. In present study the mesocosms were exposed to 500 µg/l chlorothalonil, while in Sherrard et al. (2004), the exposure concentration was 326 µg/l. Lower degradation rates are also obtained when microbial activity is limited and hydrological residence times are short (US EPA 1999).

The median half-life time of chlorothalonil in water in present study was 0.7 days for both vegetated and non-vegetated mesocosms. This falls within the range of half-lives in water of 0.18-8.8 days given in Chaves et al. (2007), but is higher than the 0.1 day given in the PPDB database of the University of Hertfordshire (2016). The half-life time (18.7 h) estimated by means of a linear regression showed to be somewhat higher than the time found (around 4 h) for 50% decrease of the initial concentrations (at time 0 h) in vegetated mesocosms (Figure 6-3). However, considering the high R^2 values obtained for that regression, half-life time values will likely be around 18.7 h. In the study of Sherrard et al. (2004), the half-life was around 0.13 days (3.4 h). That study also revealed that repeated exposure of mesocosms with chlorothalonil did not improve the removal rate significantly. Although, the amount of chlorothalonil in *P. australis* was not quantified in the study of Casas-Zapata et al. (2013), they suggest that effective removal of chlorothalonil can be achieved up to concentrations of 385 µg/l in horizontal subsurface flow constructed wetlands planted with *P. australis*.

The amount of chlorothalonil detected in *P. australis* in present study was 0.9 µg/g DW. No comparable results were found in other reports using *P. australis*. Only amounts detected in the shoots and leaves of lettuce (1.4 µg/g DW) and radish (1.3 µg/g) are available (Żebrowski et al.

2008). In present study, the major portion (about 19 times higher than in roots) was also detected in the plant upper part (shoots and leaves). Pesticides are transported and concentrated in the harvestable part (e.g. shoots and leaves) of the plant by means of phytoextraction/accumulation (Truu et al. 2015).

6.4.2. Lambda-cyhalothrin

Overall, a fairly good and rapid dissipation was observed for lambda-cyhalothrin from the water phase, with the exception of batches 3 and 4 (for both vegetated and non-vegetated mesocosms). According to Fogarty and Tuovinen (1991), application of a higher dose of lambda-cyhalothrin results in temporary high aqueous concentrations, which inhibit microbiological activity. In Munoz-leoz et al. (2009), the ability of the pyrethroid, deltamethrin to inhibit soil microbial respiration under anaerobic conditions was proven. A rapid dissipation of lambda-cyhalothrin and other pyrethroids from the water phase is mentioned in several articles (Moore et al. 2001; Bouldin et al. 2005; Cooper et al. 2004; Mahabali and Spanoghe 2014). This is mainly linked to their physico-chemical properties such as a very low water solubility and a high log K_{ow}. Faster dissipation was not obtained in the repeated batches in present study, this in contrary to the previous experiment described in chapter 5 (Mahabali and Spanoghe 2014), in which lambda-cyhalothrin was exposed to low (10 µg/l) and high (30 µg/l) concentrations. For both concentrations repeated batches resulted in a faster removal of lambda-cyahlothrin e.g. 48 h instead of 72 h. The mesocosms in present research were exposed to much higher concentrations (500 µg/l) and complete removal occurred for e.g. batch 1 and batch 2 only at the end of the experiment (time 334 h).

The average dissipation or degradation rate constant (k) was 0.0099 h^{-1} for *P. australis* mesocosms exposed to 500 µg/l lambda-cyhalothrin, while in previous chapter (results described in Mahabali and Spanoghe (2014) this was on average 0.031 h^{-1} for *Nymphaea amazonum* mesocosms and 0.030 h^{-1} for *Eleocharis mutata* mesocosms exposed to 30 µg/l lambda-cyhalothrin. The lower reaction rate constant may be the result of lower temperatures (18-22 ^{o}C), as the experiment was conducted in Belgium, in contrast to the 26-28 ^{o}C range measured in Suriname. In general, the rate of microbial degradation or transformation doubles for every 10 °C increase in temperature (Yu et al. 2007). Microbial degradation can also be temporarily impaired because of sedimentation or strong sorption, making pesticides less bio available for degradation (O'Geen et al. 2010). It may also be related to the differences in wetland substrate characteristics (e.g. OM content, pH) and to the much higher pesticide application rates, which according to US EPA (2012) results in lower removal rates. Although comparable soil types (sandy loam) were used, the values for organic matter (3.5% compared to 0.46% of dry solids) and pH (7.2 compared to 6.5) were higher in present study and may have resulted in different microbial communities as observed in the study of Elsayed et al. (2015). Different microbial communities in that study involving the degradation of chloroacetanilide herbicides were linked to factors such as sediment organic matter, nutrient cycling and different redox conditions. However, assessment of the different microbial communities present in mesocosms was not done in present study. Higher removal rates and lower half-live

times were found for the non-vegetated mesocosms compared to the vegetated ones, suggesting that vegetation does not play a role in the removal of lambda-cyhalothrin. In several studies (Leistra et al. 2003; Moore et al. 2009A; Mahabali and Spanoghe 2014), the main pathway for lambda-cyhalothrin's (and other pyrethroids) dissipation was through sedimentation, while in other studies (Moore et al. 2001; Bennet et al. 2005; Lizotte et al. 2014) uptake by plants played a major role. Higher plant densities resulted in higher pesticide uptake and therefore caused a reduced amount adsorbed to the sediment fraction. Compared to Mahabali and Spanoghe (2014), lambda-cyhalothrin's uptake by *P. australis* in present study, was much higher than for both plants used in that experiments. An explanation for that can be a different effect of the plant's density and type on the uptake of lambda-cyhalothrin.

The median half-life time of lambda-cyhalothrin in water was 3 days in vegetated and 2 days in non-vegetated mesocosms and shows to be somewhat higher than the results derived from Figure 6-4, for batch 1, lower for batch 2 and much higher for batch 5, in vegetated systems. Results in batch 3 showed much higher values (around 14 days) for a 50% dissipation for both vegetated and non-vegetated systems, while batch 4 could not be evaluated because of increasing concentrations with time. The linear regression also resulted in rather low R^2 values. The results from present study are slightly lower than the DT_{50} of 4.6 days, found by Moore et al. (2001), in which study a drainage ditch was exposed to a comparable concentration of 460 µg/l lambda-cyhalothrin. Much lower half-lives (≤ 1 day) were found by Hand et al. (2001) and Mahabali and Spanoghe (2014). They observed that higher half-lives were related to the higher application rates of pesticides (Kanrar et al. 2006; US EPA 2012). Comparison with previous studies (Bennett et al. 2005; Boudin et al. 2005; Mahabali and Spanoghe 2014) shows that the amount of lambda-cyhalothrin per gram DW of plant material (11.4 µg/g DW) in this study is much higher. In those studies the maximum measured lambda-cyhalothrin concentrations were 8.79 µg/g DW (mixture of *Ludwiga*, *Lemna* and *Polygonum*); 0.86 µg/g DW (*L. peploides*) and 2.02 µg/g DW (*Nymphaea amazonum*) respectively.

6.4.3. Imidacloprid

In all batches, imidacloprid concentrations in the water phase were reduced to more than 75%. The highest removal (100%) was obtained in batch 5 (both vegetated and non-vegetated mesocosms) at time 240 h, which might be caused by enhanced biodegradation caused by repeated exposures (Singh et al. 2003). Other wetland studies also describe this trend (Weaver et al. 2004; Rose et al. 2006; Budd et al. 2011; Mahabali and Spanoghe 2014).
The amount found in plants was 1.6 µg/g DW, with the highest amount present in shoots and leaves of *P. australis*. Similar trends in occurrences of pesticides in shoots and leaves were also found in other studies (Wilson et al. 2000; Alsayeda et al. 2008; Mahabali and Spanoghe 2014). However, in Mahabali and Spanoghe (2014), the total amount found in *Nymphaea amazonum* (2085 µg/g DW) and in *Eleocharis mutata* (13.5 µg/g DW) was much higher. According to Truu et al. (2015) wetland plants are capable of removing contaminants e.g. pesticides from soil by their roots (phytoimmobilization or phytostabilization and rhizofiltration), but also transporting and concentrating them in e.g. shoots and leaves (phytoextraction/accumulation).

The median half-life times of imidacloprid in water were 3 days and 2 days for respectively vegetated and non-vegetated mesocosms, which is somewhat higher than the 1-2 days derived from Figure 6-4, for 50% removal from the water phase of vegetated mesocosms, while results for non-vegetated mesoscosms were comparable. The median values fall within the range (1-9 days) of the half-life times found in Mahabali and Spanoghe (2014). However, for the target concentration of 180 µg/l (batch 4) in that study, the half-life time was between 7.1 and 7.7 days and the average degradation rate, 0.0050 h^{-1} compared to the degradation rate of 0.0125 h^{-1} in present study. This difference can however not be explained by the effect of temperature, which was lower in current study and differences might be related to other factors. The target concentration (180 µg/l) is close to the 250 µg/l used in present study.
Similar to higher values obtained for the degradation rate, the DT_{50} in present study was approximately 3 fold lower and was not in agreement with Kanrar et al. (2006), that higher exposure concentrations results in higher half-lives, and might be related to differences in wetland matrix and exposure to sunlight. Imidacloprid is also sensitive towards photolytic degradation (PPDB of the University of Hertfordshire 2016).

In the previous mesocosm study (chapter 5), the same set of vegetated mesocosms were exposed to increasing pesticide concentrations during 2 consecutive batches. From the results, the role of plants was assessed, in the removal/retention of 2 frequently used pesticides (lambda-cyhalothrin and imidacloprid). The amounts removed from water and present in plants and sediment were used to describe the fate of the pesticides. In current study, the same types of pesticides, together with chlorothalonil, were added in a constant and high dose to vegetated and non-vegetated mesocosms during 5 consecutive batches. By comparing the amounts found in different plants for both studies, a selection was made of which plant to choose for pesticide removal in the proposed field wetland.
For design considerations, the half-life time values from present study were used, because of the high exposure concentrations, which are comparable to a worst-case scenario of a high pesticide load in runoff, in Suriname. The inflow of CW receiving agricultural runoff is highly variable in time (Zhang et al. 2015) and it is therefore necessary to evaluate different exposure concentrations in wetland systems such as conducted in the previous chapter. Pollutant removal is often accomplished by manipulating the system's hydraulic and hydrologic conditions and by selecting the appropriate type of (dominant) vegetation (Vymazal 2007; Kadlec and Wallace 2009). It is better to investigate the removal of pesticides using multiple types of plants, because this gives a better representation of the system in nature and plant survival rates are much higher compared to systems with monocultures (Kadlec and Wallace 2009). Calculation of the dimensions of a field scale wetland (375 m^2) was based on an HRT of 9.7 days and a lambda-cyhalothrin reduction of 90% in mesocosms planted with *P. australis.* In Budd et al. (2009), lambda-cyhalothrin's removal was 90% in a much bigger SFCW wetland (area 2.5 ha) with a much lower inflow concentration of 3.3 ng/l and an HRT of 18 hours. In Lizotte et al. (2014), the removal of the pyrethroid cis- and trans permethrin was evaluated in a vegetated (mixture of *P. australis* and *J. effusus*) wetland (area 227 m^2). Removal efficiencies were between 98-100% for an inlet concentration of 5.24 µg/l (see appendix 1), however the HRT

was not provided. Dissipation was achieved by means of sorption to (dense) vegetation, suspended solids (SS) and dissolved organic carbon (DOC) and particle settling. In none of these studies the values for the soil porosity were given. According to Bendoricchio et al. (2000), wetland porosity (ε) has proven difficult to be accurately measured in the field. As a result, wetland porosity values reported in literature are highly variable. Kadlec and Knight (1996) reported that average wetland porosity values are usually greater than 0.95, and ε=1.0 can be used as a good approximation. That was also done in present study (see chapter 2, section 2.7.3).

6.5 Conclusions

The dissipation of 3 types of pesticides, chlorothalonil, lambda-cyhalothrin and imidacloprid was investigated in vegetated *(P. australis)* and non-vegetated mesocosms. The highest and fastest removal was observed for chlorothalonil, followed by imidacloprid and lambda-cyhalothrin. No significant differences in removal between vegetated and non-vegetated mesocosms were observed. This finding is in agreement with the DT_{50} values found for these pesticides in vegetated and non-vegetated systems. To design a CW and calculate its dimensions, the half-life time and the reaction rate constant found for the dissipation of lambda-cyhalothrin were used. A HRT of 9.7 days and a wetland area of 75 m (length) x 5 m (width) x 1m (depth) were calculated. This study indicates that *P. australis* has a relative high uptake towards lambda-cyhalothrin compared to the uptake by other wetland plants in the previous mesocosm study (see chapter 5). It is therefore reasonable to perform wetland studies with a mixture of plants (e.g. a combination of *N. amazonum* and *P. australis*) for the efficient treatment of agricultural runoff containing lambda-cyhalothrin and imidacloprid. Several mechanisms might have played a role in pesticide degradation and must be further investigated. These mechanisms are likely related to pesticides physico-chemical properties (e.g. vapour pressure (lambda-cyhalothrin), solubility, log K_{oc}) and environmental conditions (e.g. type of plants, substrate characteristics, pH) and desorption or remobilization in the wetland. Desorption or remobilization can further be investigated by means of batch sorption experiments

Chapter 7 Feasibility of a field scale constructed wetland to mitigate agricultural pesticides pollution

Abstract
CWs can serve as a cost effective best management practice to decrease non-point source pesticide pollution. The aim of this research is to investigate the removal of the insecticides lambda-cyhalothrin and imidacloprid, in a field scale constructed wetland with a monoculture (phase 1) and using multiple wetland plants types (phase 2). Lambda-cyhalothrin was not found in the water phase of the surface flow wetland, with a few exceptions in phase 2, mainly at the start of the experiment (0-48 h). A good overall removal from the water phase (> 95%) was observed for imidacloprid during both phases 1 and 2, with a somewhat higher and constant pattern in phase 2. Different plant uptake mechanisms were observed for pesticides during both phases such as, the higher accumulation of lambda-cyhalothrin in phase 2 compared to phase 1, which indicates a possible enhancement towards the uptake of this pesticide, in the presence of a multiple plant types. For both pesticides, lower amounts of lambda-cyhalothrin were found in sediment during phase 2 compared to phase 1. Batch sorption experiments showed the highest distribution coefficient (K_d) for lambda-cyhalothrin in soil from the agricultural field, this compared to soil from the inlet of the wetland and the drainage ditch. Although the clay and organic matter content of this soil was much lower, the sorption of lambda-cyhalothrin from aqueous solution onto different soils using batch kinetic experiments, showed a constant pattern in time for lambda-cyhalothrin, while for imidacloprid an irregular pattern was found, indicating the phenomenon of desorption or remobilization. The field scale constructed wetland designed from input data from mesocosm experiments shows good capabilities for removal of lambda-cyhalothrin and imidacloprid from the water phase, especially during repeated exposure and with multiple types of plants.

7.1 Introduction

The ability of wetland mesocosms to mitigate pesticide pollution originating from agricultural runoff is demonstrated in several studies (Moore et al. 2001; Cooper et al. 2004; Bennett et al. 2005; Moore et al. 2011; Dordio and Carvalho 2013: Moore et al. 2013; Mahabali and Spanoghe 2014; Vymazal and Březinová 2015). Frequently used pesticides (chlorothalonil, lambda-cyhalothrin and imidacloprid) in the Commewijne district were studied making use of vegetation that was abundantly present in the research area. In chapter 6, mesocosm experiments were performed in 2010 at Ghent University, Belgium, by making use of *Phragmites australis,* which is one of the frequently used plants in wetland studies (Vymazal 2013C). Another mesocosm experiment was performed in Suriname (year 2012), by making use of 2 other types of plants, (*Eleocharis mutata* and *Nymphaea amazonum*), which were abundantly present in the main ditches of the research area and addressed in chapter 5. The results from those experiments were further used to design a field scale wetland for the

removal of 2 frequently used pesticides e.g. lambda-cyhalothrin and imidacloprid. In Chapter 2, the use of constructed wetlands for the removal of pesticides e.g. pyrethroids such as lambda-cyhalothrin, is partially highlighted, while information on imidacloprid removal in CW is scarce. The mesocosm experiments resulted in important data such as: pesticide half-life times and amounts of pesticides in different environmental compartments. However, research on a larger scale is necessary to validate results obtained for mesocosm experiments. These experiments may contain intrinsic artifacts which may confound extrapolation of results to conditions in larger scale systems (Ahn and Mitsch 2002). The main goal of this research was to investigate the removal/retention of two insecticides e.g. Karatox (a.i. lambda-cyhalothrin) and Imidox (a.i. imidacloprid) in a vegetated field scale surface flow constructed wetland. This research aimed to investigate the removal of these two insecticides with a monoculture of *Nymphaea amazonum* or Water Lily (executed during phase 1) and with multiple types of plants (e.g. *Nymphaea amazonum* (Water lily) and *Echinochloa polystachya* (Aleman grass)), executed during phase 2. In addition, batch sorption experiments were carried out to assess sorption kinetics and sorption isotherms. Batch kinetic sorption experiments are essential in describing the non-equilibrium phase of pesticide sorption on 3 soils from the field scale wetland area, while sorption isotherms provided data of sorption partition or distribution coefficients.

7.2 Material and methods

7.2.1 Design of the field scale wetland

In chapter 6, a calculation is presented for the design of a field scale constructed wetland for the treatment of runoff containing the pesticides chlorothalonil, lambda-cyhalothrin and imidacloprid using data obtained from the *P. australis* mesocosms study. For the design of a field scale wetland for the treatment of runoff with lambda-cyhalothrin and imidacloprid, the formulas used in chapter 6 were applied. Instead of a 90% reduction from previous study, a minimal reduction of 50% and a depth of 1 m were used for calculations. The reaction rate constant was derived from mesocosms experiments in Suriname, instead of those from Belgium (chapter 6). Reasons for that were the fact that the field scale wetland study is planned with multiple plant types; results derived from the *P. australis* mesocosm studies do not cover that; higher reaction rates were found in Suriname and the flow used is the actual inflow of the field scale wetland.

The total water flow (Q expressed in m^3/s) entering the wetland was derived by making use of Formula 7-1 based on information provided in the flow meter user manual.

$$v = (F\ x\ 0.000845) + 0.05 \qquad \textit{(Formula 7-1)}$$

In which:

v: *velocity (m/s)*

F: *average flow counts (F) were 259 (n=5), (derived from field measurements*)

From Formula 7-1, the calculated velocity (v) is 0.269 m/s. The calculated water flow (Q in m^3/s) is derived from Formula 7-2.

$$Q = v \; x \; L \; x \; W \qquad \textit{(Formula 7-2)}$$

In which:

Q: *water flow in m^3/s*
v: *velocity in m/s*
L: *length of the narrowing of the ditch in m (0.1 m)*
W: *width of the narrowing of the ditch in m (0.1 m)*

From Formula 7-2, Q becomes 0.00269 m^3/s. The average time for irrigation is 2 h per day. For two hours of irrigation, the total discharge can be calculated by means of Formula 7-3.

$$Qdischarge = Q \; x \; h \; x \; c \qquad \textit{(Formula 7-3)}$$

In which:

Qdischarge (m^3/d): *the total discharge taking the hours of irrigation into account.*
h: *the average time (hours) for irrigation on a daily basis*
c: *conversion factor from seconds to hours (3600)*

Based on Formula 7-3, Qdischarge (m^3/h) is 19.4 m^3 per 2 h per day, which equals 19.4 m^3 per day.

The reaction rate constant (k) of 0.228 $days^{-1}$ was used in present study and was based on mesocosm experiments described in chapter 5 for imidacloprid (0.0095 h^{-1}). Taking into account the pesticide with the highest DT_{50} (imidacloprid) and making use of the formulas for designing a wetland, described in chapter 6, the calculated wetland area became 118 m^2, with a HRT of 3 days. In order for sufficient removal of pollutants to take place in surface flow constructed wetlands, these systems should be designed to retain water for 3 to 3.5 days (Vymazal 2010). The practical dimension of the wetland was: Length (25 m) x Width (5 m) x Depth (1 m). For the actual construction it was decided to increase the length of the wetland to 50 m (section 7.3), based on the results in Moore et al. 2001, in which study a ditch length of 50 m was proposed for reduction of lambda-cyhalothrin to a no effect level of ≤ 0.02 µg/l aqueous concentrations. In several other studies, (Bennett et al. 2005; Kröger et al. 2009; Moore et al. 2009A) the ditch length was accessed as a tool to provide an adequate reduction of lambda-cyhalothrin. For the wetland porosity, ε = 1.0 was used, which is according to Kadlec and Wallace (2009) a good approximation.

In June 2012, a field scale constructed wetland (Figure 7-1) with dimensions 50 m (L) x 5 m (W) x 1 m (H) was built with the help of a local contractor and field officers from the Ministry of Agriculture in the Alkmaar area (district Commewijne (location represented by the green star in Figure 4-2) by adjusting the existing landscape (drainage canal). This canal receives water from several small ditches and is drained into the nearby Commewijne River. To regulate the water reaching the inlet, a dam consisting of clay; was built (Figure 7-1, dam 1), followed by a second one (dam 2) at a distance of 6 m from the first. This area is in connection with two drainage ditches (1 and 2). These ditches drain water from an agricultural field (area 459 m^2). By means of a handmade trench (40 cm (L) x 15 cm (W) x 30 cm (H)) in dam 2, water from the inflow was transferred to the wetland (point V1, "start" of the wetland or inlet). A third dam was built 25 m from dam 1, and the outflow was placed there by means of a PVC tube. Another tube functioned as overflow tube; in case the water level would increase due to heavy rainfall. To apply a simulated rainfall; the existing water reservoir upstream from the proposed wetland location, was excavated to twice its depth. All other drainage ditches besides 1 and 2 and streams flowing to the wetland area, were disconnected. To facilitate flow measurements, the outflow of drainage ditches 1 and 2 was adjusted by narrowing it to a length of 10 cm and a width of 10 cm. The planted area between drainage ditch 1 and ditch 2 was cut down to a length of a few centimetres.

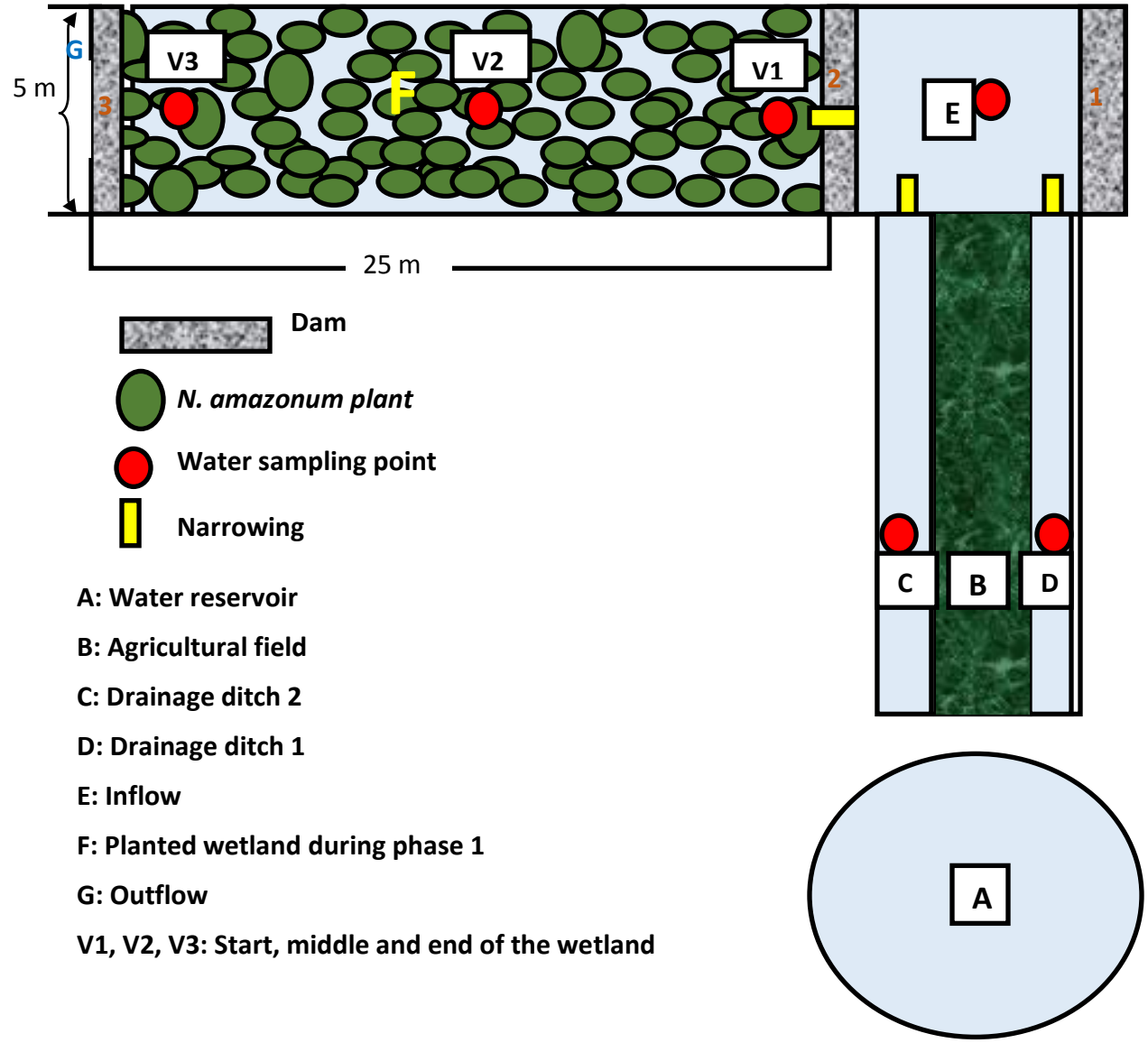

Figure 7-1: Schematic view of constructed surface flow wetland located at Alkmaar, Commewijne. Point V1 "start" of the wetland indicates the inlet, while point V3 "end of the wetland" represents the measuring point near the outlet

Figure 7-2: Images of the surface flow wetland after the acclimatisation period with Nymphaea amazonum plants (phase 1) and with multiple plant types (Nymphaea amazonum and Echinochloa polystachya plants (phase 2))

7.2.2 Vegetation establishment

Before planting, water from the wetland was drained out with a 3-inch water pump. A total of 67 plants of *Nymphaea amazonum* were harvested from a nearby sump upstream of the wetland and planted in the wetlands by hand (plant density was around 4 plants per m^2), followed by an acclimatization period of 6 weeks (Kadlec and Wallace 2009). The average temperature in the sump was 32.9 ˚C, the pH 6.67, S 0.174 ppt, the EC_{cond} 0.315 µS and the TDS 0.261 g/l. The experiments were conducted in 2 phases, phase 1 with a monoculture of *Nymphaea amazonum* and phase 2, with multiple plant types (*Nymphaea amazonum* and *Echinochloa polystachya).* Phase 1 was executed from November 20, 2012 up to December 3, 2012, while phase 2 was executed from March 5, 2013 up to March 18, 2013. Each phase consisted of 14 days, from which 9 days were used for field measurements. Before the start of phase 2, 78 *Echinochloa polystachya* plants were harvested from an upstream ditch nearby (T 31.8 ˚C; pH 7.01; S 0.187 ppt, EC_{cond} 0.326 µS and TDS 0.288 g/l) and planted in the wetland according to the same procedure as for the *Nymphaea amazonum* plants. Figure 7-2 gives a view of the wetland at the start of both phases.

7.2.3 Pesticide application

On day 1, of each phase, a pesticide mix was applied, based on high application rates of farmers in the research area and the frequency of pesticide use. In a spraying tank, 550 ml of imidacloprid (Imidox 20% SL, purchased at H.J. De Vries Agro) and 250 ml of lambda-cyhalothrin

(Karate 2.5 EC purchased at Agrimex N.V.) was mixed with water, to a total volume of 25 litres and applied to the agricultural field (459 m^2) resembling an application rate of respectively 2.4 kg/ha and 0.14 kg/ha. Application of pesticides was done by means of a hand spray. The different types of samples and the time during which they were taken are presented in Table 7-1.

7.2.4 Precipitation measurements

The amount of rainfall was measured daily by means of a rain gauge which was attached to a piece of wood stick that was placed in the open field adjacent to the agricultural field.

7.2.4.1 Simulated rainfall

A simulated rainfall was performed by means of a water pump (2 inch) in case the rainfall was less than 10 mm/day. A minimum value of 10 mm/day is needed for runoff to occur. The water used for this purpose, was taken from the water reservoir (Figure 7-1). On day 1, the agricultural field was irrigated for 30 up to 40 minutes immediately after pesticide addition. This time and the amount of rainfall needed, was derived from earlier field measurements (data not shown). During very dry periods (phase 2), this time was increased to 1½ and 1¾ h, allowing the possibility to sample the water and to measure the general water quality parameters directly in the different locations.

7.2.4.2 Water level measurements

The water level in the drainage ditches was measured twice (before and after the simulated rainfall) on a daily basis by making use of a graduated piece of wood mounted in the soil. The measurements were done in 10 measuring points e.g. in the “start” or near the inlet (V1), the middle (V2) and in the end or near the outlet (V3) of the wetland and in the inlet, midpoint and outlet of both drainage ditches.

7.2.5 Water quality measurements

The water quality was determined by daily measurements of the following parameters; pH, EC_{cond}, S, TDS and T, by means of a field meter (brand Extech). The water quality measurements were done in ditch 1 and 2, the inlet, the “start”, middle and end of the wetland by placing the electrode directly in the water and by means of reading the scale. For sampling within the wetland, (the inlet, the “start”, middle and end of the wetland) this was not feasible and water from these locations was first transferred to a water bucket (4 liters capacity); the electrode was placed in the bucket, followed by direct measurements with the field meters.

7.2.6 Flow measurements

The water velocity (v) was measured immediately after the simulated rainfall and in 3 points: narrowing ditch 1, narrowing ditch 2 and narrowing between the inflow and the point near the inlet of the wetland (V1) by means of a similar procedure as described in section 6.2.5.4.

7.2.7 Water, plant and sediment sampling

For the determination of the amount of lambda-cyhalothrin and imidacloprid in water, composite samples were prepared at different time-intervals in 6 measuring points (Figure 7-1 and Table 7-1); ditch 1, ditch 2, inflow wetland, near the inlet (V1), middle (V2) and near the outlet (V3) of the wetland. Water was sampled by means of a water bucket (ditch 1 and 2) and in the wetland by means of an uncapped 1-L Nalgene bottle by collecting water at four locations within the vicinity (radius of 0.5 m) of each measuring point and combining them to achieve a representative sample. Before pesticide addition, blanks were prepared in a similar manner, to determine if pesticides were already present and in which concentrations. Similar procedures were executed for the sediment and plant samples. Plant samples (3 plants per location) were harvested near the inlet (V1), middle (V2) and near the outlet of the wetland (V3) and roots separated from the plant upper part for further analyses. Sediment samples were collected in the following points: B (planted area), ditch 1 and 2, inflow, V1, V2 and V3 by combining four subsamples (top 3 cm layer) to receive a composite sample (1 kg) of each location.

7.2.8 Calculations

7.2.8.1 Amount of pesticides in ditches and runoff

The amount of pesticides in ditches 1 and 2 was calculated by multiplying the concentration of pesticides in water (at time x h) with the total volume present in the ditch. The total volume was determined by multiplying the water level (at time x h) with the area of the ditch (14 m (L) x 1.4 m (W)), assuming a homogeneous concentration throughout the ditch. The amount of pesticides in runoff was determined by multiplying the total amount of pesticides in the ditches with a conversion factor. This factor is the ratio: $\frac{\text{volume increase after simulated rainfall}}{total\ volume\ ditch}$. This amount is further expressed in percentage of the total amount of pesticide added to the field (see appendix 7, Tables g-4 up to g-11).

7.2.8.2 Removal efficiencies

Two types of removal efficiencies were investigated in this wetland study (1) temporal and 2) spatial); 1) the removal in one measuring point over a specific time-interval (e.g. 2 h-312 h) and 2) the removal over distance between two measuring points (inlet versus outlet) in the end of the experiment (312 h). The Formula used to calculate the removal in one measuring point is similar as the Formula used in chapter 5, Formula 5-1. With $C_{0,}$ representing the concentration at time 0 h and C_x the concentration at time x h for a specific sampling location. For the overall efficiency, the inlet concentration was compared with the outlet concentration and substitutes respectively C_0 and C_x.

7.2.9 Statistical analysis

The following statistical tests using Sigma Stat. 2.03 were executed:

1) A two way ANOVA to assess; a) significant differences between inlet and outlet concentrations of phase 1 and phase 2 and b) significant differences between the pesticide's spatial removal from water in phase 1 and phase 2.
   The test was performed, to assess significant differences of pesticide removal with 1 type (phase 1) and 2 types of plants (phase 2). To perform these experiments, datasets were checked for fulfilment of the assumptions for normality and equal variance. A confidence level of 95% was used and a p-value of 0.05.
2) A Mann-Whitney Rank Sum Test (Because data of pesticide uptake by plants was not normally distributed) to determine significant differences in plant uptake (shoots and leaves versus roots) in phase 1.
3) A Three Way ANOVA to assess significant differences between the dependent variable amount (µg) of pesticide/g DW plant (phase 2) and the sources of variation (time, location in wetland and plant part).

7.2.10 Sampling scheme for water, sediment, plants and water quality (WQ) parameters

The sampling schemes in phase 1 (November 20, 2012-December 3, 2012) and phase 2 (March 5, 2013-March 18, 2013) are presented in Table 7-1.

Table 7-1: Scheme of water, sediment (Sed) and plant sampling for pesticides (lambda-cyhalothrin and imidacloprid abbreviated as lam and imi) residue analysis and for the measurement of the general water quality (WQ) parameters during phase 1 and phase 2 of the field wetland study

Time (hours)	Water lam	Water imi	Plant lam	Plant imi	Sed lam	Sed imi	WQ par.
0[a]	x[c]	x	x	x	x	x	x
2[b]	x	x	x	x	x	x	x
24	x	x	-	-	-	-	x
48	-[d]	x	x	x	x	x	x
72	-	x	-	-	-	-	x
96	x	x	x	x	x	x	x
144	-	x	x	x	x	x	x
192	-	x	x	x	x	x	x
240	-	x	-	-	-	-	x
312	x	x	x	x	x	x	x

[a] *Start of the experiment, water samples (blanks) taken prior to pesticides application to the field*
[b] *2 h after pesticide application to the field;*
[c] *Sampled*
[d] *Not sampled*

7.2.11 Sorption experiments

The main objective of the sorption experiments was to assess sorption kinetics and sorption isotherms. Results obtained for mesocosm experiments (e.g. the increase in imidacloprid concentration especially in batch 5, at the start of the experiment (chapter 5)) indicated possible remobilization or desorption of pesticide to be occurring. Therefore batch kinetic sorption experiments were used to study possible desorption of pesticides. These kinetic experiments describe the non-equilibrium phase of pesticides (lambda-cyhalothrin and imidacloprid) sorption on 3 different soils (S1, S2, and S3) in the vicinity of the field scale wetland in Alkmaar, Commewijne. Sorption isotherms provided data of sorption at equilibrium, allowing to assess the partitioning or distribution coefficients *(*K_d value) at equilibrium for each pesticide-soil combination. Soil S1 was collected from the inlet (Figure 7-1, inlet); S2 was collected from ditch 2, and S3, from the planted area or the agricultural field (point B, Figure 7-1). Pesticide sorption on 3 different soils was studied using the batch equilibrium technique according to the OECD guideline106. Different tests (results not shown) were performed to derive the solid (g air dried soil) to liquid (ml of calcium chloride ($CaCl_2$) solution ratio suitable for sorption studies.

7.2.11.1 Sorption isotherm experiments for lambda-cyhalothrin

The experiment was performed in duplicate as follows: A concentration series of 0, 2, 4, 8 and 16 mg/l was made by pipetting 0, 0.5, 1, 2 and 4 ml of 100 mg/l of lambda-cyhalothin stock solution (Dr. Ehrenstorfer GmbH) in centrifuge tubes, followed by the addition of 2.5 ml, 2g/l

sodium azide (NaN3, Sigma-Aldrich, St-Louis, MO, USA) solution. The calcium chloride solution was used as a background electrolyte to simulate an ionic strength similar to that of a natural soil solution, while sodium azide solution was added to minimize biological activity. Each tube contained 0.125 grams of air dried soil. The flasks were filled up to a total of 25 ml with calcium chloride solution (Merck, Darmstadt, Germany), 0.01 M. Two blanks were performed without the addition of soil. The obtained mixtures were placed for 24 h in the dark on an orbital shaker (type SM 30 B Control provided by Edmund Buhler GmbH) at 150 rpm (rates per minutes) at room temperature, followed by separation of the soil from the water phase by means of a centrifuge (Heraeus) and filtration over an ash less paper filter (Whatman 589/1, pore size: 12-25 µm). Batch sorption experiments were performed to determine the amount of sorption of pesticides on the filter paper used. No significant sorption on filter paper was observed (data not shown). From the filtrate, 10 ml was pipetted, extracted with 2 times 25 ml ethyl acetate (VWR, Leuven, Belgium). The organic layer was evaporated until almost dry in a Heidolph Laborota 4000 Rotary Evaporator at 40°C and re-dissolved in 3 ml hexane (VWR, Leuven, Belgium) and analyzed by GCMS.

Because of the lack of sufficient soil (S3) and negative results (no detections in the water phase after equilibrium after different tests (results not shown) using the OECD guideline 106), experiments with S3 were repeated using the method described hereunder. The experiment was performed in duplicate as follows. A concentration series of 0.01, 0.05, 0.5, 1 and 10 mg/l was made by pipetting respectively 5, 25, 250, 500 and 5000 µl of lambda-cyhalothrin 100 mg/l in centrifuge tubes, followed by the addition of 5 ml NaN_3 solution (2 g/l). Each tube contained 0.5 grams of air-dried soil. The flasks were filled up to a total of 50 ml with calcium chloride solution of 0.01 M. Two blanks were performed without the addition of soil. These solutions were placed on an orbital shaker for 24 h. After this, they were centrifuged for 20 min at 13,000 rpm. From the supernatant, 10 ml was pipetted and extracted with 2 times 25 ml ethyl acetate and the organic layer was then evaporated until dryness and re-dissolved in 2 ml hexane and analyzed by GCMS.

7.2.11.2 Sorption isotherms of imidacloprid

The experiment was performed in duplicate as follows. A concentration series of 0, 4, 6, 8 and 10 mg/l was made by pipetting 0, 0.4, 0.6, 0.8 and 1.0 ml of 100 mg/l of imidacloprid stock solution (Dr. Ehrenstorfer GmbH) in centrifuge tubes, followed by the addition of 1 ml sodium azide solution (2g/l). Each tube contained 2 grams of air-dried soil. The flasks were filled up to a total volume of 10 ml with calcium chloride solution 0.01 M. The tubes were sealed and agitated for 24 h on an orbital shaker at 150 rpm at 25 °C. After equilibrium the mixtures were centrifuged for 20 min at 13,000 rpm and the filtrate was filtered through a 0.2 um syringe filter and injected into the HPLC.

7.2.11.3 Sorption kinetic experiments for lambda-cyhalothrin

In a centrifuge tube, 0.1 g air-dried soil (S1, S2, and S3) was added to 5 ml 100 mg/l lambda-cyhalothrin stock solution, 5 ml sodium azide solution (2 g/l) and the final volume was brought to 50 ml with calcium chloride solution of 0.01 M. A blank without soil was also carried out. To study sorption kinetics, suspensions were filtered over an ash less paper filter (Whatman 589/1, pore size: 12-25 µm) after 2, 6, 17, and 25 h of placement on an orbital shaker. Prior to this, the mixtures were centrifuged for 20 min at 13,000 rpm. After filtration, 10 ml of the supernatant was extracted with 2 times 25 ml of ethyl acetate. After evaporation to near dryness, the residue was re-dissolved in 3 ml hexane and concentrations were determined by means of GCMS.

7.2.11.4 Sorption kinetic experiments for imidacloprid

In a centrifuge tube, 5 grams of air dried soil, 0,5 ml of imidacloprid stock solution (10 mg/l) was added and 1 ml of sodium azide solution (2 g/l). The total volume was brought up to 10 ml with calcium chloride 0, 01 M. A blank was carried out without the addition of soil. Experiments were conducted in duplicate and mixtures were placed on an orbital shaker for 0, 6, 24 and 72 h. After reaching equilibrium, the mixtures were centrifuged for 20 min at 13,000 rpm and the supernatant was filtered through a 0.2 µm syringe filter and injected into the HPLC to determine the residual pesticide concentration.

7.3 Results and discussion

7.3.1 Physical-chemical parameters

The results for the physico-chemical parameters are presented in Table 7-2.

Table 7-2: Range[a] of physical-chemical water quality parameters for the 6 measuring points

Phase		pH	Temperature (◦C)	TDS (g/l)	Salinity (ppm)	Conductivity (µS)
Phase (n=66[b])	1	7.4-9.9	25.5- 34.8	202-577	101- 486	202-577
Phase (n=66)	2	7.5-9.8	29.0-32.3	300-591	226-499	309-588

[a] *Measured values for water quality parameters presented from the start up to the end of the experiment*
[b] *For each water quality parameter a total of 66 measurements were evaluated*

All water quality parameters increased in value over time. This was observed in all measuring points. This might have been caused by the photosynthetic activity of the vegetation in the wetland combined with intense sunlight (Ahn and Mitsch 2000; Maltby and Barker 2009).

7.3.2 Flow measurements

The results for the flow measurements (expressed in m^3/d) are presented in Table 7-3.

Table 7-3: Measured flow (m^3/d) in a surface flow constructed wetland under N.amazonum only (phase 1) and N.amazonum + E. polystachya (phase 2) conditions

	Phase 1			**Phase 2**		
Time (h)	**Ditch 1 to inflow**	**Ditch 2 to inflow**	**Inflow to wetland**	**Ditch 1 to inflow**	**Ditch 2 to inflow**	**Inflow to wetland**
0	4.76	6.56	44.6	12.16	12.32	45
2	8.64	6.2	35.6	18.72	11.02	38.2
24	N.M.[a]	N.M.	N.M.	19.8	14.62	72
48	8.72	11.02	48.2	24	18	72
72	8.92	9.64	50.6	21.8	25.6	82.8
144	9.28	10.08	50	21.8	25	85.6
192	9.44	10.66	50.6	21.6	18.44	69.8
312	9.44	9.8	50.6	N.M.	N.M.	N.M.

[a] *not measured*

The water flow varies between 4.8-50.6 m^3/d in phase 1 and between 12.2-86.0 m^3/d in phase 2. The values in phase 2 were in all sampling points higher than in phase 1, because more water had to be used for irrigation purposes. Phase 2 was executed in the small dry season, which later gave rise to a dry field. The measured values for the flow are in agreement with the inflow rates (15.8-21.5 m^3/d) mentioned in the design manual of Stantec Consulting Ltd (1999) for CW for rural applications in Ontario, Canada for a SFCW of 33 m (L) x 33 m (W) and a depth of 0.1 m.

According to Budd et al. (2009), pesticide removal efficiencies have shown to decrease quickly with increasing flow through wetlands. Decreased flow, increases retention time and water/macrophyte contact in agricultural drainage systems, and removes suspended solids from the water column (Bouldin et al. 2005). In Stearman et al. 2003, 14 small constructed wetlands cells (area 6-12 m^2) collecting runoff from a 465 m^2 gravel bed nursery were loaded at 3 flow rates of 0.240, 0.120, and 0.060 m^3/d. At the lowest flow rate, which corresponds to lower mass loadings and greater hydraulic retention times (HRTs), a greater percentage of pesticides was removed (90.2% metolachlor and 83% simazine) compared to the higher flow rate (82.4% metolachlor and 77.1% simazine).

7.3.3 Pesticide concentrations measured in water

7.3.3.1 Lambda-cyhalothrin

The lambda-cyhalothrin concentrations in all 6 water-sampling points (Table 7-4) were below the limit of detection (0.001 µg/l) between timeframe 0 h-312 h in phase 1. In phase 2, most of the measured concentrations were also below the limit of detection, with a few exceptions in the sampling points, ditch 1 and ditch 2 (timeframe 2 h-48 h), and V3 (near the outlet, timeframe 192 h-312 h). The percentage of pesticide found in runoff (phase 2) was 0.6% (time 2h) and 0.2% (time 48 h) in ditch 1, while in ditch 2 these values were respectively 0% and 0.1%. In Bouldin et al. (2005), 33 of the 56 measured lambda-cyhalothrin concentrations in water of wetland's microcosms were below the limit of detection (0.001 µg/L). The physico-chemical properties of lambda-cyhalothrin e.g. low solubility and a high K_{oc} value determine its behaviour in water. The high K_{oc} value indicates a high affinity to the soil (Hand et al. 2001). Lambda-cyhalothrin is a hydrophobic pesticide and comparable results for its removal/retention such as a very fast dissipation from the water phase, accumulation in wetland plants and adsorption to soil/substrate are observed in other studies (Chapman et al. 1981; Moore et al. 2001; Lee et al. 2004; Bennett et al. 2005; Moore et al. 2009A). Other factors of importance, especially for field studies, are the time of application of the pesticide and the rainfall, especially intense rainfall (Jergentz et al. 2005). The shorter this time difference, the higher the amount of pesticides that ends up in agricultural runoff.

Table 7-4: Measured lambda-cyhalothrin concentrations in 6 water sampling points in a surface flow constructed wetland in phase 2, under N. amazonum and E. polystachya conditions

Time (h)	Ditch 1 mg/l	Ditch 2 mg/l	Inflow mg/l	Inlet (V1) mg/l	Midpoint (V2) mg/l	Outlet (V3) mg/l
0	ND[a]	ND	ND	ND	ND	ND
2	0,0934	ND	ND	ND	ND	0,0149
24	0,0163	0,0069	0,0078	ND	ND	ND
48	0,0283	0,0156	0	0,0032	ND	ND
144	ND	ND	ND	ND	ND	ND
192	ND	ND	ND	ND	0,0022	ND
312	ND	ND	ND	ND	0,0420	ND

[a]*Not Detected (LOD 0.001 µg/l)*

For both phase 1 and 2, pesticide measurements were conducted immediately after a simulated rainfall. All concentrations in phase 1 were below the LOD, while in phase 2 detections were done in both drainage ditches (see Table 7-4). This might be caused by desorption or remobilization (Passeport et al. 2013; Vallee et al. 2014) of lambda-cyhalothrin accumulated in the sediment from the ditches during phase 1 and the longer duration of the simulated rainfall during phase 2, allowing the flushing of the pesticide from the agricultural field. In Moore et al. (2013), between 1.0-2.0% of the initial pyrethroid (permethrin) amounts applied, were released after flushing performed 48-51 h after the start of the experiment. This

is higher than the amount of lambda-cyhalothrin released (between 0.3-0.6%) after application of a simulated rainfall performed between 2 h-48 h after the start of the experiment in phase 2 of present study. This is expected, because the solubility of permethrin is around 40 times higher than that of lambda-cyhalothrin (PPDB 2016). Lambda-cyhalothrin's detection in the water phase is also related to the amount applied and the way of applying this e.g. directly in the mesocosms or applying it firstly to the farmland, followed by a simulated runoff. In the second mesocosm experiment executed in Belgium (chapter 6), mesocosms were directly amended with a high lambda-cyhalothrin's concentration (500 µg/l). This resulted in longer detections up to 334 h in the water phase, while for the study described in chapter 5, (mesocosms amended with much lower concentrations of 10 and 30 µg/l), lambda-cyhalothrin was detected only up to 48 h.

A study comparable to present study is presented in CDPR (2008). In that study lambda-cyhalothrin was first applied to a 0.40 ha alfalfa field by means of an aerial application of 0.14 kg/ha. After a 4 days irrigation regime, water was sampled and lambda-cyhalothrin analyzed. The application rate was the same as in present study, but comprised of an aerial application (possibility of spray drift) and involved a return ditch, which was not present in current study. Lambda-cyhalothrin concentrations ranged between 0.018-0.077 µg/l in runoff, while in present study these values were much higher (2.2-93.4 µg/l), This is possible, because the agricultural field was around 9 times smaller and droplet drift occurred during application. The highest concentration was measured at time 2 h in ditch 1 (see Table 7-4). In addition, the range of lambda-cyhalothrin concentrations (0.71-3.8 µg/l) measured in small ditches near the farmland (chapter 4) is much lower. That is expected, because in present study a much higher dose (0.14 kg/ha) was applied, than the recommended field dose used by farmers in the research area (0.01-0.02 kg/ha).

7.3.3.2 Imidacloprid

Figure 7-3 presents the results for the dissipation of imidacloprid in the 6 measuring points during phase 1 and 2.

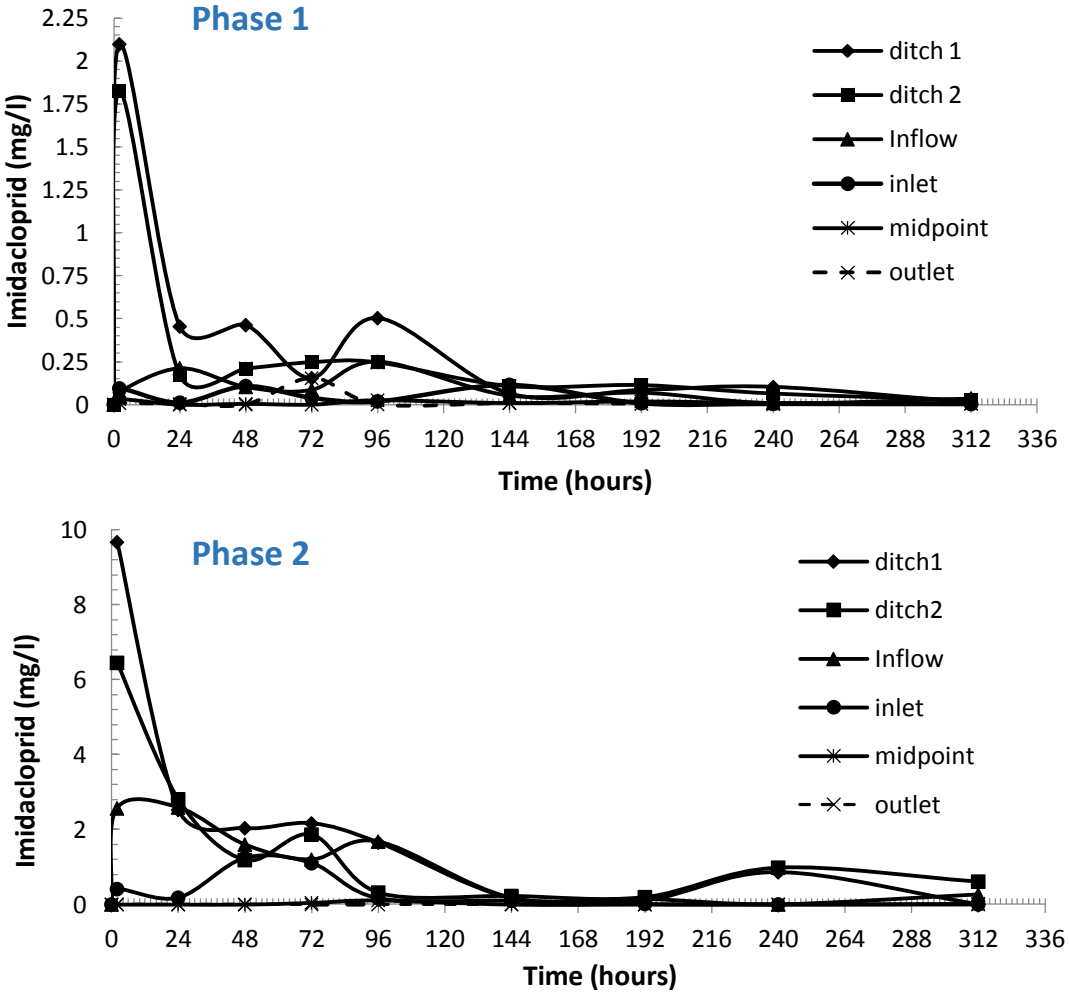

Figure 7-3: Imidacloprid aqueous concentrations in a field scale constructed wetland under N.amazonum only (phase 1) and N.amazonum + E. polystachya (phase 2) conditions

Results for phase 1, show that at time 2 h after pesticide addition, imidacloprid was detected in all 6 measuring points, with the highest concentrations in ditch 1 (2.10 mg/l) and ditch 2 (1.82 mg/l) and the lowest concentration in the outlet of the wetland (0.0085 mg/l). At the end of the experiment, concentrations were reduced up to 99.5% (ditch 1), 98.5 (ditch 2), 52.0% (inflow), 96.3% (inlet or "start" of the wetland), 78.7% (middle of wetland) and 47.1% at the end of the wetland (near the outlet). Result obtained for phase 2, showed the same pattern for the 4 measuring points i.e. ditch 1 and 2, inflow and inlet. However, at the midpoint and the outlet, no imidacloprid was detected at time 2 h and 312 h. For ditch 1, the measured concentration of imidacloprid was 9.68 mg/l at time 2 h and decreased with 99.1% at time 312 h. For ditch 2, these values were 6.45 mg/l (90.7% reduction), while for the inflow and inlet of the wetland, the measured concentrations were 2.57 and 0.41 mg/l and their respective reduction 89.8% and 95.4%. Another aspect is that in phase 2 more imidacloprid was detected in all points at time 2 h after application of rainfall. Results from the statistical analysis showed significant

differences between concentrations found in the inlet and outlet for both phases 1 and 2. (Table 7-5). The amount of imidacloprid in runoff was between 0.2-1.4% in phase 1 and 1.1-5.7% in phase 2 for the time interval 2 h-48 h, with the highest amount obtained 2 h after the start of the experiment (see Appendix 7, Tables g-4 up to g-10). Although, comparable amounts of pesticides were applied in the two phases, a longer irrigation time (3 to 4 times) applied during all water-sampling regimes in phase 2, was probably the cause of the higher amounts of pesticide found in runoff. The high solubility of imidacloprid also allows a good interaction with the water applied to the field. In the mesocosm study, described in chapter 5, complete removal of imidacloprid in a mesocosm planted with *Eleocharis mutata* and exposed to a target concentration of 60 µg/l imidacloprid was found at time 216 h, while exposing these mesocosms to higher imidacloprid concentrations (1000 µg/l) resulted in a lower removal. The mesocosm experiment described in chapter 6 shows that a 100% removal of imidacloprid from the water phase for vegetated (*P. australis*) and for non-vegetated mesocosms was obtained in batch 5 (time 240 h). This occurred after repeated exposure (in batches 1 up to 4) to the same concentration of pesticides (250 µg/l), while in the study described in chapter 5, mesocosms were exposed to increasing imidacloprid concentrations. The higher removal obtained in batch 5, indicates an enhanced biodegradation as described in Singh et al. (2003), Weaver et al. (2004), Rose et al. (2006), Budd et al. (2011).

A higher removal of imidacloprid in one measuring point was not observed in this field wetland during repeated exposure (phase 2) and with a combination of 2 plants. However, the results for the spatial or overall efficiency of the field scale wetland (Figure 7-4) show that compared to phase 1, the overall removal of imidacloprid in phase 2 was higher. In phase 1, the removal efficiency was 88.7% at time 2 h and that it increased up to 100% at time 48 h. Between time 144 h and the end of the experiment (312 h) the removal varied between 70.0-94.3%. In phase 2, a more constant pattern was observed for the dissipation of imidacloprid from the water phase compared to phase 1. Starting from time 24 h, the removal was already near 100%. This can be the result of the presence of different plant species (a combination of *Nymphaea amazonum* with *Echinocloa polystachya)*. The positive effect of vegetation on pesticide dissipation is also described in Elsaesser et al. (2011). In their study, peak concentrations (18-5904 ng/l) were reduced up to 91% in vegetated cells for a mixture of 4 pesticides (dimethoate, dicamba, triflozystrobin and tebuconazole), while for the non-vegetated cells the reduction was 72%. The vegetated cells were dominated by *P. arundinacea L.*, *T. latifolia L.* and *P. australis.* However, results of pesticide uptake by plants was low (up to 4%), with *P. arundinacea* cells being more effective than *T. latifolia* cells.

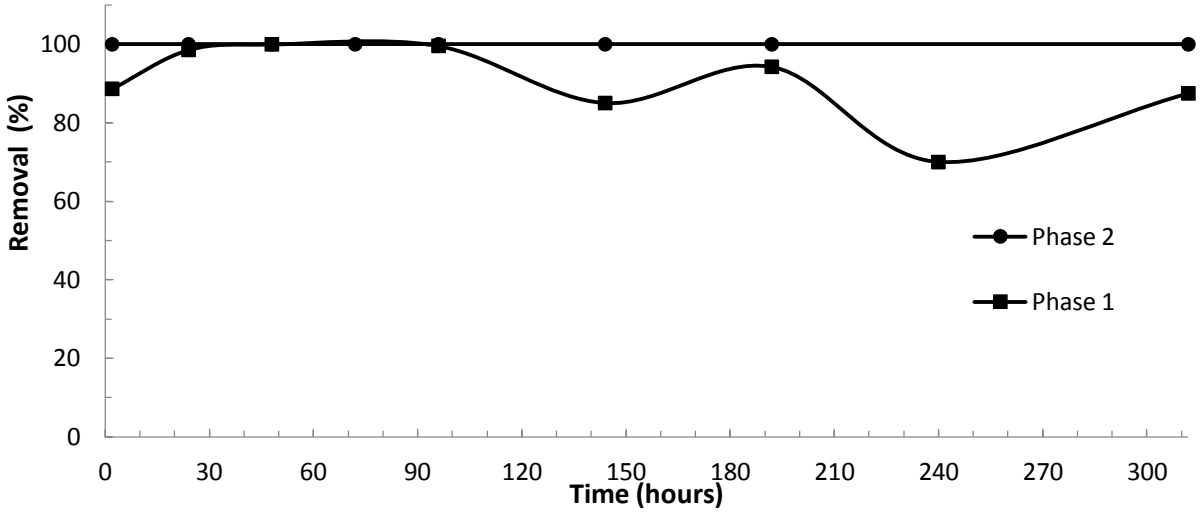

Figure 7-4: Spatial removal involving the comparison of inlet- with outlet concentrations of imidaclorpid from the water phase of a field scale constructed wetland under N. amazonum only (phase 1) and N. amazonum + E. polystachya (phase 2) conditions. Inlet concentrations were measured in V1 and outlet concentrations in V3

Table 7-5: Results of statistical analysis performed for a surface flow constructed wetland under N.amazonum only (phase 1) and N.amazonum + E. polystachya (phase 2) conditions

Experiment	Two-Way Anova Analysis	p-value at α=0.05 level	Significant differences
Phase 1	Inlet vs. outlet imidacloprid concentrations	0.043	Yes
Phase 2	Inlet vs. outlet imidacloprid concentrations	0.0462	Yes
Phase 1 and phase 2	Phase 1 vs. phase 2 (overall removal)	0.058	No

Although a relatively higher value was found for the spatial wetland efficiency during phase 2 (Figure 7-4), statistical analysis stated that this value was not significantly higher than the results obtained in phase 1. However, the p-value (0.058) did not differ much from 0.05, and indicates that the conditional probability of a type I error, given that the null hypothesis is true, is 5.8% instead of 5% (α=0.05).

7.3.4 Comparison of wetland effluent concentration with the EU WQ guidelines

Table 7-6 presents the wetland effluent concentration measured in phase 1 and 2 in comparison to the EU water regulation guideline. Concentrations in phase 1 are still higher than the annual average environmental quality standard (AA-EQS), while in phase 2 imidacloprid was not detected or below the limit of detection (0.5 µg/l). Taking into consideration that very high concentrations, detected near the inlet have been efficiently removed during both phases, and that the actual concentrations (between 0.5-3.9 µg/l) measured in drainage ditches of farmers

in the research area (see chapter 4) were much lower, it is expected that the guideline will be met.

In the study of Phong et al. (2009) imidacloprid was applied (application rate 10 kg/ha) as a commercial granular formulation (2%) to nursery boxes (2 with an area of 90 m^2) planted with 14-day-old rice seedlings. Immediately after pesticide application, the rice seedlings were transferred into paddy soil, which was ~2.5 cm below the soil surface. The highest imidacloprid concentrations measured in the paddy water 1 day after application of pesticide were between 58.6-73.9 µg/l and were reduced to < 1 µg/l after 14 days. Present experiment was also executed for 14 days (application rate 2.4 kg/ha) during each phase, but the minimum imidacloprid concentration during phase 1 was higher (4.5 µg/l). In Sanchez-Bayo and Goka (2006), 1 month was needed to reduce concentrations of 240 µg/l to 1 µg/l during a similar experiment as in Phong et al. (2009). Therefore instead of 14 days, a longer time might be required in present study (phase 1) to reach the environmental quality standard of 0.2 µg/l.

Table 7-6: Comparison of aquaous imidacloprid concentrations measured in the outlet of the field scale wetland with WQ guidelines, which are presented in green

Imidacloprid concentration in water	**(µg/l)**
Outlet concentration Phase 1 (wetland with *N. amazonum*)	4.5
Outlet concentration Phase 2 (wetland with *N. amazonum* and *E. polystachya)*	ND[b]
Annual Average Environmental Quality Standard (AA-EQS)[a]	**0.067**
Maximum Allowable Concentration-Environmental Quality Standard (MAC-EQS)[a]	**0.2**

[a] *Vijver and van den Brink (2014)*

[b] *Not Detected (LOD imidacloprid in water: 0.5 µg/l)*

7.3.5 Pesticide concentrations in shoots and leaves, roots and sediment of the wetland

7.3.5.1 Lambda-cyhalothrin in shoots and leaves and roots

Lambda-cyhalothrin was measured in the inlet (V1), midpoint (V2) and outlet (V3) of the wetland. The results are presented in Figure 7-5 and a detailed overview of the amounts detected is presented in Appendix 7, Table g-1. At time 2 h, lambda-cyhalothrin was not detected in shoots and leaves of *Nymphaea amazonum* in the inlet and outlet during phase 1 and phase 2. In the root part, detections were done only in the inlet. This was the case for both phase 1 and 2, with a 3 times higher detection during phase 2. The detection in *E. polystachya* was more in the plant upper part e.g. lambda-cyhalothrin was not detected in the roots of plants sampled in the inlet and the midpoint of the wetland. At time 48 h the highest concentration of lambda-cyhalothrin was detected in V1, in which increased amounts were found and detections done up to time 96 h. For the middle part (V2) of the wetland, lambda-cyhalothrin was only detected shortly after application (time 2h), while near the outlet (V3) only 1 detection was done at time 48 h.

In phase 1, the highest amount of lambda-cyhalothrin was found in V2, 2 h after application and in phase 2, at time 48 h in shoots and leaves of *Nymphaea amazonum.* For root samples, a

relatively higher uptake was measured in phase 2 compared to phase 1, indicating a possible adaptation towards the uptake of lambda-cyhalothrin by plants (combination of *Nymphaea amazonum* and *Echinocloa polystachya*). After statistical analysis no significant differences were found between the median values in roots of *N. amazonum* in phase 1 and phase 2 (a result 'Do Not Test' was found, indicating 'Not Significant')The highest amount of lambda-cyhalothrin was measured at time 312 h, in V3 of the wetland.

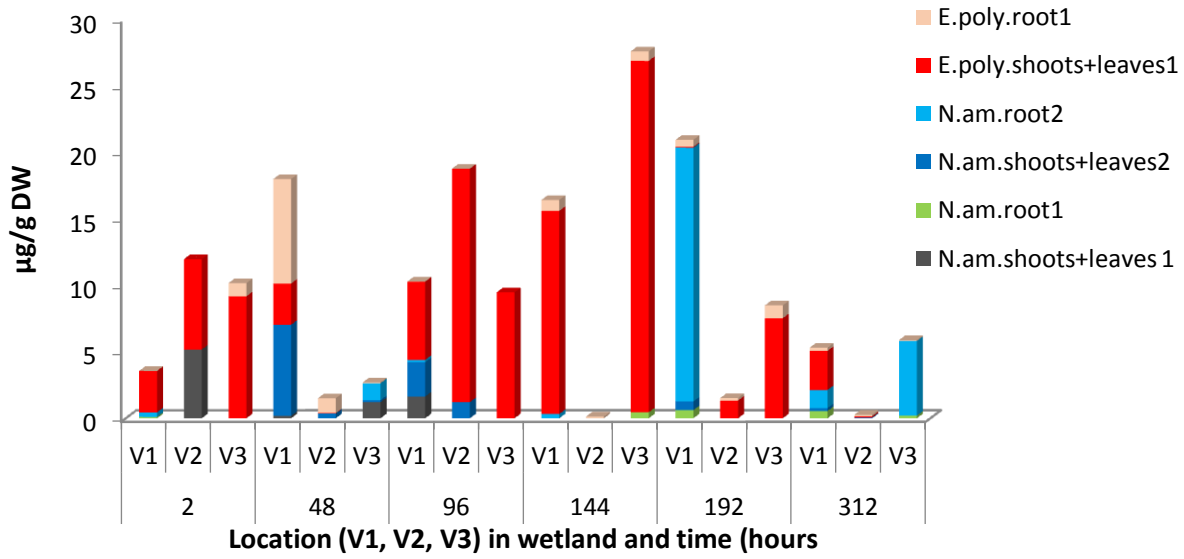

Figure 7-5: Lambda-cyhalothrin detected in N. amazoum (N. am.) and E. polystachya (E. poly.) shoots and leaves and root parts from different locations (V1 or 'start', V2 or "middle" and V3 or "end" of the wetland) in a field scale constructed wetland. N.am.root1 indicating uptake by the root part of N. amazonum in phase 1 and N. amazonumroot2, uptake by the root part of N. amazonum in phase 2

Compared to *Nymphaea amazonum* plants, lambda-cyhalothrin was detected two times more frequently in the *Echinocloa polystachya* plants. Amounts in plants increased from time 2 h up to time 144 h, while no detections were done for this timeframe, in the *Nymphaea amazonum* plants. The amount detected in roots was much lower in all measuring points except in the inlet, time 48 h and 192 h. The highest uptake was in shoots and leaves near the outlet (point V3) at time 144 h.
Statistical analysis (see appendix 4, statistical outputs for chapter 7) confirmed ($p<0.05$) the higher amounts of lambda-cyhalothrin in *E. polystachya* plants (plant part/shoots and leaves) compared to *N. amazonum* plants (both shoot and leaves and roots), while no significant differences (a result 'Do Not Test') were found between the uptake by roots of *E. polystachya* plants and those of *N. amazonum plants* in phase 2.

7.3.5.2 Imidacloprid in shoots and leaves and roots

The results for the measured imidacloprid concentrations in plants and root samples during phase 1 and 2 are presented in Figure 7-6 and a detailed overview is given in Appendix 7, Table

g-2. Results show a strong increase of the amount of imidacloprid in shoots and leaves from the inlet (V1) at time 2h up to time 312 h during phase 1. Increased amounts in roots were also observed during phase 1. The highest uptake of imidacloprid was at time 192 h in measuring point V2 in roots. During phase 2, this trend was not observed for plants and a more irregular pattern was found. In addition, less imidacloprid was detected in the roots of *Nymphaea amazonum* plants. Compared to *Nymphaea amazonum*, significantly more ($p<0.001$) imidacloprid was detected in the plants ($p<0.001$) and roots ($p=0.010$; see appendix 4, statistical outputs for chapter 7) of *Echinocloa polystachya* plants in all measuring points, indicating a higher uptake towards imidacloprid. The highest amount (1293 µg/g) was found in the inlet (V1) at the end of the experiment at time 312 h. In the mesocosm experiment (Chapter 5) the highest amount of imidacloprid in *N. amazonum* plants after 336 h was 2085 µg/g of which 82 % was attributed to the plant upper part. In present study the highest amount detected in *N. amazonum* was 1072 µg/g also in the plant upper part at time 48 h (phase 2). This lower value might be related to exposure of the wetland to full sunlight, which might have resulted in photolytic degradation of imidacloprid. Although these experiments were conducted under different conditions these values indicate a high uptake potential towards imidacloprid, while the uptake towards lambda-cyhalothrin in both phase 1 (5.16 µg/g at time 2 h) and phase 2 (26.4 µg/g at time 144 h; uptake by *E. polystachya*) was low.

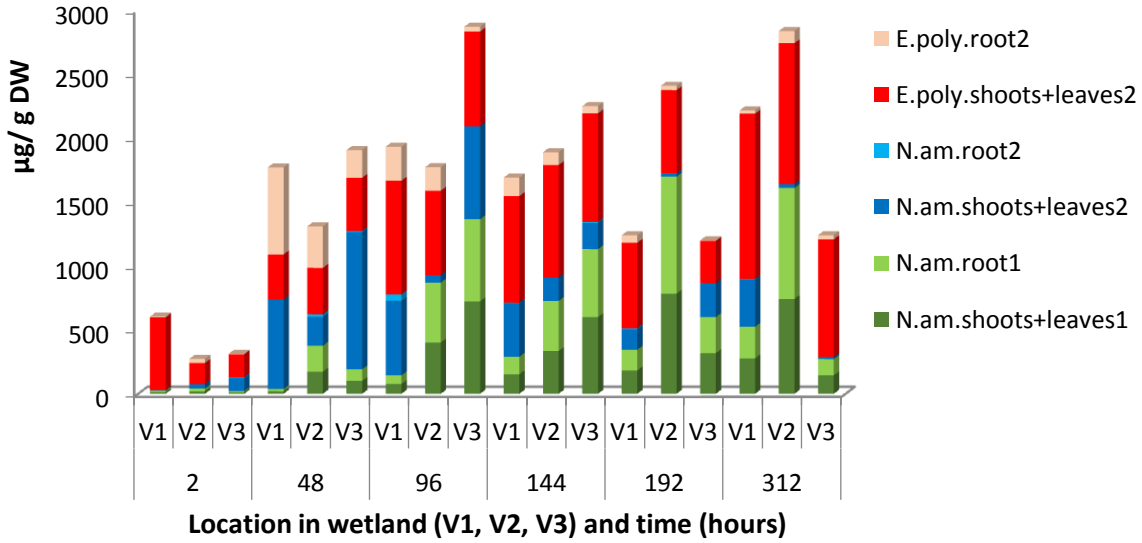

Figure 7-6: Imidacloprid detected in N.amazoum (N.am.) and E. polystachya (E.poly.) shoots and leaves and root parts from different locations (V1 or 'start', V2 or "middle" and V3 or "end" of the wetland) in a field scale constructed wetland. N.am.root1 indicates uptake by the root part of N. amazonum in phase 1 and N.am.root2, uptake by the root part of N. amazonum in phase 2

Significant differences were found for the uptake of the above plant part of *N. amazonum* compared to the root part of the same plant. In addition, more pesticide was taken up by the above plant part (shoots and leaves). The same trend was found for the *E. polystachya* plants. Significant differences were also found between the amounts detected in roots of both plants.

The average values (n=18) for imidacloprid uptake were 6.1 µg/g DW (roots of *N. amazonum*) and 128 µg/g DW (roots of *E. polystachya).*

7.3.5.3 Pesticides detected in sediment during phase 1 and phase 2

The pesticide concentrations measured in sediment samples during phase 1 and 2 are presented in Figure 7-7 and a detailed overview given in Appendix 7, Table g-3.

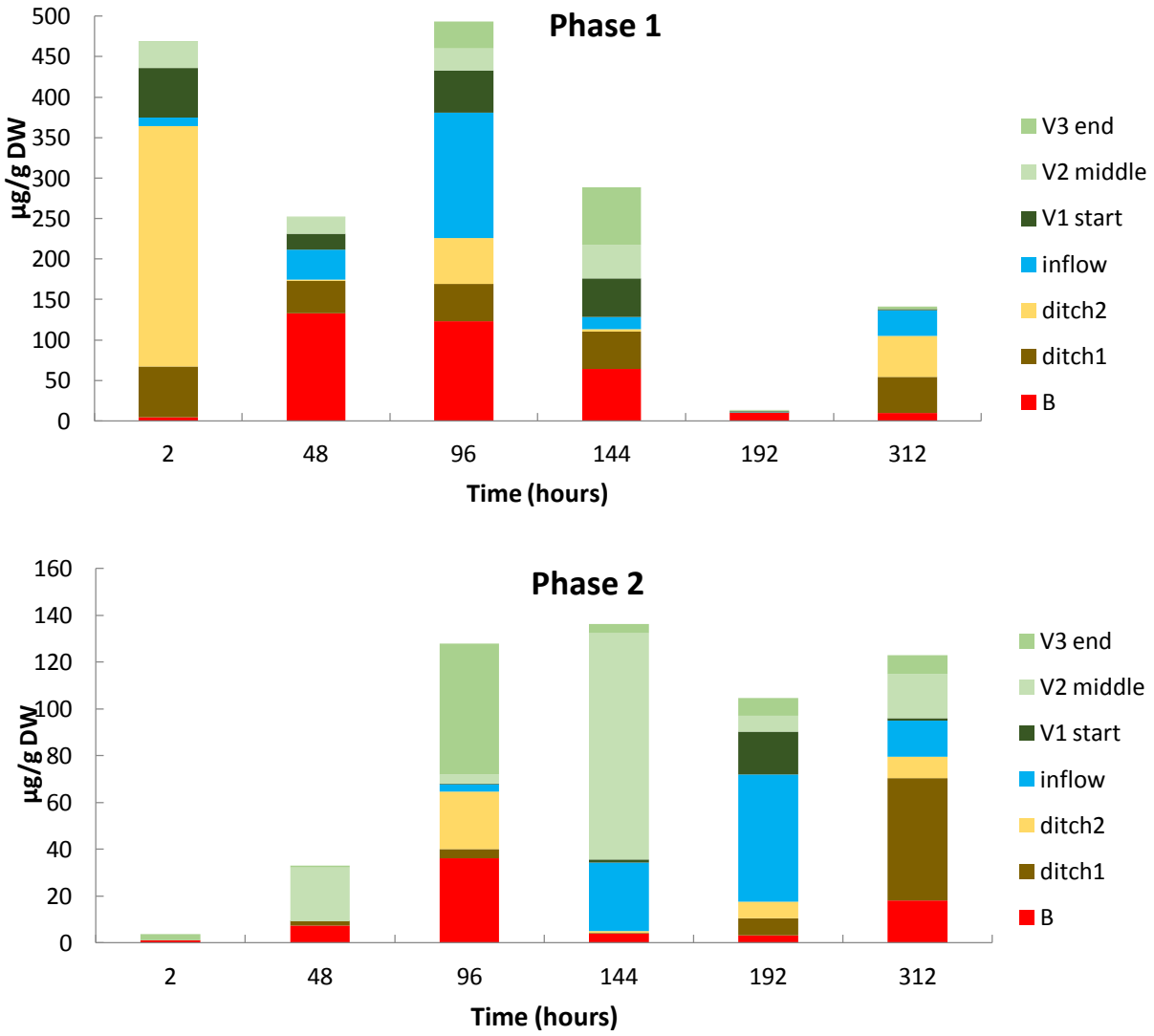

Figure 7-7: Amount of Lambda-cyhalothrin detected in sediment from different measuring points in a field scale constructed wetland under N.amazonum only (phase 1) and N.amazonum + E. polystachya (phase 2) conditions. Point V1 represents the "start" or inlet, V2 the "middle" and V3 "end" or outlet. B represents the agricultural field to which pesticide was applied

All 7 measuring points show that for the agricultural field (point B) relatively high concentrations of lambda-cyhalothrin were found 48 h after pesticide addition in phase 1. In the end of the experiment, these amounts declined considerably. This was expected because of the effects of rainfall and the repeated runoffs. With the exception of a few points, lower amounts of lambda-cyhalothrin were found in sediment samples during phase 2 compared to phase 1.

During phase 1, peak concentrations of lambda-cyhalothrin were found in ditch 2 at 2 h, in point B at 48 h and in the inflow at 96 h. In phase 2, these peak concentrations were found in point V2 at 144 h, in point V3 at 96 h and in ditch 1 at 312 h. During phase 1, peak concentrations of imidacloprid were found in ditch 2 at 2 h, in point B at 48 h and in the inflow at time 96 h. In phase 2, these peak concentrations were found in point V2 at 144 h, in point V3 at 96 h and in ditch 1 at 312 h.

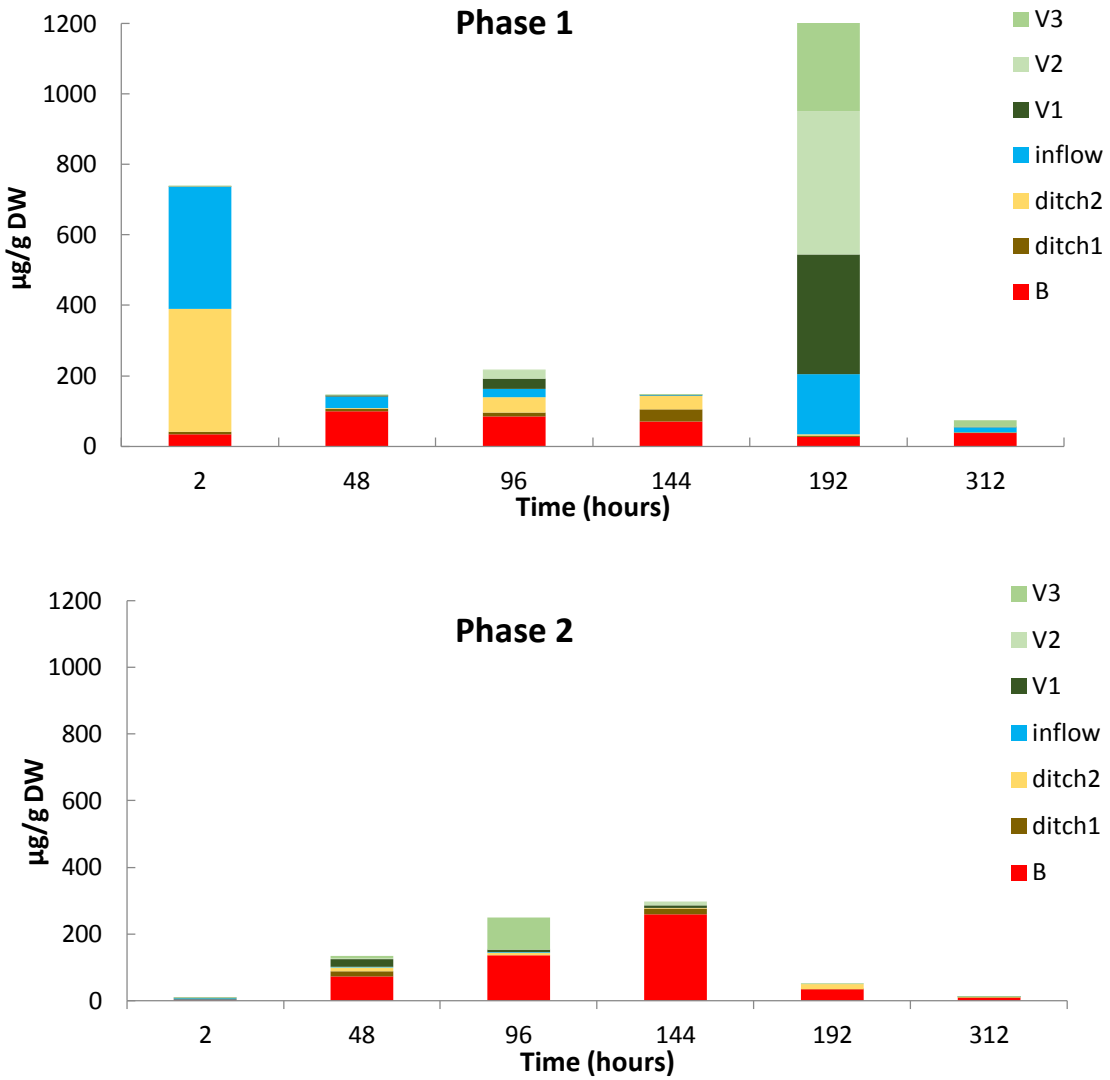

Figure 7-8: Amount of imidacloprid detected in sediment from different measuring points in a field scale constructed wetland under N. amazonum only (phase 1) and N. amazonum + E. polystachya (phase 2) conditions. Point V1 represents the "start" or inlet, V2 the "middle" and V3 "end" or outlet. B represents the agricultural field to which pesticides were applied.

Much higher amounts of imidacloprid in sediment were detected in phase 1 compared to phase 2. Peak concentrations were measured in phase 1 at time 192 h in the inflow, V2 and V3 and at time 312 h in V1 and V3 while in these same points very low amounts were detected in phase 2 (Figure 7-8 and Appendix 7, Table g-3). These results indicate that during phase 2, in the presence of two types of vegetation, more imidacloprid might be taken up by plants or be present in the water phase. The presence of vegetation and the duration of the simulated runoff may explain the higher concentrations found for the highly soluble imidacloprid (phase 2), but not the lower amount found in sediment. It seems likely that plants (higher density and multiple plant types) influence this uptake. Similar findings for the influence of wetland's vegetation density are presented in Lizotte et al. (2014), Vallée et al. (2014) and Vymazal and

Březinová (2015). Imidacloprid is a systemic pesticide which is soluble in water so that it can be absorbed by plants and transported in plant tissues (Vymazal and Březinová 2015).

7.3.6 Sorption isotherms experiments for three different soils from Suriname

The results obtained for sorption isotherms and kinetics are presented in Figure 7-9 up to Figure 7-12. The different properties of the soils are given in Table 7-7.

Table 7-7: Different properties of soils used in sorption experiments

Soil	Origin of soil	Depth (cm)	pH-H_2O	% of Moistue	%OC of Dry Solids	%OM of Dry Solids	% Sand	% Silt	% Clay	Texture
S1	Inlet	0-30	7.41	43.28	1.96	3.91	51.6	28.9	19.4	Loam
S2	Ditch 2	0-30	7.57	43.38	1.61	3.22	29.8	34.0	36.2	Heavy loam
S3	Planted area	0-20	6.53	34.06	0.23	0.46	98.3	0.4	1.3	Sand (loamy)

7.3.6.1 Sorption isotherms for lambda-cyhalothrin

The sorption isotherms obtained for the adsorption of lambda-cyhalothrin to three different types of soils (S1, S2, S3) are presented in Figure 7-9, together with the distribution coefficient, K_d (equal to the slope; $K_d = \frac{Cs}{Ce}$) and the regression coefficient (R^2), which indicates a good correlation for a linear curve fit. According to Yaron and Saltzman (1972) and Goldberg (2005), this linear model is only valid under a certain concentration range. In this study, the concentration range was between 0.0000-0.0602 mg/l for S3 and between 0.0000-0.3890 mg/l for S1 and S2 soils. A better linearity of the obtained isotherms in Figure 8-9 is obtained for S3. This is because of the lower concentration range and the higher R^2 value obtained for the experiment with S3 soils. Although a higher concentration range was used in the experiments using S1 and S2 soils, the obtained distribution coefficient (K_d) is the highest for soil S3. This can be explained by the properties (texture) of the three soils. Soils S1 and S2 have a much higher clay content compared to S3, which is a sandy soil. On the other side, the organic carbon content for S1 and S2 soils is much higher compared to S3 soils, but this was not the determining factor for the higher distribution coefficient as described in Vallée et al. (2014) (see Chapter 2).

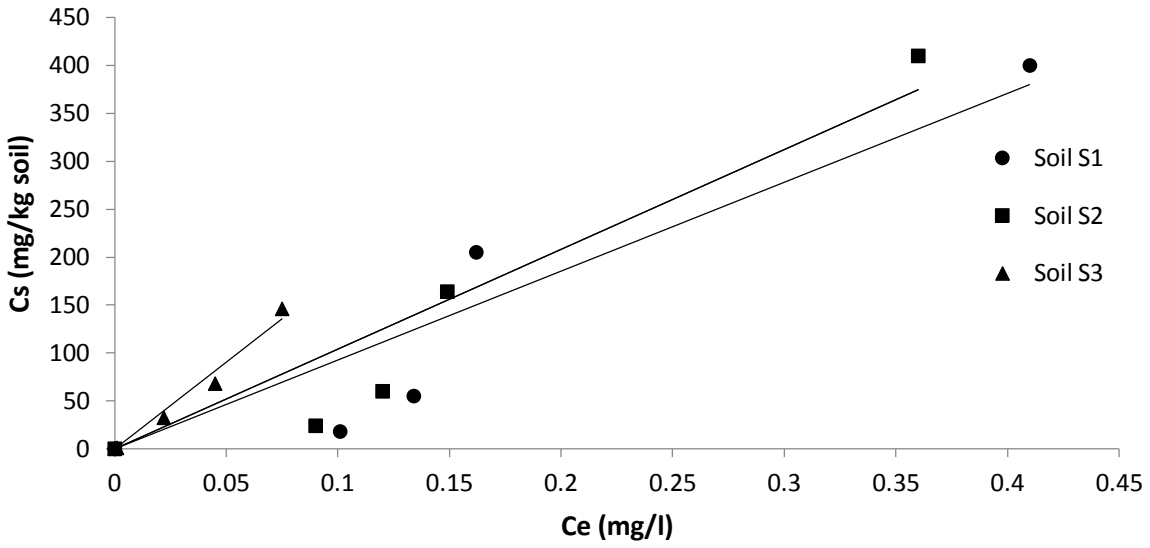

Figure 7-9: Sorption isotherms, presented as the sorbed concentration Cs (mg/kg) versus the concentration in the liquid phase C_e (mg/l) obtained for lambda-cyhalothrin when in contact with soil S1, soil S2 and S3, (n=2: experiment conducted with a paralell replicate)

Similar findings were done in the adsorption study with radio-labelled lambda-cyhalothrin (concentration range 0.019-0.306 mg/l) on 10 mineral soils. Average K_d values ranged from 1.970-7.610, while the coefficient of determination (R^2) for the relationship K_d vs. organic matter was 0.04, indicating that lambda-cyhalothrin's sorption is not dependent on the soil organic matter content (US EPA 2001).

Table 7-8: Correlation (R^2) and distribution coefficients (K_d) obtained from lambda-cyhalothrin adsorption isotherms in Figure 7-9

Soil	R^2	K_d (l/kg)
S1	0.9263	1028
S2	0.913	1080
S3	0.9539	2194

7.3.6.2 Sorption isotherms for imidacloprid

The adsorption isotherms for imidacloprid obtained by investigating the adsorption of this pesticide on 3 types of soils (S1, S2 and S3) from the wetland area are presented in Figure 7-10.

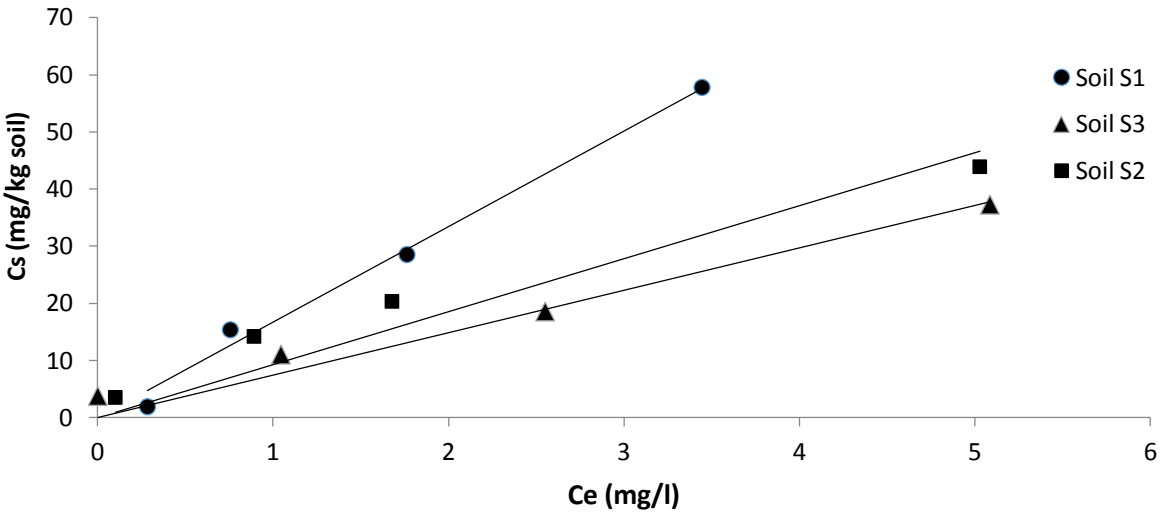

Figure 7-10: Adsorption isotherms, presented as the sorbed concentration Cs (mg/kg) versus the concentration in the liquid phase C_e (mg/l) obtained for imidacloprid when in contact with soil S1, soil S2 and S3 (n=2, experiment conducted with a paralell replicate)

The adsorption of imidacloprid follows a linear sorption model. From the obtained equations, the distribution coefficient, K_d (equal to the slope; $K_d = \frac{Cs}{Ce}$) and the regression coefficient (R^2) are presented in Table 7-9. The greater the R^2, the better the correlation for a linear model. In this study, the concentration range was between 0.00-10.00 mg/l. The linearity of the obtained isotherms in Figure 7-10 fits better for S1 (R^2: 0.9904), followed by S3 (R^2: 0.9592). The obtained K_d (16.7) for imidacloprid in S1 (soil from the inlet) is much higher than the one for S2 (soil from ditch 2) and S3 (soil from the agricultural field). The highest value is found for the soil with the highest moisture content (S3). This result was not found in the wetland experiment, except at time 2h for both phases 1 and 2. A higher K_d value was found for the soil with the highest organic carbon value (S1). Similar outcomes are presented in the studies of Krohn and Hellpointner (2002) and Yasgan et al. (2005). Based on the K_d values found (Table 7-8 and 7-9), much more lambda-cyhalothrin (a factor 295 times for S3) is found in the sediment compared to imidacloprid, which explains the higher affinity of lambda-cyhalothrin towards the sediment phase.

Table 7-9: Correlation and distribution coefficients obtained from imidacloprid adsorption isotherms in Figure 7-10

Soil	R^2	K_d (l/kg)
S1	0.9904	16.7
S2	0.9173	9.27
S3	0.9592	7.42

7.3.6.3 Sorption kinetics for lambda-cyhalothrin

Results for the sorption kinetics of lambda-cyhalothrin are presented in Figure 7-11. The results show an increase followed by a decrease from time 17 h up to time 25 h. The highest sorption for lambda-cyhalothrin was at time 17 h in all three soils. The values ranged between 1125 and 1193 mg/kg dry soil and were the highest for soil S2 (1191 mg/kg) and S3 (1193 mg/kg). The lowest values were measured at time 25 h (end of experiment) for S1 (831 mg/kg) and S2 (730 mg/kg), while the lowest value for S3 (816 mg/kg) was found at time 6 h. Compared to the wetland experiment the results for the sorption of lambda-cyhalothrin by wetland soil did not show a constant pattern e.g. increase or decrease.

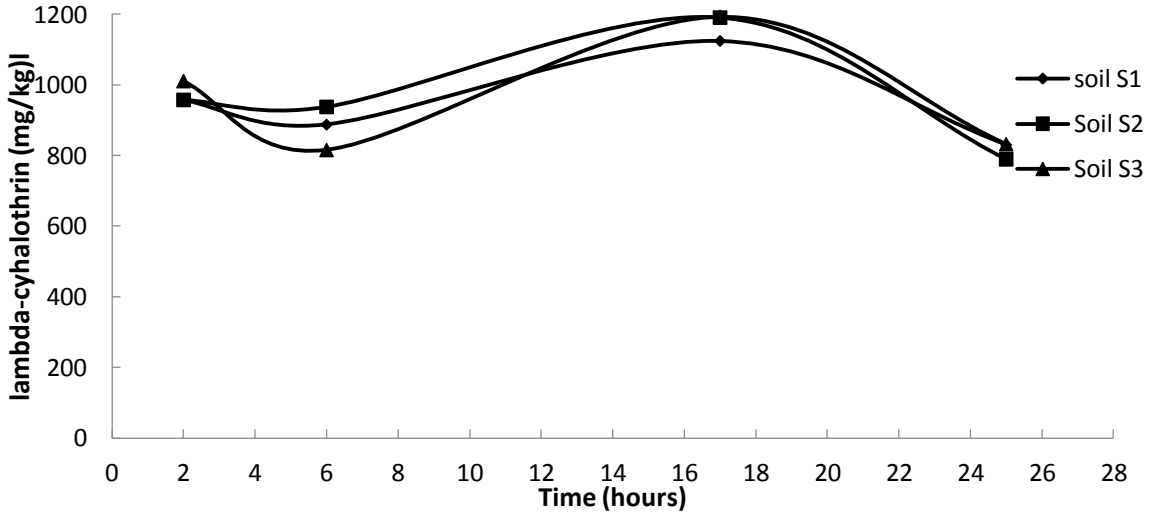

Figure 7-11: Sorption kinetics of lambda-cyhalothrin for three different soils of Suriname

According to Passeport et al. (2013); Tournebize et al. (2013) and Vallée et al. (2014) it is related to desorption or remobilization. However, for point B (comparable to S3 in the kinetic experiments) during phase 1, a constant decrease was observed starting from time 48 h up to the end of the experiment, while in phase 2 this was not the case.

7.3.6.4 Sorption kinetics of imidacloprid

The results for the sorption kinetics of imidacloprid are presented in Figure 7-12. Contrary to the results obtained for lambda-cyhalothrin, results show an increasing trend in sorption on the different soils over time. Calculation of the amount imidacloprid attached to soil, shows that at time 72 h (end of experiment) 68% (S1) and 88% (S2) was adsorbed. For S3, 79% was adsorbed at time 48 h, while results for lambda-cyhalothrin show that already at time 6 h, 95%, 100% and 87% was adsorbed to respectively S1, S2 and S3.

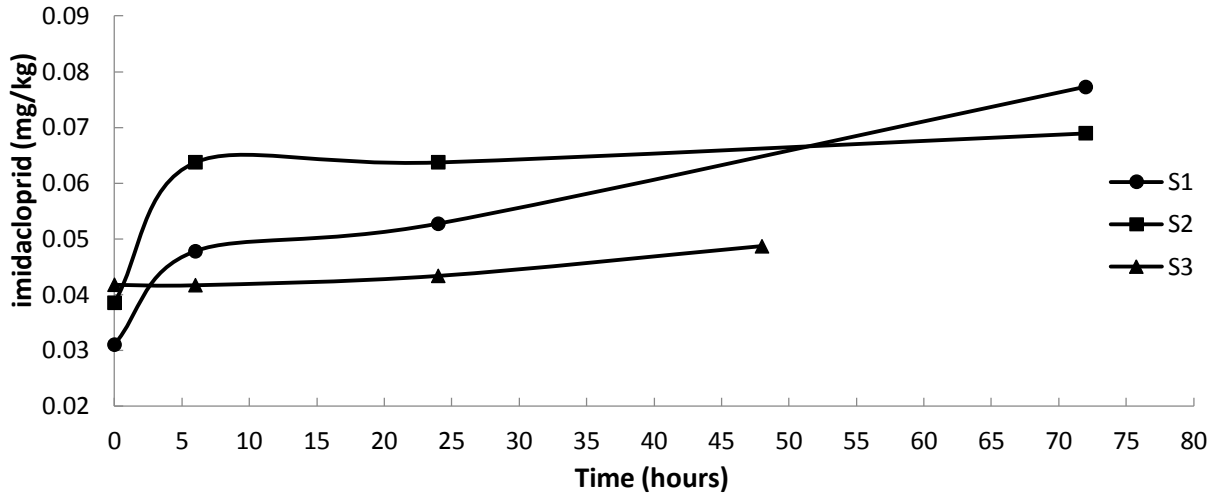

Figure 7-12: Sorption kinetics of imidacloprid for three different soils of Suriname

7.4 Conclusions

The detection of lambda-cyhalothrin in the water phase of both phase 1 (short rainy season) and phase 2 (dry season) was low with more detections in phase 2, were a longer irrigation regime was followed. The detection was not only depended on the irrigation regime, but also on the amount of pesticide applied and the way it reached the water phase. In the field scale wetland, pesticides were first applied to the agricultural soil and not directly in the water phase. In the previously conducted mesocosm studies, lambda-cyhalothrin was applied directly to the water phase in high concentrations (chapter 6) and was measured throughout the whole experiment (336 h), while lower amounts applied (chapter 5) in a similar manner as in chapter 6, resulted in detections in water up to 72 h. For both phase 1 and phase 2, a good dissipation of imidacloprid was found from the water phase. Repeated exposure (phase 2) to the pesticide did not result in higher removal rates in one specific point in time, while for the removal over the distance, a complete and stable removal was obtained during phase 2, starting at time 24 hours. Based on statistical analysis the overall wetland efficiency during phase 2 was not significantly higher than the results for phase 1, however this must be interpreted with caution because of the little difference of the obtained p-value with 0.05. Hence, it is carefully concluded that the wetland with a combination of two plants (*Nymphaea amazonum* and *E. polystachya*) shows a clearly higher removal of pesticides compared to the removal with one type of plant (*Nymphaea amazonum).* This should be confirmed with more repetitions of the experiment. One objective must also be to obtain imidacloprid effluent concentrations which are lower than the environmental quality standard needed for the protection of the aquatic environment.

Calculated amounts of pesticides in runoff are in agreement with values from the literature and can be explained by the pesticides properties, fate characteristics, duration of rainfall and

amount of pesticides applied. Higher amounts of imidacloprid were found in runoff in phase 2 because of e.g. a longer irrigation regime applied

Relative higher lambda-cyhalothrin concentrations were measured in root samples during phase 2 compared to phase 1, indicating a enhanced accumulation towards uptake of lambda-cyhalothrin by plants (combination of *Nymphaea amazonum* and *Echinocloa polystachya*). This, contrary to the mesocosm experiment (chapter 5) were lambda-cyhalothrin was not detected in roots.
Significantly more detections were done for imidacloprid in *Echinocloa polystachya* (shoots and leaves) compared to *N. amazonum,* while the amount detected in roots of *Echinocloa polystachya* was much lower than the amount detected in the plant part, but still significantly higher than the amount found in the roots of *N. amazonum*, indicating a higher uptake towards imidacloprid.

With the exception of a few points, lower amounts of lambda-cyhalothrin were found in sediments during phase 2 compared to phase 1. Perhaps, this relates to the presence of vegetation (higher plant density and multiple plant types) during phase 2. Sorption experiments may explain why the amount of lambda-cyhalothrin found in the sediment of measuring point B (comparable to S3) was much higher than that in measuring points ditch 2 (comparable to S2) and the inlet (comparable to S1). Sorption of lambda-cyhalothrin showed to be independent of the soil organic matter and was based on the highest K_d found for the soil with the lowest organic matter content (S3).Sorption kinetics indicated desorption from the sediment or remobilization in the water phase.

Similar as for lambda-cyhalothrin, much higher amounts of imidacloprid were found in sediment in phase 1 compared to phase 2 and might be related to higher plant densities in phase 2. Contrary to lambda-cyhalothrin, imidacloprid sorption was dependent on the soil organic matter. Sedimentation was not the major pathway of dissipation. The latter was overruled by plant uptake which is influenced by the systemic properties of imidacloprid and its high water solubility.

This field scale constructed wetland designed from input data from mesocosm experiments shows good capabilities for removal of lambda-cyhalothrin and imidacloprid from the water phase, especially during repeated exposure and with multiple plant types.

Chapter 8 General discussion, conclusions and recommendations

The world's population is projected to increase by 30% to 9.2 billion by 2050, therefore increasing the demand for food production and food security (Popp et al. 2013). The beneficial outcomes from use of pesticides provides evidence that pesticides will continue to be a vital tool in maintaining and improving food production worldwide (Zhang et al. 2007; Popp et al. 2013). The intensive use of pesticides however, leads to an increased risk of contamination of the environmental compartments and of harmful effects on humans, water resources and biodiversity (Schwarzenbach et al., 2010, Malaj et al., 2014; EFSA 2015). The risk of pesticide contamination by means of e.g. agricultural runoff can be reduced by applying best management practices and routines such as the implementation of constructed wetland systems within the agricultural landscape (Moore et al. 2006; Gregoire et al. 2009; Elsaesser et al. 2011; Elsayed et al. 2014; Vallée et al. 2015b; Vymazal and Březinová 2015). At the start of this doctoral research, there was very little information available on pesticide usage, pesticide residues in different environmental compartments and the possible contamination of these compartments in Suriname. In addition, no research was previously conducted, involving the mitigation of pesticide pollution and on the estimation of design parameter values for a field scale wetland. This research was therefore divided in two major parts: 1) evaluation of good agricultural practices by farmers in Commewijne, Suriname and 2) the use of constructed wetlands for the treatment of agricultural runoff. This chapter takes into consideration the research questions derived from the specific objectives (chapter 1), it discusses and gives the conclusions about the main findings of this dissertation and presents recommendations for future research.

8.1 General discussions and conclusions

8.1.1 Do farmers in district Commewijne, Suriname apply good agricultural practices (G.A.P.) in the use of pesticides and do vegetables comply with applicable maximum residue limits (MRL) values? What is the risk of human exposure to pesticides?

A face-to-face questionnaire was worked out and the consumer exposure to pesticides was estimated by means of the international estimated short-term intake (IESTI). Results showed that most of the farmers applied a dose, which was within the regulated label or EU dose. The misuse of pesticides by a minority of G.A.P. registered farmers however raises concern about the effectiveness of the G.A.P. program, which was implemented by the Ministry of Agriculture in 2003. In addition, the use of cocktails and non-authorized pesticides was observed. They raise concerns as some of the applied pesticides belong to the highest toxicity class of the WHO and therefore put consumers and the environment at risk. Misuse of pesticides was related to: the low educational level of farmers (Hurtig et al. 2003; Recena et al. 2006 and Polidoro et al.

2008); the lack of a registration system and training programs on pesticide use (Salameh et al. 2004; Ibitayo 2006 and Abhilash and Singh 2009); the absence of a meteorological station and weather forecasting; the easiness to buy non-authorized pesticides from e.g. neighbouring countries and the lack of legislation and control mechanisms (Popp et al. 2013).

Good results were obtained for the use of some of the PPEs (mouth cap, boots and protective clothes). The IESTI was below the EU ARfD for imidacloprid. Interpretation however must be done cautiously, because pesticide residue analysis showed that 80% of the imidacloprid detections were above the EU MRL value and that comparison with the newly proposed ARfD will raise the IESTI to values near 100%. The MRL-value is a trade norm checking if pesticides have been applied according to G.A.P. Values above this norm have economic consequences for Suriname, and result in the rejection of imported crops by the Netherlands (biggest export market). It must be emphasized that from the 24 most frequently used pesticides in Suriname, only 6 were quantified in this study. With the recent acquirement of new analytical equipment (LCMSMS) by the laboratory of Crop Protection Chemistry, analytical limitations are not expected to be an issue anymore and outcome of future research will provide a better overview of pesticide residues in food and the possible exposure risk to humans.

8.1.2 What is the status of the pesticide contamination of the different environmental compartments (surface water and sediment) and the bacteriological quality of irrigation water, and what are the types of wetland plants present in the main ditches of the research area?

In less than 20% of the surface water samples, residues of the pesticides α-endosulfan, lambda-cyhalothrin, chlorothalonil and imidacloprid were found with concentrations much higher than international norms for the protection of surface water. They therefore pose a health threat to e.g. humans and aquatic life. Multiple detections of imidacloprid were done in both main ditch water and sediment in concentrations higher than the environmental norms, while α-endosulfan and lambda-cyhalothrin detections were done in respectively surface water and sediment. For the drainage ditches on farm level, imidacloprid and lambda-cyahlothrin were detected multiple times and gave an estimate of the concentration range that can be expected in runoff within the research area (Alkmaar) for the two pesticides. These concentrations could however be underestimated when information provided by the farmers about the time of application is not correct (pesticide was not applied recently) and simulated runoff is not applied or rainfall does not occur shortly after the application of pesticides. Sampling of these drainage ditches was based on information provided by farmers and should therefore first be validated by applying runoff experiments using the recommended label dose. Chlorothalonil was not detected in runoff and results indicate a fast dissipation from the water phase. The fast dissipation from water (around 0.7 days) was observed for the mesocosm experiments and it can be expected that for field experiments this will even be much lower (around 0.1 days; PPDB 2015). Another explanation is that this pesticide, in contrary to the information provided, was not applied recently or not applied at all. A registration system could have provided more details on e.g. the type of pesticides applied, application rates, and frequency of applications.

However, such a system, which is obligate in Europe, does not exist in Suriname. Similar as for the analysis of the crops only a few of the pesticides were quantified in the compartments investigated. This assessment was done in the long rainy season and dilutions of pesticide concentrations might have occurred in the water by the influence of rainfall. The microbiological quality of irrigation water in the research area was poor. Ground- and surface water tested positive on total - and fecal coliforms and *Escherichia coli*, with more detections for surface water. The latter was because groundwater is mostly better protected (confined by soil layers) from stray cattle and other sources of pollution. The microbial quality was determined during the long dry season and the start of the small dry season. The microbial quality of irrigation water may exhibit both diurnal and seasonal variability as well as be affected by precipitation events (Pachepsky et al. 2011). It s therefore advisable to do the microbial quality assessment during other seasons. In Pachepsky et al. (2011), there was a clear trend to a decrease of *E.coli* concentrations from morning to mid-day. Rainfall events may increase concentrations of pathogens and indicator organisms in surface water due to runoff and release of bacteria from bottom sediments, while on the other hand, extreme rainfall events may dilute concentrations. The vegetation diversity assessment provided valuable information on the abundance of plants in the main drainage ditches. Selection of plants for wetland studies must be done with care e.g. plants should be used which are abundant, however they should not become invasive (Kadlec and Wallace 2009). Also multiple plant types are preferred above monocultures. Not all pesticides (around 25%) belonging to the group of most frequently used pesticides (G.A.P. survey) could be validated in current study.

8.1.3 What is the potential of vegetated and non-vegetated wetland mesocosms to remove selected pesticides in agricultural runoff?

Different removal/retention mechanisms exist for the most important types of contaminants including pesticides in agricultural runoff. For the more hydrophobic pesticides and the water-soluble pesticides different mechanisms of removal were found. Water-soluble pesticides are mainly removed by microbial degradation in the water phase and retention by the water-plant interphase, and some have depending on their properties (high K_{oc}) also very good sorption capabilities, which can be influenced by the vegetation density. For the more hydrophobic pesticides, adsorption to particles and sedimentation is one of the main mechanisms. The potential of wetlands to remove selected pesticides was investigated by two mesocosm studies, which were presented in chapter 5 and chapter 6.

8.1.3.1 Mitigation of lambda-cyhalothrin and imidacloprid pollution by wetland plants

A complete dissipation of lambda-cyhalothrin and imidacloprid from the water phase was obtained for the low target concentrations for both types of mesocosms (*N. amazonum* and *E. mutata*). Contrary to imidacloprid, higher removal efficiencies were obtained from the water phase for lamda-cyhalothrin (time interval 24-48 h) in repeated batches, which might be caused by an enhanced biodegradation due to repeated exposure. This enhanced biodegradation was

also observed for imidacloprid, but only during exposure to the highest dose and was only during the time interval 24-72h (imidacloprid in *N. amazonum* mesocosms). Dissipation of lambda-cyhalothrin from the water phase was independent of the dose applied and type of plant. In contrast to that, the dissipation of imidacloprid was dependent on the type of plant, but only for the highest dose. DT_{50} values obtained by linear regression were not consistent (much higher) compared to the 50% dissipation found for imidacloprid in mesocosms exposed to the highest dose and were consistent with the low R^2 values found. Because the DT_{50} of imidacloprid in the repeated batch (5) was significantly higher than those obtained for batch 1, it is unlikely that an enhanced biodegradation occured, because that would have resulted in lower DT_{50} values. The main pathway of lambda-cyhalothrin was adherence to sediment particles (consistent with other findings in the literature) and possibly to the tub walls. Other studies however showed a preference towards plants uptake and relate this to the plant density.

The amount of imidacloprid detected in plants of *Nymphaea amazonum* was over 3320 times higher than the amount of lambda-cyhalothrin. Lambda-cyhalothrin was observed in the shoots and leaves of *Nymphaea amazonum*, while for *Eleocharis mutata* mesocosms, accumulation occurred in the roots. For imidacloprid, both parts of the plants were responsible for its uptake, but this was more pronounced for the above mass of plants. A good correlation was found by linear regression to estimate the DT_{50}'s of lambda-cyhalothrin (around 1 day) and imidacloprid (1-9 days) in the water-phase. The DT_{50} of imidacloprid was lower for low target concentrations compared to higher target concentrations. Statistical analysis showed significant differences in imidacloprid half-life times in mesocosms exposed to the 60 µg/l and mesocosms exposed to 180 µg/l.

According to Kadlec and Wallace (2009) and O'Geen (2010), biodegradation of pesticides can be enhanced in constructed wetlands when repetitive exposure occurs to the same pesticides over time, due to induction and adaptation of microbes, leading to establishment of organisms capable to degrade the pesticides more rapidly in the wetland. This might also be the case in present study, because the same mesocosms were exposed to the pesticides in consecutive batches and therefore further research is needed. Quantification of the amount of pesticides in harvested plants and in sediment from wetland mesocosms, showed that less than 50% of lambda-cyhalothrin is recovered. Imidacloprid was almost comletely recovered in *Nymphaea amazonum* mesocosms, while only a small amount (around 16%) was traced back in plants and sediment of *Eleocharis mutata* mesocosms. These differences which are possibly related to photolytic decay, (bio) degradation and formation of metabolites must be further investigated. Sorption to tubs will not likely occur because both types of mesocosms were exposed to imidacloprid and around 96% was traced back in the *N. amazonum* mesocosms.

8.1.3.2 Fate of chlorothalonil, lambda-cyhalothrin and imidacloprid in wetland mesocosms

In this mesocosm experiment a different set-up was used compared to that in the previous chapter. The set-up consisted of exposure of vegetated (*P. australis*) and non-vegetated mesocosms with a constant and much higher exposure concentration of pesticides during 5 consecutive batches. The high exposure concentration in present study might have resulted in the temporary drawback in removal especially of lambda-cyhalothrin observed in batch 3 and batch 4 of the experiment, while for chlorothalonil and imidacloprid a good removal was found. For the 3 pesticides an enhanced removal was observed during repeated exposure (batch 2 (chlorothalonil and lambda-cyhalothrin) and batch 2 and batch 5 (imidacloprid)), which might be related to an enhanced biodegradation. Contrary to imidacloprid, lower degradation rates were found for chlorothalonil and lambda-cyhalothrin, which might be caused by the higher application rates causing inhibition of microbiological activity. However, when compared to the previous mesocosm experiment (chapter 5) a much higher degradation rate and also lower half-life values were found for imidacloprid, despite of the lower temperatures measured in present study, which indicates that other factors contrary to the temperature e.g. photo degradation (PPDB 2016) might be responsible for imidacloprid removal. Higher removal rates and lower half-life times found for the non-vegetated mesocosms, suggest that vegetation did not play a significant role in the removal of lambda-cyhalothrin, while comparison with the previous mesocosm study suggest that temperature was a determining factor in lambda-cyhalothrin's removal.

Chlorothalonil has a half-life time, which was consistent with findings from the literature and is volatile (Sherrard et al. 2004; PPDB 2016). In present study, exposure concentrations of 500 µg/l chlorothalonil, when applied immediately to the water phase can be measured up till 18-20 h for respectively vegetated and non-vegetated systems, while in chapter 4, chlorothanlonil was not detected in drainage ditches near the farmland, immediately after application and irrigation. Because of this short time-span (up to 20 h) of chlorothalonil dissipation from water, it is advisable to validate results (concentrations found in runoff) by means of pesticide applications at the field and to start the water sampling immediately after a simulated irrigation. Compared to the previous mesocosm study, higher half-life times were obtained for lambda-cyhalothrin when higher target concentrations were applied. The uptake of chlorothalonil and imidacloprid was mainly in the plant upper part (shoots and leaves), while lambda-cyhalothrin was situated in the roots part and consistent with the uptake pattern of *E. mutata* in previous mesocosm experiment. In addition, amounts of lambda-cyhalothrin (µg/g DW) were much higher compared to plants uptake in previous mesocosm study and other wetland studies and shows good capabilities for use in wetlands abating pesticide pollution.

8.1.4 What is the potential of a field scale wetland to mitigate pesticide pollution originating from agricultural runoff under 2 conditions: 1) wetland planted with a monoculture and 2) wetland planted with multiple plant types?

To answer the last research question a reflection is made of chapter 7. A good overall dissipation of imidacloprid (> 95%) was found in the water phase under conditions with a monoculture of *N. amazonum* (phase 1) conditions with multiple plants (*N. amazonum* and *E. polystachya (phase 2)*.
Lambda-cyhalothrin was not detected in the water phase of the surface flow wetland, with a few exceptions for phase 2 (conditions with multiple plant types) and was mainly detected in the first receivers of agricultural runoff, ditches 1 and 2. Lambda-cyhalothrin was detected in water only after a much longer irrigation time performed in phase 2. In addition, more of the water-soluble imidacloprid was found in runoff in phase 2 (1.9-5.7%) compared to phase 1 (0.2-1.4%) within 2 hours after the application of pesticides and applying a simulated rainfall. These calculations are a theoretical estimation using the amounts applied, the water level rise in the ditch and the total ditch volume. Taking into consideration the influence of other factors/processes such as volatilization, sorption, photolytic degradation, microbiological degradation, accumulation in plants and sediment, but also more frequent sampling of all compartments, can provide a better estimation of the amounts of pesticides found in runoff and surface water. Repeated exposure to the pesticide did not result in higher removal rates in one sampling point in time, but the overall efficiency of phase 2, showed a higher and more constant pattern of removal for imidacloprid, which started at time 24 h.
Lambda-cyhalothrin's and imidacloprid uptake by plants was the highest for the upper plant part for both types of plants during phase 1, however much more imidacloprid was detected in the plants and roots of *Echinocloa polystachya* (phase 2). Also significantly more lambda-cyhalothrin was found in the roots of *N. amazonum* plants during phase 2 compared to phase 1. It seems like with a combination of two plants (phase 2) a certain competition for pesticide accumulation is present. The results for this field scale wetland are contrary to those from the mesocosm experiment described in chapter 5, in which no lambda-cyhalothrin was detected in the roots of *N. amazonum*. Lower amounts of lambda-cyhalothrin and imidacloprid were found in sediment samples during phase 2 compared to phase 1 in most of the sampling locations. Findings were in agreement with the higher amounts found in plants especially for *Echinocloa polystachya*, and might be related to a higher plant density. Although the plant density was not measured by counting the plants, it can be concluded based on viewing the pictures taken during the experiment, that for phase 2, the plant density in the water was higher.

Based on the K_d values found, lambda-cyhalothrin's dissipation is faster from the water phase to the sediment compared to imidacloprid and independent of the type of soil (sand to heavy loam). Sorption kinetics showed a better insight in the dissipation of lambda-cyhalothrin from the water phase compared to imidacloprid and might explain the irregular pattern of imidacloprid accumulation in sediment over time.

Based on the high removal efficiencies for high initial imidacloprid concentrations in phase 1, it is expected that actual (much lower) effluent concentrations in ditches (presented in chapter 4) are further reduced and environmental norms are met. Although the DT_{50} value was not derived through linear regression, imidacloprid removal from the ditch water (ditch1 and 2, Figure 7-3) to half of its concentration was around 1 day and consistent with the results found for the experiments described in chapter 5 and 6. The design of this field scale wetland was based on input data from mesocosm experiments.

From the results obtained from the wetland mesocosm studies it can be concluded that for the three pesticides investigated (chlorothalonil, lambda-cyhalothrin and imidacloprid) in general a high removal was found in the water phase. The results from mesocosm studies (chlorothalonil excluded) were validated in a field scale wetland and results obtained, show comparable findings for e.g. the dissipation of pesticides in water and the accumulation in plants (shoots and leaves) and sediment. The use of multiple plant types resulted in an improved removal of different types of pesticides from the water phase and might be the reason for the lower amounts of pesticides found in sediment during phase 2.

8.2 Future perspectives

Limitations were found during the research. This section displays additional information to carry out the research more targeted. Research perspectives are recommended and management options are presented.

8.2.1 Research perspectives

The G.A.P. survey should be extended to other agricultural areas in Suriname (e.g. the Kwatta area) and the survey questions should be broadened to the use of e.g. irrigation water and the type of irrigation method used. This is because of the concerns on the microbial quality of the water. Other used pesticides/vegetables combinations must be analyzed. Photolytic degradation experiments, especially for imidacloprid must be executed to determine pesticides losses through this pathway. The low educational level of farmers was a restriction not only during the survey (right answering of questions) but also for the adequate use of pesticides in daily live. Only a few pesticides were quantified in the different compartments. The lack of good analytical methods and instruments to analyse more pesticides has to overcome.

Water should be sampled during a complete agricultural season and under different weather conditions and simultaneously with the sediment fraction and plants. This approach can facilitate mass balance calculations of pesticides in e.g. runoff. For instance during the dry season a higher concentration of pesticides is expected in waterways. Groundwater and rain (is collected and used for drinking purposes) must also be taken into consideration. The leaching potential of pesticides to groundwater must also be assessed by determining the GUS (Groundwater Ubiquity Score). Studies must also focus on the microbial quality of irrigation

water. Answers to the questions on how to improve the present situation in district Commewijne, and how to treat the contaminated irrigation water simultaneously with runoff containing pesticides in constructed wetlands should be looked for.

For both mesocosm experiments, monocultures were applied, while for the field-scale wetland a mix-culture was used during phase 2. From the latter, preferences in pesticides uptake were seen and it is therefore recommended to do future research involving mesocosms with both mono- and multiple types of plants, by making a selection from the list obtained in the vegetation diversity assessment study described in chapter 4 and Appendix 2, Table b. Results of this assessment show that the species *Ludwigia stolonifera*, belonging to the genus *Ludwigia* was found and seems interesting for future wetland research involving lambda-cyhalothrin. Research should be extended to investigate pesticides fate by means of sedimentation in *P. australis* and non-vegetated mesocosms, microbial degradation of pesticides and formation of possible metabolites. Different application rates, plants, types of mesocosms (flow-through) and repetitions and duration of experiments has to be addressed. In addition, batch experiments following respiration or quantification of microbial activity and sorption on different types of substrate (e.g. straw, soil with varying organic carbon content, sediment) but also to the tubs in mesocosm experiments can be used to assess the fate of pesticides in soil-water (plant) systems. Hydroponic cultures can be used to investigate pesticide-plant interactions, this without the influence of soil/sediment. Design configurations can be adjusted and other parameters such as porosity and density of plants and type of substrate (especially for subsurface flow wetland systems) can be incorporated.

Prior to the execution of a field runoff experiment, the vegetation diversity has to be determined in different parts of the ditch. Sampling in different compartments of the ditch provides a better insight in the dissipation of the pesticides in water and the uptake especially by plants and sediment. The ditch length required for aquatic toxicity reduction can be determined. Batch sorption experiments using substrates with different organic carbon content can be executed, to investigate the sorption behaviour of lambda-cyhalothrin. Runoff is also an important parameter for the design of a constructed wetland and experiments using different simulated runoff regimes, pesticides and soils in combination with infiltration studies must be conducted. The leaching potential of pesticides can be investigated by means of column experiments using different types of soil and runoff treatments.

Measurement of different water quality parameters (especially for the field scale wetland experiment), was very time-consuming and labour intensive. It is recommended to work with automated field measuring stations. In the real situation, wetlands are with a mix of plants and for a better representation, a mix of more than two plants abundant in the research can be used, which can firstly be investigated in mesocosms studies. A possible solution to measure lambda-cyhalothrin much longer in water is to add it, immediately to the inflow of the wetland. Further recommendations are to observe the water of the wetland including the uptake by plants and the deposit in the sediment for a longer period.

For future studies, pesticides can be investigated along with other runoff constituents/contaminants (nutrients, trace metals, dissolved organic matter (DOM), suspended solids and pathogens), for which a good removal has been found in other wetland studies, especially of pathogens. This by taking the recommendations presented in chapter 2 into consideration, such as the use of a hybrid system for efficient pathogen removal.

8.2.2 Management options

Pesticide labels must be written in the spoken or native language in order for farmers to understand them and a registration system for pesticide use should be made mandatory. Research programs should be executed to improve the knowledge of farmers and other stakeholders involved in the selling and handling of pesticides towards pesticides and G.A.P. and to improve sustainable agricultural practices.

A meteorological station and weather forecast in the research area will improve the efficient application of pesticides and will reduce the amount of pesticides ending up in the receiving water bodies. It will also help in a better interpretation of obtained runoff data or fluctuations in measured pesticide concentration. Having a well-functioning and operational pesticide residue laboratory should be a priority for the Surinamese government and other research institutions. Studies must focus on adequate pesticide legislation, control mechanisms and border control. The AHFSU unit of the ministry of Agriculture should provide MRLs and toxicological limits for different pesticides and should execute food-monitoring studies.

The Ministry of Agriculture in Suriname must promote sustainable agricultural practices incorporating IPM (Integrated Pest Management) and ICM (Integrated Crop Management). This will reduce the use of pesticides and the emission to the environment.

Future research should result in a prototype wetland at farmer's hand. This wetland must be able to efficiently treat a selected waste stream containing pesticides and other contaminants in runoff. Researchers and students can all benefit from wetland research projects. Integrated pest management studies can also be conducted e.g. the use of flooding to remove invasive plant species instead of the use of herbicides and the use of mosquito fish or bacteria to control mosquitoes in surface flow wetlands.

Wetlands are a sustainable and environmentally sound approach to abate water pollution. Instead of discharging polluted agricultural water into surface water without any form of treatment (as which is the case not only for district Commewijne, but also for the whole country), this water can be diverted to such a system. As mentioned in the introduction, many of such systems already exist in the research area and can be easily adjusted to meet design criteria. That will also reduce the costs needed for extra land. Supervision and guidance of farmers in Suriname by e.g. the Ministry of Agriculture, Animal Husbandry and Fisheries and monitoring studies are essential tools in sustaining these systems and to protect the environment.

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Appendix 1: Literature review of removal/retention of pesticides in wetland treatment systems

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Acetochlor (H)	Four VEG columns of borosili-cate	Inner diameter 15 cm, H: 65 cm. Substrate layer 5 cm gravel (Ø 1-2 mm) and sand layer 52 cm (Ø 0.40–0.63 mm)	1.9 µM	Mass removal; dissipation; bacterial populations under different environmental conditions	Mean mass removal: 61 ± 14%; Dissipation: 56-90%	Dissipation mainly in root part together with nitrate and sulphate reduction. Root channelling to top part of column; ESA (Ethane Sulfonic Acid) degradation product formed.	Duration 112 days; HRT: 9.3 days; Q: 0.33 ml/min. Different microbial communities formed in different segment of column	Elsayed et al. 2015
Alachlor (H)	Four VEG columns of borosili-cate	Inner diameter 15 cm, H: 65 cm. Substrate layer 5 cm gravel (Ø 1-2 mm) and sand layer 52 cm (Ø 0.40–0.63 mm)	1.9 µM	Mass removal; dissipation; bacterial populations under different environment-tal conditions	Mean mass removal 52 ± 12%; Dissipation 32-64%	Dissipation mainly in root part together with nitrate and sulphate reduction. Root channelling to top part of column; ESA (Ethane Sulfonic Acid) and oxanillic acid (OXA) degradation product formed.	Duration 112 days; HRT: 9.3 days; Q: 0.33 ml/min. Different microbial communities formed in different segment of column	Elsayed et al. 2015
Ametryn (H)	SSFCW VEG	24 m^2	1 g m^{-3}	inclination	39%	Biodegradation; Plant uptake and desorption processes	Duration: 11weeks; HRT (days): 3.8; Sol_{H2O} (mg/l): 204; K_{oc} (cm^3/g):316-445	Borges et al. 2009
Atrazine (H)	Micro-cosms	0.78 l(sm) 202 l (lm)	15-18 µg/l	Substrate; Size microcosm	sm: 87.5%; lm: 43.3%	Hydrolysis; Oxidation; Sorption; s	Duration (days): 41-50. Metabolites	Laabs et al. 2007

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Atrazine (H)	Micro-cosms Veg/NON VEG	0.0042 m^3 Sediment**:** 10cm Water: 8.0 cm	2.23 kg a.i. /ha Com-bined with lambda-cyhalo-thrin: 0.028 kg a.i./ha area: 2.03 ha	Uptake of pesticides by vegetation; - Reduction of toxicity	(*J. effuses):*6.4 µg/kg (day 7)/Sediment: 24 hours: 1902.6 µg/kg (L. peploides): 2461.4 µg/kg (day 7)/Sediment: 24 hours: 752.1 µg/kg	Partitioning to plants and sediment; Highest uptake by l.peploides (in microcosms amended with Atrazine and combined microcosms	Duration 56 days Sol_{H2O}: 32 mg/l Log K_{oc}: 1.97; DT_{50}: 30 days; Precipitation 0.64 cm	Bouldin et al. 2005
Atrazine (H)	SSFCW; VEG	0.045 m^3 static batch CW with gravel beds	0.1 mg/l	Salinity (0-15 g/l) influence on degradation	44-47%	Increase in salinity; decrease in degradation and neg. influence micro-organisms; Main removal: biodegradation and metabolite formation	DT_{50} :16 days	Lin et al. 2008
Atrazine (H)	SFCW	(59-73)x14x0.3 m^3	73 µg/l and 147 µg/l	Atrazine removal; Partitioning; Required design parameters	For 73 µg/l: 66-70% For 147 µg/l: 34-37%	Limited uptake by plants or sediment; Main removal from the water phase (transferred or transformed)	Duration: 35 days DT_{50}: 16-48 days	Moore et al. 2000
Atrazine (H)	SFCW	5400 m^2; 2 cells (deep and shallow)	42 ml 0.48 kg/l; Aatrex 4L per 1893 litres	Environmental fate	Shallow cell: 89%; Deep cell: 70%	Dissipation affected by HRT; Higher removal in shallow cell	Duration: 21 days; Shallow (0.3 m) and deep (1 m)	Locke et al. 2011

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Atrazine (H)	Micro-cosms	Jars of 300 ml	2 mg/l (amend-ments with and without algae and lindane (2 mg/l)	Interaction between algae and pesticides	Retention water without algae: 51 ± 5%; Water with algae: 36 ± 3%; Mixture with lindane; Without algae: 64 ± 9%; With algae: 44 ± 6%	Atrazine with or without lindane; no sig. removal difference; Algae provides: sorption sites, facilitates pesticide degradation ,	Jars placed in growth chamber with algae culture for 11 days; Algae acts as photo catalyst: mechanism not fully understood	Friesen-Pankratz et al. 2003
Atrazine (H)	High density Poly-ethylene containers . (VEG and NONVEG meso-cosms	1.32 mx0.70 mx0.66 m. Substrate 16 cm silt loam sand above 22 cm of sand. Water depth 5.4-15 cm	Target concen-tration: 20 µg/l (exposu-re for 6 h followed by a 42 h rest period. After this additio-nal 6 h of flushing with unamen-ded well water	Pesticide load decrease after: Initial runoff; Flushing (investigate pesticide sorption to plant); total load decrease	Total load decrease 31 ± 4% (*S. americanum*); 35 ± 8% (*T. latifolia*); 45 ± 7% (*L. oryzoides*). NONVEG mesocosms (13 ± 20%) significantly different from *T. Latifolia and L. oryzoides*	Mass retention and plant sorption (reversibel)	Plant adaptation period 6 weeks; HRT 6 hours;	Moore et al., 2013
Azinphos-Methyl (AZP) (I)	SFCW; VEG	4400 m^2	0.40 ± 0.03 µg/l	Retention, fate and effects on *Copepoda Cladocera*	90 ± 1% (water); 61 ± 5%: mass retained	water phase: uptake by plants; no uptake by sediments; volatilization, photolysis, hydrolysis, metabolic degradation	5 spray drift trials; duration of sampling: 1week; $K_{oc::}$1000 L/kg; $Sol_{H2O:}$ 29 mg/L (25 °C) Henry's constant : $9.5x10^{-11}$ atm m^3 mol^{-1}	Schulz et al.

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Bifenthrin (I)	SFCW	4.8 ha	Below detection limit 4.8 ng/l	Phase partitioning; Dissipation using two sediments; Removal as a function of particle size	C_{ss}:1.0x10^{6} ng/kg; K_d (L/kg): 8.83x10^{4}; Retention highest in 0.7-2 µm clay fraction, (± 70% of concentration); pos. correlation particle size and concentration for > 53 µm particles, correlation with high OC content	Aging of sediments; decreased availability of microbial degradation; Strong sorption to sediments with high OC	DT_{50} (flooded + anaerobic): stable; DT_{50} (dry, aerobic): stable, more persistent	Budd et al. 2011
Bifenthrin (I)	VEGSFCW 1; VEGSFCW 2; Sediment basins (SB)	CW1: 2.3 ha; CW2: 2.5 ha	CW1: 2.0 ng/l; CW2: 2.6 ng/l; SB: 3.1 ng/l	Efficacy of flow through CW; Evaluate factors of importance	CW1: 69%; CW2: 84%; SB: 6%	Removal in CW1:mostly in part with more vegetation; CW2: larger flow path, more dense vegetation which promotes particle settling	Duration: irrigation season (3½ months); HRT CW1: 1h; HRT CW2: 18h; Adsorption and physical trapping also important	Budd et al. 2009

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Boscalid (H)	1 ditch (CW1 with 16 different plants) and 1 triangle pond (CW2 with 17 different plants);	CW1: 13 m (l)x6 m (W)x1 m; V equals 4 m^3; CW2: 20.5 mx15.5 mx11 m (triangle) and a 22 m (L)x1 m (W) lake	Year 2012-2013, Inlet CW1 46 mg/ha; CW2, Inlet 76.7 mg/ha; year 2013-2014: inlet CW1, 10.4 mg/ha; CW2, 77.8 mg/ha; CW1 received drainage from a 5 ha plot with a 12.5% slope, CW2 received from a 9 ha plot with a 5.0% slope	Effectiveness of two CW during two growing seasons (2012-2013; 2013-2014) for abate-ment of pesticides; effect pesticide properties on removal/reten-tion	2012-2013: efficiency CW1 0.3%; CW2 - 12.1%; year 2013-2014; efficiency CW1 33.1%; CW2 5.3%; max. Concentration in substrate 75 µg/kg;	High sorption and retention to substrate and plants due to high K_{oc} and low solubility; Negative efficiency CW2 related to runoff and remobilization. Boscalid was detected at low and constant concentration during each season because of desorption	HRT: CW1 0.25-55 h; CW2 5.6-415 h; Q_{max} CW1: 100 m^3/d/ha; CW2 50 m^3/d/ha.	Vallée et al. 2015b
Chlorothalonil (F)	Meso-cosms, VEG	1.85 m (L)x 0.63 m (W)x 0.63 m (H) or 0.73 m^3	148-326 µg/l	Extent and rate of removal over 72 hours; Toxicity towards *P. promelas* and *C. dubia*	> 99.5%	Sorption and biodegradation; Aquatic aerobic degradation DT_{50}: 8d; Anaerobic degradation: DT_{50}: 5d	Duration: 72 hours Log K_{oc}: 3.14 Sol_{H2O}: 0.60mg/l	Sherrard et al. 2004

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Chlorpyrifos (I)	VEGSFCW 1, VEGSFCW 2; Sediment basin (SB)	CW1: 2.3 ha; CW2: 2.5 ha	CW1: 3.9 ng/l; CW2: 3.0 ng/l; SB: 5.1 ng/l	Efficacy of flow through CW; Evaluate factors of importance	CW1: 61%; CW2: 52%; SB: not provided	Smaller reductions compared to pyrethroids; lower Koc's; vert. transport; Sedimentation; plant uptake	Duration: irrigation season (3½ months); HRT CW1: 1h; HRT CW2: 18h	Budd et al. 2009
Chlorpyrifos (I)	Meso-cosms, VEG	1.85 m (L)x 0.63 m (W)x 0.63 m (H) or 0.73 m^3	0.9-19.9 µg/l	Extent and rate of removal over 72 h; Toxicity towards *P. promelas* and *C. dubia*	± 100% removal; first order kinetic removal rate: 0.067 h^{-1}, r^2 : 0.83	Sorption and biodegradation; Aquatic aerobic degradation; DT_{50}: 12-41 days; Anaerobic degradation: 35 days	Duration: 72 hours; Log K_{oc}: 3.95; Sol_{H2O}: 1.40 mg/l	Sherrard et al. 2004
Chlorpyriphos (I)	SSFCW	55 m^2 with gravel bed	100 mg/40 l	Behavior in SSFCW	80-90%; 0-20%: uptake by gravel	Biodegradation; Plant uptake probable elimination pathway	Duration; 450 hours; Log K_{ow} : 4.96	Matamoros et al. 2007
Chlorpyriphos (CPF) (I)	Micro-cosms; Sediment-water; 2 types: flooded + drain-fill	0.0018 m^3 16 cm sediment + 6 cm water	Flooded: 400 \µg/l; Drainfill: 800 µg/l	Drainfill cycling on CPF mineralization relative to permanently flooded conditions	Drain fill: < 25%; Relative to permanently flooded: as high as 50% (less CPF removal)	Flooded systems: more microbial activity (two-fold at the end of experiment); Lower CPF hydrolysis rate; High sorption rate	Duration: 53 days; Log K_{ow}: 5; Sol_{H2O}: 1.39 mgl^{-1} (20° C); DT_{50} : 5-6 years (complete mineratlization)	Gebrema-riam et al. 2010
Chlorpyriphos (CPF) (I)	SFCW and SSFCW (fiberglass three of each type)	1.0 m (L)x0.6 m (W)x0.6 m (H) Substrate layer for SFCW: 0.1 m; SSFCW: 0.3 m. Water layer depth SFCW: 0.4 m; SSFCW: 0.2 m.	Assay 1: 0 µg/l; Assay 2: 478 µg/l; Assay 3: 589 µg/l; Assay 4: 788 µg/l	Removal efficiency of chlorpyrifos and Dissolved Organic Carbon (DOC) in *P. australis* mesocosms	Removal of CPF (DOC) (%) in SFCW/SSFCW: Assay 2: 94% (DOC 96%)/91% (DOC 92%); Assay 3: 95% (DOC 96%)/93% (DOC 93%); Assay 4: 96% (DOC 94%)/96% (DOC 89%).	Biological degradation; absorption/adsorption into plants roots and biofilm. Adsorption to substrate. Mineralization and possible fermentation	Q: 4.6 ml/min. HRT: 7 days Metabolite 3,5,6-trichloro-2-pyridinol (TCP) measured in influent and effluent of both types of CW.	Agudelo et al. 2010

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Diazinon (I)	Ditches 2 VEG (U and V-shaped) + 1 NONVEG (Vshaped)	U-shape: 3.33 m^3 (3mx3mx0.37 m); V-shaped: 0.26 m^3 (1.8mx0.6mx 0.24m)	$UVEG_{t:oh}$: 580 µg/l; $VVEG_{t:oh}$: 1320µg/l; $VNONVEG_{t:oh}$:1100 µg/l	The use of veg.drainage ditches as BMP	Water phase: UVEG: 38%; VVEG: 38%; VNONVEG: 50%; Sediment: UVEG: 54%; VVEG:57%; VNONVEG: 50%; Plant: UVEG: 8%; VVEG: 5%	High water-solubility Diazinon; Chemical hydrolysis; Microbial degradation; Sedimentation	Duration: 5days; DT_{50}: 4.5-6.4 hours; Log K_{oc}: 1007-1842; Sol_{H2O}: 40 mg/l; halfdistances: (55 m for V-vegetated and 158 m for V-NONVEG)	Moore et al. 2008
Diazinon (I)	High density Poly-ethylene containers. (VEG and NONVEG meso-cosms	1.32 mx0.70 mx0.66 m. Substrate 16 cm silt loam sand above 22 cm of sand. Water depth 5.4-15 cm	Target concen-tration: 20 µg/l (exposu-re for 6 h followed by a 42 h rest period. After this additio-nal 6 h of flushing with unamen-ded well water	Pesticide load decrease after: Initial runoff; after flushing (investigate pesticide sorption to plant); total load decrease	Total load decrease 50 ± 5% (*S. americanum*); 25 ± 47% (*T. latifolia*); 61 ± 7% (*L. oryzoides*). NONVEG mesocosms (28 ± 20%) not significantly different from VEG mesocosms	Mass retention and plant sorption (reversibel)	Plant adaptation period 6 weeks; HRT 6 hours;	Moore et al., 2013
Diazinon	SFCW on a 1.2 ha parcel; contribu-ting watershed 39,000 ha	Wetland channel 280 m (L)x6.5 m (W)x0.3 m (H); L:W ratio 43:1	C_{inlet}: 0.038-0.13 ppb; diazinon decay rate: 0.097-0.289 d^{-1}. (substan-tial because zero not included in confi-dence interval);	Establish a 95% credible interval in decay rate of pesticides by modelling hydrology based on a five parameters model	Removal efficiency based on in- and outlet concentrations between 0-71%	Decay/reten-tion equals hydrolysis, photolysis, biodegrada-tion, sorption and volatilization (not significant for diazinon e.g. low Henry's Law constant)	Q = 100 m^3/d; HRT = 4 days; Spray-drift was not considered in the model. Based on model, an area between 2860 m^2 - 8522 m^2 is required to reach the of 0.1 ppbv (normation) assuming a depth of 0.33 m and 4 tanks:	Krone-Davis et al. 2013

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Dicamba (H)	SFCW; 2 VEG and 1 NONVEG	L(40m)xW(3 m)xH (0.05-0.5m)	Peak conc. (ng/l); Cell 1: 4039;	Influence of vegetation	VEG cell 1: 66%; VEG cell 2: 90%;	Sorption: low; related to low K_{oc} and low OC soil;	Duration: 17 hours; K_{oc} (ml/g): 12; DT_{50}: 40 days	Elsaesser et al. 2011
Dicamba (H)			Cell 2: 5904; NONVEG: 4190	Toxicity reduction (not discussed)	NONVEG cell: 48.3%	Short HRT: Sorption on plant surface; Photolytic decay	HRT cell 1:280 min; HRT cell 2: 330 min; HRT NONVEG cell: 132 min	
Dimethoate (I)	SFCW; 2 VEG and 1 UN-VEG	L(40m)xW(3 m)xH (0.05-0.5m)	Peak conc. phase (ng/l); Cell 1: 158; Cell 2: 18; NONVEG cell: 244	Influence of vegetation; Toxicity reduction; (not discussed)	VEG cell 1: 100%; VEG cell 2: 100%; NONVEG cell: 75%; Sorption to sediment for NONVEG cell: 21 µg of 330 µg added (6.3%)	Sorption to sediment and plants; Photolytic decay	Duration: 17 hours; K_{oc} (ml/g): 30; DT_{50}: 45 days; HRT cell 1:280 min; HRT cell 2: 330 min; HRT NONVEG cell: 132 min	Elsaesser et al. 2011
Diuron (H)	SFCW VEG	0.11 km^2	17-240 ng/l	Long term removal	43-55%	Dependent on: wetland design, HRT, type of vegetation and redox potential	Duration: 7d, 28 days and 34 days over a three years period	Page et al. 2011
Diuron (H)	VEG and NONVEGCW	VEGCW: 200m^2, depth 0.5 m NONVEG-CW: 100 m^2,depth 1m	± 45 µg/l	Influence of vegetation	27 -55%	No influence of Inletconcentra-tion; Wetland aging e.g. higher removal during second season; Uptake by plants;	Duration: 7 to 13 days (DT_{50}: 21.3 ± 4.2 days; Adaptation of microorganisms. Enhanced Rhizosphere degradation	Rose et al. 2006
Endosulfan-α (A/I)	Micro-cosms	0.78 litres (sm); 202 litres (lm)	15-18 µg/l	Substrate; Size microcosm	sm: 97.3%; lm: 96.3%	Oxidation; Sorption; Hydrolysis; Metabolite formation	Duration: 41-50 days; Metabolite (endosulfan sulphate)	Laabs et al. 2007

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Endosulfan-α (A/I)	SSFCW	55 m^2 + gravel bed	100 mg/40 l	Assess behavior in SSFCW	> 90% removed	Biodegradation; Plant uptake	Duration: 450 hours; Log K_{ow}: 3.84	Matamoros et al. 2007
Esenvalerate (I)	Agricul-tural drainage ditch	600 m; 0.27-0.31 m water depth	0.15 mg/l	Assess behavior	Water reduced to 4 µg/l; Sediment: 17 ±12 µg/kg; Plant: 784 ± 426 µg/kg	Sorption by vegetation from the water column	Duration: 112 days; DT_{50} (water): 0.12 days; DT_{50} soil: 9 days; DT_{50} plant: 1.3 days	Cooper et al. 2004
Fluometuron (H)	VEG and NONVEG CW	VEGCW: 200m^2; depth 0.5 m; NONVEG-CW:100 m^2, depth 1m	± 10 µg/l	Influence of vegetation	First incubation: no removal; Second incubation; Veg. pond: 58%; NONVEG pond: 41%	Reduced DT_{50}: 13.8 days to 5.5 days; Possible influence of higher T (second incubation: no enhhanced degradation)	Duration: 7 to 13 days (Ten incubi-tion periods), 2 seasons; DT_{50}: 25.4 ± 8.6 days	Rose et al. 2006
Fluometuron (H)	SFCW	5400 m^2; 2 cells: Shallow (0.3 m) and deep (1 m)	42 ml 0.48 kg/l Cotoran 4L per 1893 litres	Determine environmenttal fate	Shallow cell: 81%; Deeper cell: 58%	Dissipation affected by HRT; higher removal in shallow compared to deeper cell;	Duration: 21 days; Plant uptake, with no preferences	Locke et al. 2011
HCH (I); α-HCH; β-HCH; γ-HCH; δ-HCH	SFCW-VEG	100 ha	α-HCH (ng/l): 1.58 ; β-HCH (ng/l): 4.13 ng/l; γ-HCH(ng/) : 2.16; δ-HCH (ng/l): 3.04	Influence of fraction (f) (fDOC and fPOC) on HCH removal	α-HCH: 6.3%: ffree: 5.7%, fDOC: 0.3%, fPOC: 4.3%; β-HCH: 2.9%: ffree: 3.1%, fDOC: 0.3%, fPOC: 2.8%; γ-HCH: 10. 2% ffree: 9.5%, fDOC: 0.2%, fPOC: 3.7%; δ-HCH: 4.3%: ffree: 3.5%, fDOC: 0.6%, fPOC: 3.0%	Short HRT; Removal related to low hydropho-bicity and low ratios of ffree/fDOC/fPOC	Duration; 3 months; HRT: 34 hours	Luo et al. 2009

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Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Isoproturon (H)	Simulated ditch:; Tilting flume	2.92 m^2	± 20 µg/l	Submergenc; Bed form geometry; Speed water flow on mitigation	Mass increase 30% in substrate with dunes; 3% mass increase for substrate with crenels (rectangular);	Sorption on Hemp substate; Mass of pesticides transferred is larger (± 20%) for crenels than for dunes	Duration: 7 hours;; Sol $_{H2O}$: 70 mg/l; Log K_{ow}: 2.5; DT_{50}: 6-28 days	Boutron et al. 2011
Isoproturon (H)					Increase in speed water flow; Increase (± 7%) in mass pesticides transferred. Increase of submergence increased mass transfer of 11%		form and distance between form must be further investigated	
Lambda-cyhalothrin (I)	Micro-cosms; Veg/ NONVEG	0.0042 m^3; Sediment**:** 10cmWater: 8.0 cm	2.23 kg a.i. /ha; Or mix with lambda-cyhalo-thrin: 0.028 kg a.i./ha/2. 03 ha area;	Uptake of pesticides by vegetation(*J. effuses* and *L.peploides);* reduction of toxicity	Veg microcosm (*J. effuses):* Plant: 1.33 µg/kg (day 7) Veg microcosm*(L. peploides):*Plant: 46.77µg/kg (day 7) highest concentration in sediment of *L. peploides* combined microcosm (136.78 µg/kg*)*	Partitioning to plants and sediment, with highestuptake by *l.peploides*(in microcosms amendedwith Atrazine andcombined microcosms	Duration 56 days; Sol_{H2O}: 0.005 mg/l; Log K_{oc}: 3.37DT_{50} (hydrolysis): 233: Precipitation: 0.64 cm	Bouldin et al. 2005
Lambda-cyhalothrin (I)	SFCW	4.8 ha	(1.5 ng /l)	Removal as a function of particle size;phase partitioning; Dissipation using two sediments	C_{ss}: $1.6x10^5$ ng/kgK_d (L/kg): $8.59x10^4$Reten-tion highest in 0.7-2 µm clay fraction, (± 55%): Pos. correlation particle size + concentration (> 53 µm particles)	Aging sediments; Decreased microbial degradation; Strong sorption to sedimentswith high OC content	DT_{50} (flooded and anaerobic): 156 ± 62 days	Budd et al. 2011

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Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Lambda-cyhalothrin (I)	SFCW1; SFCW2+ (Both VEG); Sediment basin(SB)	CW1: 2.3 ha; CW2: 2.5 ha	1.8 ng/l; 3.3 ng/l; SB: 6.6 ng/l	Efficacy of flow through CW; Evaluate important factors	CW1: 71%; CW2: 90%; SB: 34%	Removal in CW1: part with more vegetation; CW2: larger flow path, more dense vegetation: particle settling; Adsorption;	Duration: irrigation season (3½ months); HRT CW1: 1hour; HRT CW2: 18hours	Budd et al. 2009
Lindane (I)	Micro-cosms	Jars of 300 ml	2 mg/l (amend-ment with and without Algae and Atrazine	Interaction between algae and pesticides	Retention; Water without algae:14%; with algae:7%; Mixture with Atrazine: without algae: 41%; with: 10%	Algae provides:sorption sites, facilitates degradation , acts as photo catalysts; Possible volatilization (high V_p)	Jars placed in growth chamber with algae culture for 11 days	Friesen-Pankratz et al. 2003
Linuron (H)	SFCW VEG	840 (CW1)-1200 (CW2) m^2	$CW1_{2000}$: 22.9 g; $CW1_{2001}$: 7.29 g; CW2: 16.1 g	Retention of pesticides	$CW1_{2000}$: 30%; $CW1_{2001}$: 3%; CW2: 19%	Retention correlates to HRT	Duration:10 months; Sol_{H2O}: 75 mg/l; Log K_{oc}: 2.7; DT_{50}: 82 days	Blankenberg et al. 2006
Linuron (H)	SFCW	CW1: 440 $m^{2;}$ CW2: 880 m^2	Input year 2000: 22.9g; year 2001: 7.2 g	Wetland size; Chemical properties influencing retention	$CW1_{2000}$: 18% $CW1_{2001}$: 0%; $CW2_{2000}$: 30%; $CW2_{2001:}$ 3%	Pos. correlation conductivitywith retention; Neg. correlation DT_{50} and retention	Duration: 19 months; Log K_{oc}: 2.70; DT_{50}: 82 days; Sol_{H2O}: 75 mg/l; Doubling size wetland: 1.8 times: more retention	Haarstad and Braske-rud (2003)

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Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
MCPA-2,4/2-Methyl-4-Chloor Phenoxy Acetic Acid (H)	1 ditch (CW1) and 1 triangle pond (CW2); CW1 received drainage from a 5 ha plot with a 12.5% slope, CW2 received from a 9 ha plot with a 5.0% slope	CW1: 13 m (l)x6 m (W)x1 m; V equals 4 m^3. Wetland covered with 16 different plant species; CW2: 20.5 mx15.5 mx11 m (triangle) and a 22 m long and 1 m large oxbow lake, covered with 17 different plant speciesd	Year 2012-2013: 240 g/ha, Inlet CW1 95.7 mg/ha; CW2 5.8 g/ha, Inlet 206.2 mg/ha; year 2013-2014: inlet 10.5 mg/ha	Effectiveness of two CW during two growing seasons (2012-2013; 2013-2014) for abate-ment of pesticides; effect pesticide properties on removal/retentio n	2012-2013: efficiency CW1 towards MCPA: -67%; year 2013-2014 not measured; efficiency CW2 towards MCPA: -618.5% (2012-2013) and 100% (2013-2014)	Low sorption and retention to substrate and plants. Negative efficiency (- 67%) related to runoff and remobiliza-tion; highest MCPA detected less than 1 month ater treatment and during high rainfall	HRT: CW1 0.25-55 h; CW2 5.6-415 h; Q_{max} CW1: 100 m^3/d/ha; CW2 50 m^3/d/ha.	Vallée et al. 2015b
Mecoprop (H)	SFCW	1 ha, veg shallow and nonveg deep	7.80 ± 3.24 µg/l	Behavior: Organic pollutants in sec. effluent WWTP	Week 1: 79 ± 2%; Week 2: 91 ± 1%	Biochemical aerobic pathways: photodegra-dation (light and T dependent); Low sorption	Duration: 2 weeks; Log K_{ow} : 3.13	Matamoros et al. 2008
Metalaxyl (F)	SFCW	CW1: 440 $m^{2;}$; CW2: 880 m^2	Input 2000: 140.9g; 2001: 6.5 g	Wetland size; Chemical properties influencing retention	$CW1_{2000}$: 12%; $CW1_{2001}$: 6%; $CW2_{2000}$: 41%; $CW2_{2001}$; 11%	Degradation by soil microflora; Retention in soil; Doubling size wetland: *no positive effect*	Duration: 19 months; Log K_{oc}: 2.23: DT_{50} : 80 days; Sol_{H2O}: 8400 mg l^{-1}	Haarstad and Bras-
Metalaxyl (F)	SFCW; VEG	CW1: -840 $m^{2;}$ CW2: 1200 m^2	$CW1_{2000(year)}$: 140.9 g $CW1_{2001}$: 7.29 g; CW2: 3.8 g	Retention of pesticides	$CW1_{2000}$: 41%; $CW1_{2001}$: -11%; CW2: 3%	Sedimentation: compound detected in arable soil; Retention correlates to HRT	Duration:10 months; Sol_{H2O}: 8400 mg/l; Log K_{oc}: 2.23; DT_{50}: 80 days	Blankenberg et al. 2006

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Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Methyl-parathion (MeP) (I)	SFCW; VEG NONVEG	11mx50mx0.2m; length (11m) divided in two CW	(VEGCW): 85.6 g (± 3.2); NONVEG-CW: 63.1 g (± 4.7)	Role of vegetation	After 96 h: MeP not detected in outflow (50 m) of VEGCW; After 10d: 1.94 g in VEGCW (mostly in plant); NONVEGCW: 4.29 g; Water phase of both CW: less than 5% of mass added	Uptake by plants; Dissipationor reactions (volatilization, photodegra-dation, hydrolysis); Biotransfor-mation	Duration; 120 days; Sol_{H2O} : 55 mg/l; K_{oc}: $5{,}1x10^3$; MeP degrades slower in sediment than water and plant	Moore et al. 2007
Metolachlor (H)	SFCW; Meso-cosm; VEG	(59-73)m x14mx0.3 m	73 µg/l and 147 µg/l	Removal; Partitionning; Design	For 73 µg/l: 91%; 6% (plant); For 147 µg/l: 87%; 17% (plant)	Partitioning to plants; Removal from the water phase	Duration: 35 days; DT_{50}water: 8-13 days; DT_{50}plant: 17-61 days	Moore et al. 2001
Metolachlor (H)	SSFCW (14 cells)	12 cells: 4.9 m (L)x1.2 m (W): 5.88 m^2; 2 cells: 2.4 m (L)x4.9 m (W)	Year 1998: 2.39 kg/ha (10 g); Year 1999: 1.19 kg/ha (55 g)	Effect of flow; Pesticide loading); HRT; Vegetation; Depth and surface area	> 86% in VEG cells (HRT ≥ 5.1 days); > HRT >% removal; > loading < HRT: 60% removal, except for 2 NONVEG cells; Effect depth significant:for 45 cm: 76.6% and for 50 cm: 68.9% removed	Sorption to gravel; Plant roots enhance microbial degradation; Enhanced Aerobic degra-dation (from plant roots)	Duration: two years 1998-1999; Note: >: higher than or higher and < means lower	Stearman et al. 2003

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Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Metolachlor rac-metolachlor	Four VEG columns of borosili-cate	Inner diameter 15 cm, H: 65 cm. Substrate layer 5 cm gravel (Ø 1-2 mm) and sand layer 52 cm (Ø 0.40–0.63 mm)	1.8 µM	Mass removal; dissipation; bacterial populations under different environmental conditions	Mean mass removal:29 ± 19%; Dissipation: 8-70% At outlet (60 cm from inlet) higher amount measured than 55 cm from inlet. This is related to preferential flows and root channeling	Dissipation mainly in root part, however uptake by roots not significant. Biodegradation of chiral metolachlor; Enantiomeric fractions of metolachlor found; ESA (Ethane Sulfonic Acid) and oxanillic acid (OXA) degradation product formed.	Duration 112 days; HRT: 9.3 days; Q: 0.33 ml/min. Different microbial communities formed in different segment of column. Microbial mediated processes major contri-butors of degradation. S-enantiomer degraded under influence of specific enzymes and proteins.	Elsayed et al. 2015

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Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Metola-chlor (S-metolachlor S-MET (H) and tracers Bromide (Br), Uranine (Ur) and and tracers Bromide (Br), Uranine (Ur)	Outdoor Subsurface flow construc-ted wetlands	4m (L)x1.8m (W)x52cm (H), Substrate; 40 cm sand (grainsize 0–4 mm) above 12cm gravel (grainsize 4–8mm)	960 g/l S-MET, 87% of the *S*-enantiomer; continuous flow wetland: two time intervals of 14 days with water amended with S-Met and tracers and in between: 14 days interval without pesticide (only water); Batch mode: 4 flood–drain cycles 14 days of saturated conditions with the *S*-MET-tracersmix, followed by 7days of drainage	The impact of batch *versus* continuous-flow modes on the dissipation of the chiral herbicide *S*-metolachlor (*S*-MET) and hydrological tracers	S-MET uptake by plant and sorption to sediment of both types of wetlands: < 5%; Total mass degradation *S*-MET: 89.7 ± 4.1%i (batch mode; 59.5±7.0% (continuous-flow mode) Dissipation of *S*-MET nearly constant during batch operations, not constant in continuous-flow wetland	Plant uptake and sorption; photo- and biodegradation prominent under batch mode and redox dependent. No signifi-cant dissipation for tracers in different modes of wetland. Plant uptake was highest for Br and sediment uptake for SRB, about 1/3 of the supplied amount.	Metolachlor oxanilicacid (MOXA) mainly formed under Batch mode; meto-lachlor ethane sulfo-nicacid (MESA) prevailed under continuous flow mode; enantio-selective degrada-tion of R-enantio-mer (batch mode); Volume-tric loading rate (VLR) does not effect S-Met degra--dation	Maillard et al. 2016

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Perme-thrin: Cis-permethrin Trans-permethrin	Ditches 2 VEG; (U and Vshaped) + 1 NONVEG (Vsha-ped)	U-shaped: 3.33 m^3 (3mx3mx0.37 m); V-shaped: 0.26 m^3 (1.8mx0.6mx 0.24m)	$UVEG_{t:oh}$: 27.0 µg/l; $VVEG_{t:oh}$: 225 µg/l; $VNONVEG_{t:oh}$:117 µg/l; $UVEG_{t:oh}$: 31.6 µg/l; $VVEG_{t:oh}$: 275 µg/l; $VNONVEG_{t:oh}$: 110 µg/l	The use of veg.drainage ditches as BMPs; mass partitioning	Water phase:UVEG: 14%, VVEG: 16%, VNONVEG: 20% Sediment phase: UVEG: 64%, VVEG: 52%, VNONVEG: 80%; Plant: UVEG: 23%, VVEG: 33%	Relative low water solubility; preference forsediment phase or matrix with OC; Rapid sorption to sediment; Uptake by plants; Half-distances in V ditches: 22 m (VVEG) to 50 m (VNONVEG)	Duration: 5 days; DT_{50}: 2.4-4.1 hours; Sol_{H2O} (mg/l): 0.2; Log K_{oc}: 10,471-86,000	Moore et al. 2008
Perme-thrin (I): Cis-permethrin (I), Trans-permethrin (I)	High density Poly-ethylene containers. (VEG and NONVEG meso-cosms	1.32 mx0.70 mx0.66 m. Substrate 16 cm silt loam sand above 22 cm of sand. Water depth 5.4-15 cm	Target concentr ation: 10 µg/l (expo-sure for 6 h followed by a 42 h rest period. After this additio-nal 6 h of flushing with unamen-ded well water	Pesticide load decrease after initial runoff and flushing; pesticide sorption to plant) and total load decrease	Total load decrease Cis-permethrin 85 ± 11% (*S. americanum*); 87 ± 1% (*T. latifolia*); 85 ± 10% (*L. oryzoides*). NONVEG mesocosms (73 ± 7%) not sig. different Trans-permethrin 78 ± 3% (*S. americanum*); 88 ± 5% (*T. latifolia*); 88 ± 5% (*L. oryzoides*). NONVEG mesocosms (68 ± 8%) sig.different	Mass retention and plant sorption (irreversibel) for both Cis- and Trans - permethrin	Plant adaptation period 6 weeks; HRT 6 hours;	Moore et al., 2013

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Perme-thrin (I): Cis-perme-thrin (I), Trans-perme-thrin (I)	VEGSFCW Mesocoms divided in an east and west part	Each section 227 m^2 with emergent aquatic plants e.g. *P.australis* and *J. effuses;* East CW V=110,880-258,720 l, slope 0.92%; West CW V=99,330-247,170, slope 0.79%l	Treat-ment (without nutrients) with pesticides (atrazine and S-meto-lachlor): 0.133 g a.i. Perme-thrin. CW amended for 4h at 103 l/min; Treat-ment: Pesticide mixed with nutrients: 0.133 g a.i. Perme-thrin mixed with 357 g N and 409 g P. CW amended for 4h at 103 l/min. V total in inflow: 24,777 l in both sections.	% pesticide dissipation without nutrients mixture and with nutrients mixture; Uptake by CW sediment; DT_{50} of pesticides	Dissipation with and without nutrients comparable: 98-100% within 48 h and dependent on plant density and time and distance from inflow (the latter only applicable for treatment with nutrients)	Biodegradation; Nutrient dissipation with mixture of pesticides independent of plant density; Sorption to plant surfaces, algae, DOC and SS; Very low sorption to CW sediment	DT_{50} Cis-permethrin: 2-7.9 h; Trans-permethrin 1.4-8.1 h for for treatment without nutrients. With nutrients: Cis-permethrin 0.8-15 h; Trans-permethrin 0.5-7.5 h	Lizotte et al. 2014

APPENDIX 1 CONTINUED

Table a: Literature review of removal/retention of pesticides in wetland treatment systems

Pesticide (Type)	Type wetland	Dimension	Concen-tration/ Mass added	Studied parameters	Removal/ Retention (%)	Removal mechanisms	Other specifications	Reference
Simazine (H)	SSFCW	55 m^2 + gravel bed	100 mg/40 l	Assess behavior in SSFCW	20% (water phase); 2% adsorbed to gravel	Log K_{ow}: 1-4; Possible uptake by plants	Duration: 450 hours: Log K_{ow}: 2.2	Matamoros et al. 2007
Simazine (H)	SSFCW (14 cells)	12 cells: 4.9 m (L)x1.2 m (W): 5.88 m^2; 2 cells: 2.4 m (L)x4.9 m (W)	Year 1998: 4.78 kg/ha (220 g); Year 1999: 2.39 kg/ha (110 g)	Effect of flow (pesticide loading); HRT; Vegetation; Depth and Surface area	> 90% in VEG.cells, (HRT > 13.3 days); < 80% in NONVEG cells; > HRT >% removal; VEG cells removed 20.6% more than NONVEG cells	Sorption to gravel; Plant roots enhance microbial degradation; Enhanced aerobic degradation	Duration: two years 1998-1999; No significant effect depth and surface area	Stearman et al. 2003

Appendix 2: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%)

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

Block	***Tribal name***	***Scientific name***	***Abundance***
1	Moko moko	Araceae *Montrichardia arborescens*	25
	Busi papaya	Cecropiaceae Sp.	10
		Convolvulaceae Sp.	10
	Common reed	Poaceae *Phragmites australis*	30
	Yorka pesi	Fabaceae Sp.(Bohinia)	10
	Herb	Onagraceae *Ludwigia stolonifera*	15
2	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	10
	Moko moko	Araceae *Montrichardia arborescens*	30
A	Common reed	Poaceae *Phragmites australis*	30
	Antroewa	Solanaceae Sp.	20
	Boulanger	Solanaceae Sp.	10
3	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	10
	Common reed	Poaceae *Phragmites australis*	40
		Araceae *Pistia stratiotis*	20
	Pankoekoe	Nymphaeaceae Nymphaea sp.	30
4		Araceae *Pistia stratiotis*	10
	Common reed	Poaceae *Phragmites australis*	40
	Moko moko	Araceae *Montrichardia arborescens*	20
	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	10
	Herb	Onagraceae *Ludwigia stolonifera*	20
5	Yorka pesi	Fabaceae Sp.	40
		Minispermaceae Sp.	30
	Bolomakka	Solanaceae Sp.	30
6	Common reed	Poaceae *Phragmites australis*	40
	Moko moko	Araceae *Montrichardia arborescens*	20
	Pankoekoe	Nymphaeaceae *nymphaea sp.*	30
	Yorka pesi	Fabaceae Sp.	5
	Herb	Onagraceae *Ludwigia stolonifera*	5
7	Brede bong	Moraceae	10
	Common reed	Poaceae *Phragmites australis*	40

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Moko moko	Araceae *Montrichardia arborescens*	30
	Dagoe blad.	Convolvulaceae *Ipomoea aquatica*	20
8	Herb	Onagraceae *Ludwigia stolonifera*	50
	Common reed	Poaceae *Phragmites australis*	25
	Moko moko	Araceae *Montrichardia arborescens*	25
9	Moko moko	Araceae *Montrichardia arborescens*	5
	Herb	Onagraceae *Ludwigia stolonifera*	5
	Dagoe blad	Convolvulaceae *Ipomoea aquatica*	5
	Common reed	Poaceae *Phragmites australis*	75
	Pikin fowroe njang	Cyperaceae Sp.	10
10	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	20
	Pankoekoe	Nymphaeaceae Nymphaea sp.	10
	Common reed	Poaceae *Phragmites australis*	30
	Common reed	Poaceae *Phragmites australis*	20
	Dagoe blad	Convolvulaceae *Ipomoea aquatica*	20
11	Herb	Onagraceae *Ludwigia stolonifera*	40
	Paardegras	Poaceae Sp.	40
	Common reed	Poaceae *Phragmites australis*	20
12		*Pistia stratiotis*	20
	Moko moko	Araceae *Montrichardia arborescens*	25
	Common reed	Poaceae *Phragmites australis*	25
	Pankoekoe	Nymphaeaceae *Nymphaea sp.*	30
13	Paardegras	Poaceae *Sp.*	40
	Moko moko	Araceae *Montrichardia arborescens*	40
	Switie bonkie	Fabaceae *Sp*.	10
	Algae		10
14	Moko moko	Araceae *Montrichardia arborescens*	40
	Papaja	Caricaceae *Carica papaya*	5
	Pokai tongo	Heliconiaceae *Heliconia sp.*	20
	Busi papaja	Cecropiaceae *Sp*.	5
	Tabaka tiki varen		30
15	Moko moko	Araceae *Montrichardia arborescens*	90
	Mango trees	Anacardiaceae *Mangifera indica*	10
16	Common reed	Poaceae *Phragmites australis*	40
	Dagoeblad	Convolvulacaea *Ipomoea aquatica*	20

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Herb	Onagraceae *Ludwigia stolonifera*	10
	Pokai tongo	Heliconiaceae *Heliconia Sp*.	20
	Chinese tayer	Aracaea *Sp.*	10
17	Moko moko	Araceae *Montrichardia arborescens*	80
	Herb	Onagraceae *Ludwigia stolonifera*	
	Common reed	Poaceae *Phragmites australis*	5
	Dagoe blad	Convolvulacaea *Ipomoea aquatica*	
	Grass	Cyperaceae *Sp*.	
	Popkie grasie	Cyperaceae *Cyperus ligularis*	
18	Treated with herbicides		Not assessed
19	Boesi papaja	Cecropiaceae *Sp.*	<5
	Common reed	Poaceae *Phragmites australis*	70
	Blaka uma	Boraginaceae *Sp*.	10
	Moko moko	Araceae *Montrichardia arborescens*	10
	Sunflower	Asteraceae *Sp.*	<10
20	Pankoekoe	Nymphaeaceae *Nymphaea sp.*	10
	Dagoe blad	Convolvulacaea *Ipomoea aquatica*	5
21	Herb	Onagraceae *Ludwigia stolonifera*	10
	Moko moko	Araceae *Montrichardia arborescens*	80
	Common reed	Poaceae *Phragmites australis*	10
22	Pankoekoe	Nymphaeaceae *Nymphaea sp.*	5
	Dagoeblad	Convolvulacaea *Ipomoea aquatica*	10
23	Common reed	Poaceae *Phragmites australis*	70
	Herb	Onagraceae *Ludwigia stolonifera*	15
	Pankoekoe	Nymphaeaceae *Nymphaea sp.*	15
24	Common reed	Poaceae *Phragmites australis*	40
	Tafra bong	Boraginacea *Cordia Sp*.	20
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Grass	Gramineae *Sp.*	20
	Gado dede	Commelinaceae *Sp.*	10
25	Dagoe blad	Convolvulaceae *Ipomoea aquatica*	70
	Eendekroos	Lemnaceae *Lemna minor*	20
	Algae		10
26	Common reed	Poaceae *Phragmites australis*	50
	Grass	Gramineae *Sp*.	50

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

27	Common reed	Poaceae *Phragmites australis*	85
	Moko moko	Araceae *Montrichardia arborescens*	5
	Herb	Onagraceae *Ludwigia stolonifera*	<5
	Liemsweed	Asteraceae *Sp*.	<5
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	<5
	pankoekoe	Nymphaeaceae *Nymphaea sp.*	<5
28	Veelwortelig kroos	Lemnaceae Spirodela polyrhiza	50
	Common reed	Poaceae *Phragmites australis*	30
	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	20
29	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	10
	Common reed	Poaceae *Phragmites australis*	50
	Herb	Onagraceae *Ludwigia stolonifera*	5
	Moko moko	Araceae *Montrichardia arborescens*	10
	Yorka pesi	Fabaceae *Sp*.	5
	Eendekroos	Lemnaceae *Lemna minor*	10
	Common reed	Poaceae *Phragmites australis*	10
30	Common reed	Poaceae *Phragmites australis*	20
	Moko moko	Araceae *Montrichardia arborescens*	10
	Yorka pesi	Fabaceae *Sp*.	20
	Busi papaja	Cecropiacea *Sp*.	20
	Gado dede	Commelinaceae *Sp*.	10
	Soekroe tanta	Asteraceae *Sp*.	10
	Herb	Onagraceae *Ludwigia stolonifera*	10
31	Common reed	Poaceae *Phragmites australis*	10
	Moko moko	Araceae *Montrichardia arborescens*	80
	Yorka pesi	Fabaceae *Sp*.	<5
	Pikin fowroe ngang	Cyperaceae *Sp*.	<5
	Switi bonki	Fabaceae *Sp*.	<5
32	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	70
	Waterplant (varenachtig)	Pteridaceae *Ceratpteris cornuta*	10
	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	10
	Pistia	Araceae *Pistia stratiotes*	5
	Amandelboom	Combreataceae *Sp*.	5
33	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	5
34	Moko moko	Araceae *Montrichardia arborescens*	5

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Pankoekoe	Nymphaeaceae *Nymphaea sp.*	10
35	Eendekroos	Lemnaceae *Lemna minor*	70
	Pankoekoe	Nymphaeaceae *Nymphaea sp.*	10
	Moko moko	Araceae *Montrichardia arborescens*	10
	Common reed	Poaceae *Phragmites australis*	10
36	Pankoekoe	Nymphaeaceae *Nymphaea sp.*	60
	Eendekroos	Lemnaceae *Lemna minor*	30
	Herb	Onagraceae *Ludwigia stolonifera*	5
	Common reed	Poaceae *Phragmites australis*	5
37	Common reed	Poaceae *Phragmites australis*	10
	Yorka pesi	Fabaceae *Sp*.	40
	Papaja	Caricaceae *Carica papaya*	20
	Gado dede	Commelinaceae *Sp*.	10
38	Grass	Gramineae *Sp*.	30
	Liemsweed.	Asteraceae *Wedelia trilobata*	10
	Common reed	Poaceae *Phragmites australis*	10
	Herb	Onagraceae *Ludwigia stolonifera*	5
	Yorka pesi	Fabaceae *Sp*.	30
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	5
	Grass	Gramineae *Sp*.	5
	Moko moko	Araceae *Montrichardia arborescens*	5
39	Cleaned ditch		ND
40	Moko moko	Araceae *Montrichardia arborescens*	5
	Grass	Gramineae *Sp*.	20
41	Moko moko	Araceae *Montrichardia arborescens*	20
	Eendekroos	Lemnaceae *Lemna minor*	20
	Common reed	Poaceae *Phragmites australis*	50
	Herb	Onagraceae *Ludwigia stolonifera*	10
42	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	80
	Common reed	Poaceae *Phragmites australis*	10
	Moko moko	Araceae *Montrichardia arborescens*	5
	Liemsweed	Asteraceae *Sp*.	<5
	Cassava	Euphorbiaceae *Sp*	<5
	Herb	Onagraceae *Ludwigia stolonifera*	<5
43	Eendekroos	Lemnaceae *Lemna minor*	60

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Common reed	Poaceae *Phragmites australis*	15
	Herb	Onagraceae *Ludwigia stolonifera*	<5
	Cassava	Euphorbiaceae *Sp*.	<5
	Dagoeblad	Convolvulaceae *Ipomoea sp.*	10
	Gras	Cyperaceae sp.	<5
	Waternavel	Hydrocotyle sp.	10
44		Convolvulaceae Sp.	
	Common reed	Poaceae *Phragmites australis*	20
	Moko moko	Araceae *Montrichardia arborescens*	40
	Cassava	Euphorbiaceae *Sp*.	10
	Eendekroos	Lemnaceae *Lemna minor*	20
	Switi bonki	Fabaceae *Sp*.	10
	Gadodede	Commelinaceae *Sp.*	5
	Bamboe		5
45	Gado dede	Commelinaceae *Sp.*	10
	Herb	Onagraceae *Ludwigia stolonifera*	20
	Tafra bong	Boraginaceae *Cordia sp.*	10
	Boesi papaja	Cecropiaceae *Sp.*	10
	Soekroe tanta	Asteraceae *Sp.*	30
	Blaka uma	Boraginaceae *Macrostachya sp.*	10
	Bolo makka	Solanaceae *Sp*.	10
46	Moko moko	Araceae *Montrichardia arborescens*	30
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Gado dede	Commelinaceae Sp.	20
	Eendekroos	Lemnaceae *Lemna minor*	40
47	Eendekroos	Lemnaceae *Lemna minor*	50
	Gado dede	Commelinaceae *Sp*.	20
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Moko moko	Araceae *Montrichardia arborescens*	20
48	Eendekroos	Lemnaceae *Lemna minor*	60
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Moko moko	Araceae *Montrichardia arborescens*	20
	Gado dede	Commelinaceae *Sp*.	10
49	Moko moko	Araceae *Montrichardia arborescens*	40
	Pankoekoe	Nymphaeaceae *Nymphaea sp*	15

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Herb	Onagraceae *Ludwigia stolonifera*	10
	Gado dede	Commelinaceae *Sp*.	10
	Common reed	Poaceae *Phragmites australis*	15
	Yorka pesi	Fabaceae *Sp*.	10
50	Crasi taya	Araceae *Sp*.	10
	Common reed	Poaceae *Phragmites australis*	30
	Soekroe tanta	Asteraceae *Sp*.	20
	Sunflower	Asteraceae *Sp*.	30
	Moko moko	Araceae *Montrichardia arborescens*	10
51	Common reed	Poaceae *Phragmites australis*	40
	Moko moko	Araceae *Montrichardia arborescens*	30
	Araceae Sp.	Araceae *Montrichardia linifera*	30
52	Common reed	Poaceae *Phragmites australis*	40
	Moko moko	Araceae *Montrichardia arborescens*	30
	Araceae Sp.	Araceae *Montrichardia linifera*	30
53	Soekroe tanta	Asteraceae *Sp*.	30
	Common reed	Poaceae *Phragmites australis*	40
	Araceae Sp.	Araceae *Montrichardia linifera*	30
54	Sunflower	Asteraceae *Sp*.	40
	Moko moko	Araceae *Montrichardia arborescens*	30
	Araceae Sp.	Araceae *Montrichardia linifera*	30
55	Moko moko	Araceae *Montrichardia arborescens*	60
	Common reed	Poaceae *Phragmites australis*	30
	Wilkensbita	Apocynaceae *Mandevilla hirsuta*	<5
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	5
	Herb	Onagraceae *Ludwigia stolonifera*	<5
56	Moko moko	Araceae *Montrichardia arborescens*	60
	Common reed	Poaceae *Phragmites australis*	30
	Switi bonki	Leguminoceae *Sp*.	5
	Waterhyacinth	Pontederiaceae *Sp*.	<5
57	Moko moko	Araceae *Montrichardia arborescens*	70
	Tafra bong	Boraginaceae *Cordia sp.*	<5
	Bita wiwiri	Solananceae *Cestrum latifolium*	<5
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	5
	Cassave	Euphorbiaceae *Sp*.	10

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Cocos noot	Arecaceae *Cocos nucifera*	
	Common reed	Poaceae *Phragmites australis*	10
58	`	Nelumbonaceae *nelumbo sp.*	20
	Eendekroos	Lemnaceae *Lemna minor*	20
	Common reed	Poaceae *Phragmites australis*	50
	Cocosnoot	Arecaceae *Cocos nucifera*	5
	Algae		5
59	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	10
	Water hyacinth	Pontederiaceae *Sp*.	10
	Dagoe blad	Convolvulacaea *Ipomoea aquatica*	70
	Moko moko	Araceae *Montrichardia arborescens*	10
60	Common reed	Poaceae *Phragmites australis*	10
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	10
	Varen sp.		20
	Moko moko	Araceae *Montrichardia arborescens*	10
	Water navel	Apiaceae *Hydrocotyle umbellata*	10
	Dagoeblad	Convolvulacaea *Ipomoea aquatica*	10
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	20
61	Herb	Onagraceae *Ludwigia stolonifera*	20
	Moko moko	Araceae *Montrichardia arborescens*	20
	Common reed	Poaceae *Phragmites australis*	5
	Grass	Cyperaceae *Sp*.	<5
	Waternavel	Apiaceae *Hydrocotyle umbellata*	5
	Grass	Cyperaceae *Sp*.	5
	Varen sp.	Pteridophyta *Sp*.	<5
	Dagoeblad	Convolvulacaea *Ipomoea aquatica*	20
	Watra kanoe	Acanthaceae *Ruellia tuberosa*	<5
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	<5
	Herb	Hydrophyllaceae *Hydrolea sp.*	5
	Pistia	Araceae *Pistia stratiotes*	10
62	Dagoeblad	Convolvulacaea *Ipomoea aquatica*	20
	Paardegras	Poaceae *Sp.*	10
	Common reed	Poaceae *Phragmites australis*	50
	Moko moko	Araceae *Montrichardia arborescens*	10

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Cassave	Euphorbiaceae *Manihot sp.*	<5
	Herb	Onagraceae *Ludwigia stolonifera*	<5
	Plant (blue flower)	Hydrophyllaceae *Hydrolea sp.*	<5
	Waternavel	Apiaceae *Hydrocotyle umbellata*	5
63	Common reed	Poaceae *Phragmites australis*	50
	Waternavel	Apiaceae *Hydrocotyle umbellata*	20
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	5
	Plant (blue flower)	Hydrophyllaceae *Hydrolea sp.*	5
	Eendekroos	Lemnaceae *Lemna minor*	5
	Liemsweed	Asteraceae *Wedelia trilobata*	<5
	Moko moko	Araceae *Montrichardia arborescens*	5
		Pontederiaceae *Sp*.	<5
64	Waterhyacinth	Pontederiaceae *Sp*.	30
	Common reed	Poaceae *Phragmites australis*	30
	Dagoe blad	Convolvulacaea *Ipomoea aquatica*	20
	Algae		20
65	Typha	Typhaceae *Typha angustifolia*	40
	Common reed	Poaceae *Phragmites australis*	5
	Moko moko	Araceae *Montrichardia arborescens*	5
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Waternavel	Apiaceae *Hydrocotyle umbellata*	10
	Pankoekoe	Nelumbonaceae *nelumbo sp.*	20
	Eendekroos	Lemnaceae *Lemna minor*	5
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	5
66	Veel wortelig kroos	Lemnaceae *Spirodela polyrhiza*	5
	Pankoekoe	Nelumbonaceae *nelumbo sp.*	5
	Typha	Typhaceae *Typha angustifolia*	60
	Waternavel	Apiaceae *Hydrocotyle umbellata*	10
	Moko moko	Araceae *Montrichardia arborescens*	5
	Baboen nefi	Cyperaceae *Sp*.	5
	Common reed	Poaceae *Phragmites australis*	5
	Paardegras	Poaceae *Sp*.	5
67	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	5
	Gras	Cyperaceae *Sp.*	5
	Moko moko	Araceae *Montrichardia arborescens*	5

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Herb	Onagraceae *Ludwigia stolonifera*	40
	Common reed	Poaceae *Phragmites australis*	20
	Gras	Cyperaceae *Sp*.	10
	Pistia	Araceae *Pistia stratiotes*	5
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	5
	Waternavel	Apiaceae *Hydrocotyle umbellata*	5
68	Dagoe blad	Convolvulaceae *Ipomoea aquatica*	10
	Herb	Onagraceae *Ludwigia stolonifera*	5
	Pistia	Araceae *Pistia stratiotes*	15
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	70
69	Common reed	Poaceae *Phragmites australis*	20
	Moko moko	Araceae *Montrichardia arborescens*	40
	Blaka oema	Boraginaceae *Sp*.	20
	Yorka pesi	Fabaceae *Sp*.	10
	Herb	Onagraceae *Ludwigia stolonifera*	5
	Waternavel	Apiaceae *Hydrocotyle umbellata*	5
70	Herb	Onagraceae *Ludwigia stolonifera*	10
	Common reed	Poaceae *Phragmites australis*	10
	Dagoeblad	Convolvulaceae *Ipomoea aquatica*	10
	Moko moko	Araceae *Montrichardia arborescens*	40
	Waternavel	Apiaceae *Hydrocotyle umbellata*	5
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	5
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	20
71	Moko moko	Araceae *Montrichardia arborescens*	20
	Typha	Typhaceae *Typha angustifolia*	60
	Common reed	Poaceae *Phragmites australis*	10
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	10
72	Typha	Typhaceae *Typha angustifolia*	60
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	20
	Common reed	Poaceae *Phragmites australis*	20
73	Waternavel	Apiaceae *Hydrocotyle umbellata*	5
	Herb	Onagraceae *Ludwigia stolonifera*	<5
	Veelwortelig kroos	Lemnaceae *Spirodela polyrhiza*	40
	Moko moko	Araceae *Montrichardia arborescens*	<5
	Dagoe blad	Convolvulaceae *Ipomoea aquatica*	10

Table b: Main vegetation found in two main drainage ditches in the Alkmaar region with their estimated abundance (%). Block 1-5: drainage ditch left; Block 6-10: drainage ditch right; Block 11-15: drainage ditch left; Block 16-20: drainage ditch right; Block 71-75: drainage ditch left

	Eendekroos	Lemnaceae *Lemna minor*	40
74	Paarde gras	Poaceae *Echinochloa polystachya*	60
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Plant (blue flower)	Hydrophyllaceae *Hydrolea sp.*	20
	Plant (blue flower)	Hydrophyllaceae *Hydrolea sp.*	20
	Waternavel	Apiaceae *Hydrocotyle umbellata*	10
75	Moko moko	Araceae *Montrichardia arborescens*	10
	Typha	Typhaceae *Typha angustifolia*	50
	Paardegras	Poaceae *Echinochloa polystachya*	20
	Herb	Onagraceae *Ludwigia stolonifera*	10
	Plant (blue flower)	Hydrophyllaceae *Hydrolea sp.*	10
76	Herb	Onagraceae *Ludwigia stolonifera*	10
	Plant (blue flower)	Hydrophyllaceae *Hydrolea sp.*	10
	Water navel	Apiaceae *Hydrocotyle umbellata*	60
	Moko moko	Araceae *Montrichardia arborescens*	10
	Tabaka tiki varen	Polypodiaceae *Acrostichum aureum*	10

Appendix 3: Results obtained for the lineair regression of LN concentration versus time for chapter 5

lineair regression of LN concentrations versus time for chapter 5

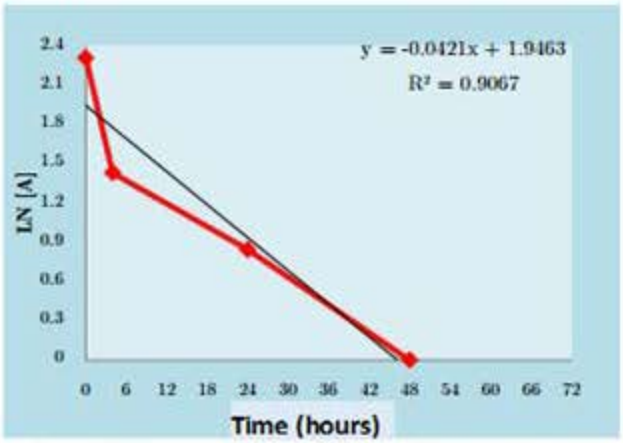

Figure a: LN concentration lambda-cyhalothrin against time, mesocosm 1, batch 1

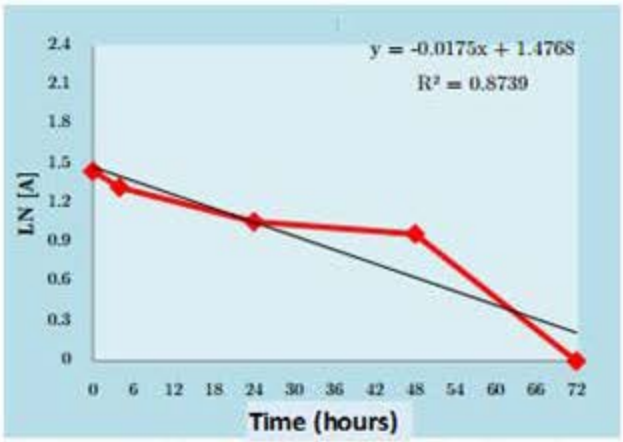

Figure b: LN concentration lambda-cyhalothrin against time, mesocosm 2, batch 1

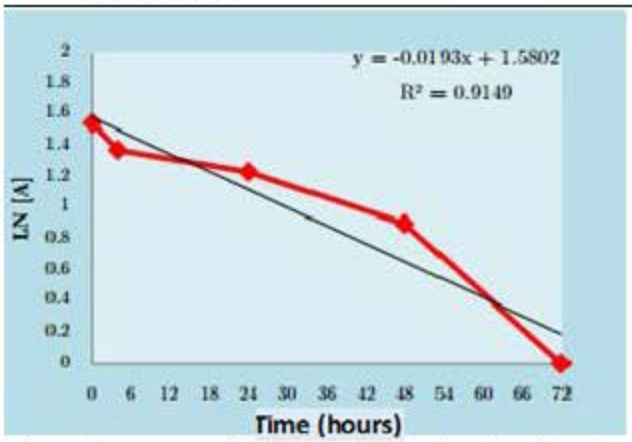

Figure c: LN concentration lambda-cyhalothrin against time, mesocosm 5, batch 1

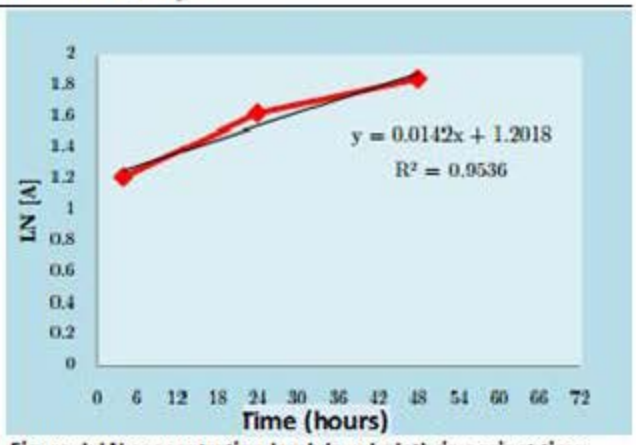

Figure d: LN concentration lambda-cyhalothrin against time, mesocosm 6, batch 1

lineair regression of LN concentrations versus time for chapter 5

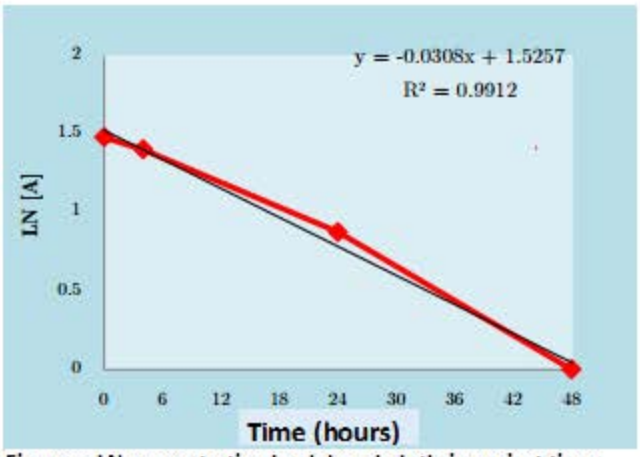

Figure e: LN concentration lambda-cyhalothrin against time, mesocosm 1. batch 2

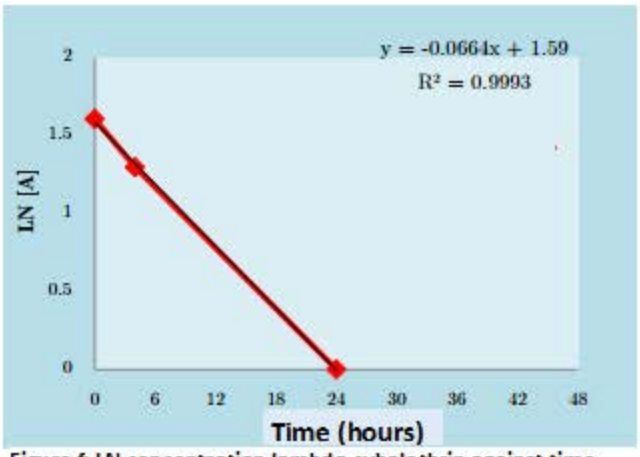

Figure f: LN concentration lambda-cyhalothrin against time, mesocosm 2, batch 2

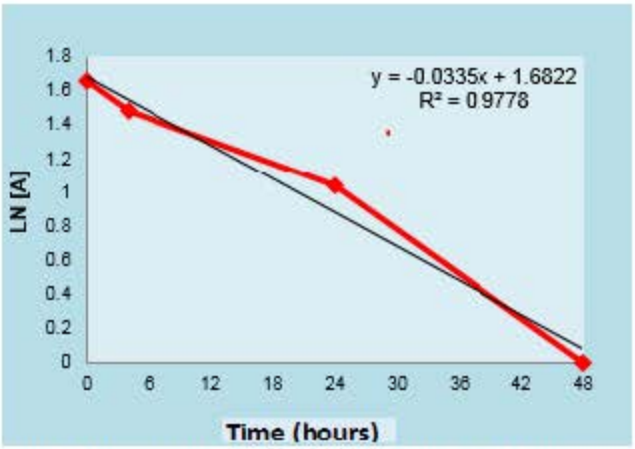

Figure g: LN concentration lambda-cyhalothrin against time, mesocosm 5, batch 2

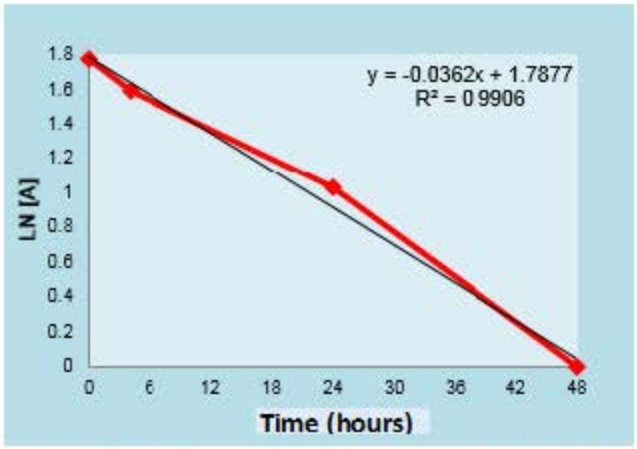

Figure h: LN concentration lambda-cyhalothrin against time, mesocosm 6, batch 2

lineair regression of LN concentrations versus time for chapter 5

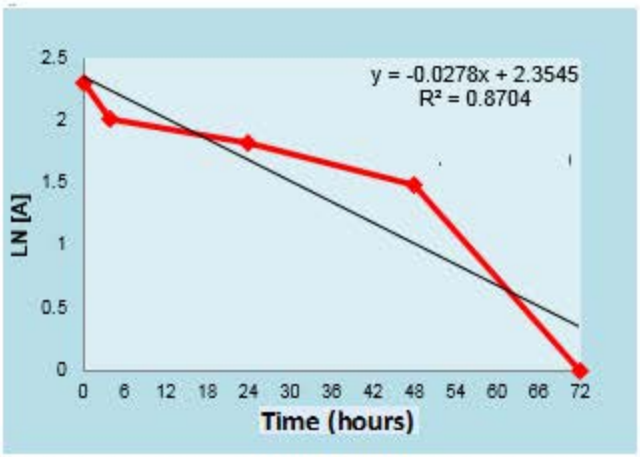

Figure i: LN concentration lambda-cyhalothrin against time, mesocosm 1, batch 3

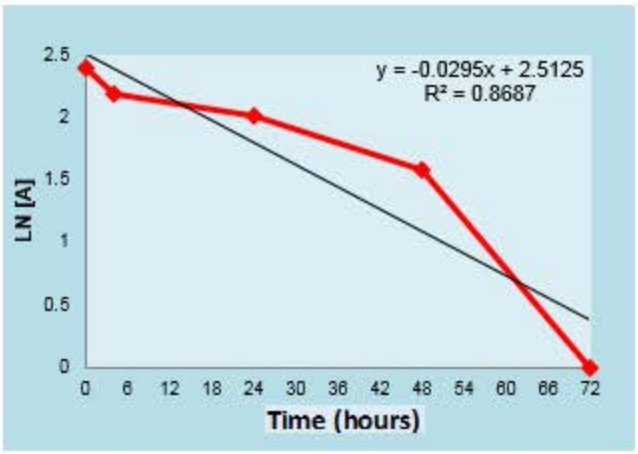

Figure j: LN concentration lambda-cyhalothrin against time, mesocosm 2, batch 3

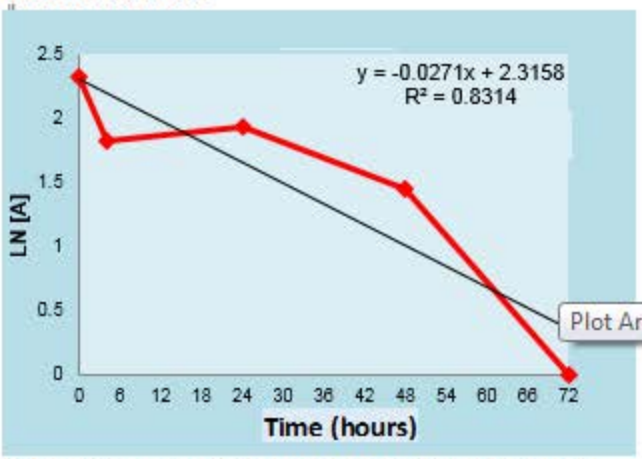

Figure k: LN concentration lambda-cyhalothrin against time, mesocosm 5, batch 3

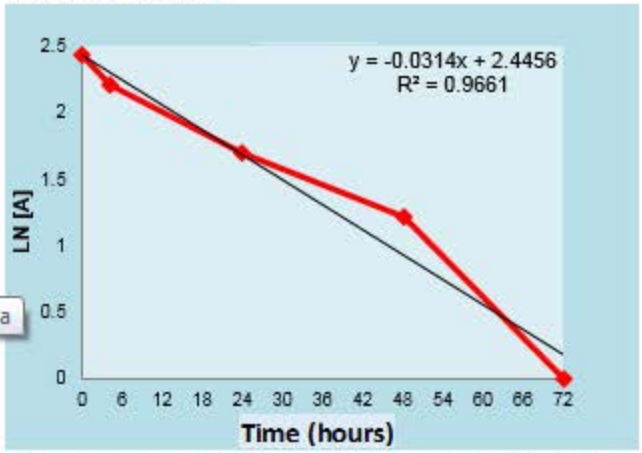

Figure l: LN concentration lambda-cyhalothrin against time, mesocosm 6, batch 3

lineair regression of LN concentrations versus time for chapter 5

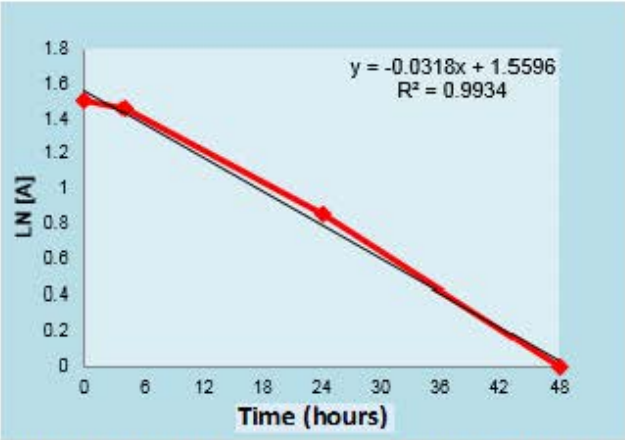

Figure m: LN concentration lambda-cyhalothrin against time, mesocosm 1, batch 4

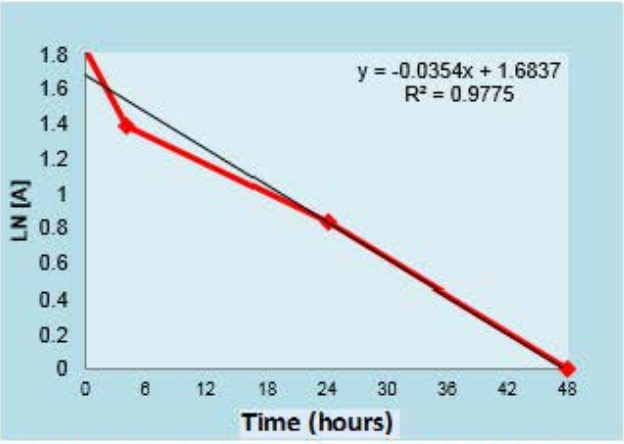

Figure n: LN concentration lambda-cyhalothrin against time, mesocosm 2, batch 4

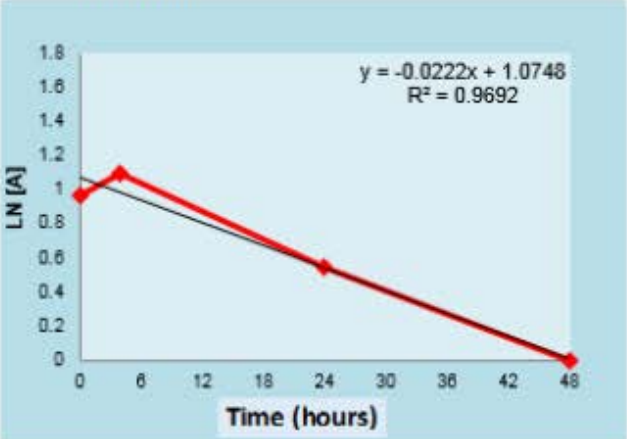

Figure o: LN concentration lambda-cyhalothrin against time, mesocosm 5, batch 4

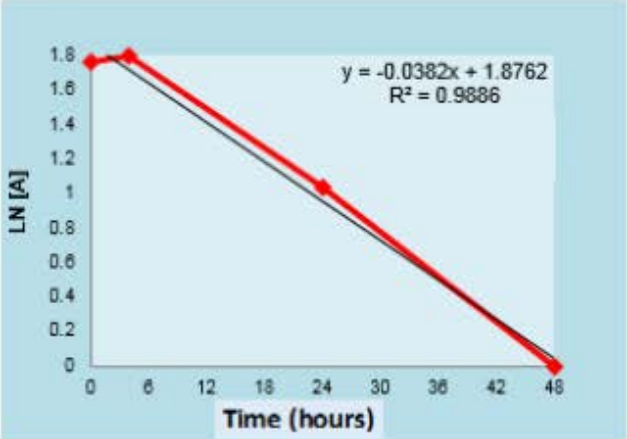

Figure p: LN concentration lambda-cyhalothrin against time, mesocosm 6, batch 4

lineair regression of LN concentrations versus time for chapter 5

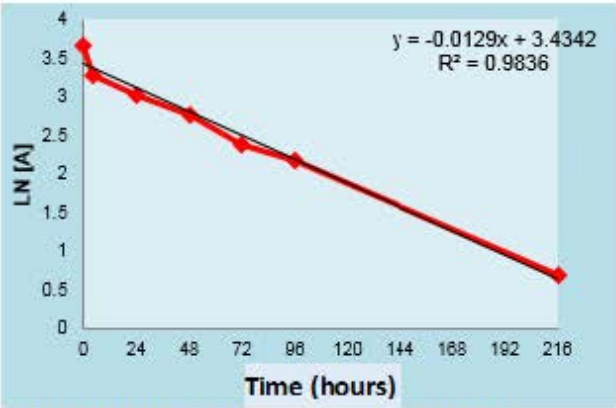

Figure A: LN concentration imidacloprid against time, mesocosm 3, batch 1

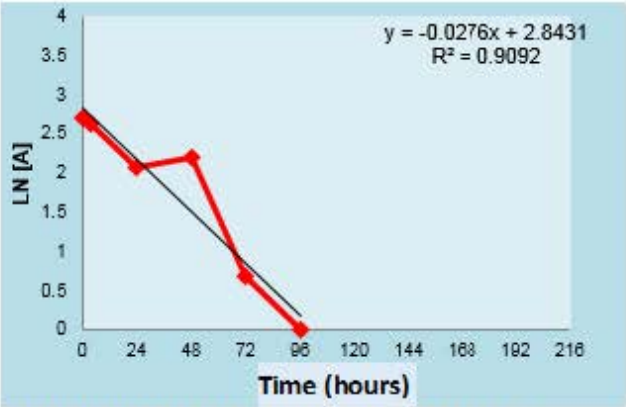

Figure B: LN concentration imidacloprid against time, mesocosm 4, batch 1

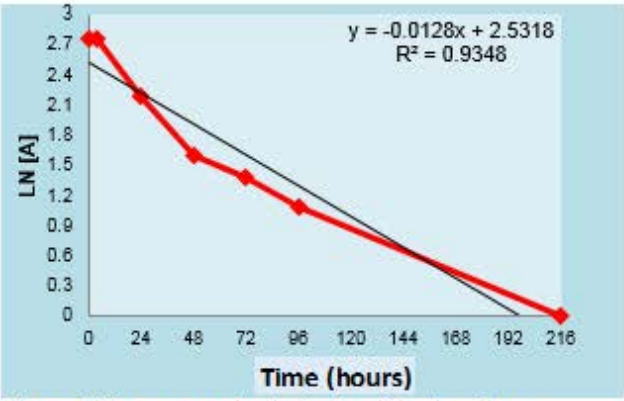

Figure C: LN concentration imidacloprid against time, mesocosm 7, batch 1

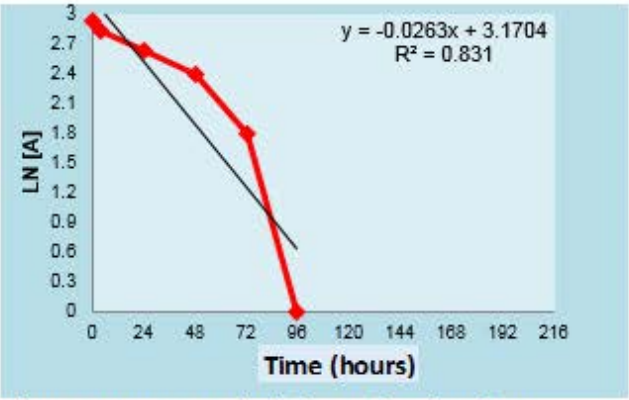

Figure D: LN concentration imidacloprid against time, mesocosm 8, batch 1

lineair regression of LN concentrations versus time for chapter 5

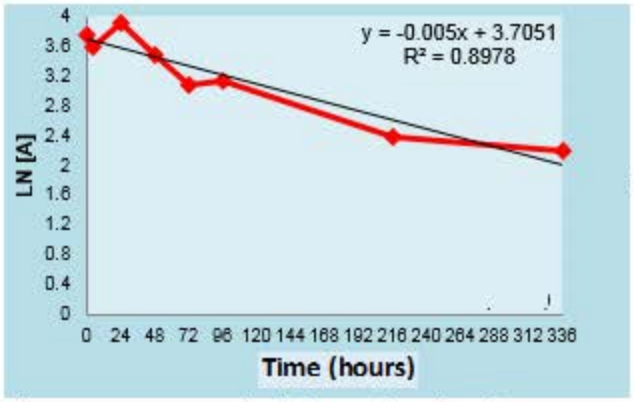

Figure E: LN concentration imidacloprid against time, mesocosm 3, batch 2

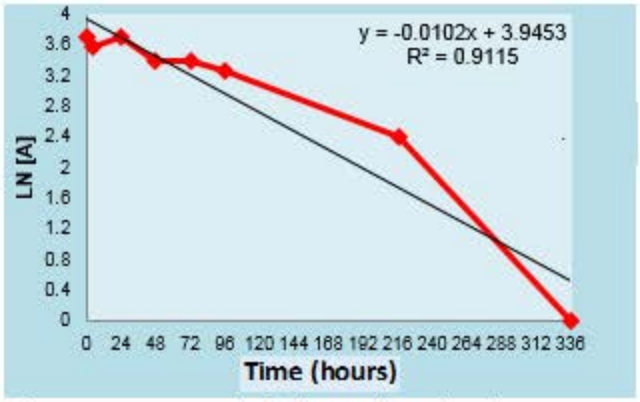

Figure F: LN concentration imidacloprid against time, mesocosm 4, batch 2

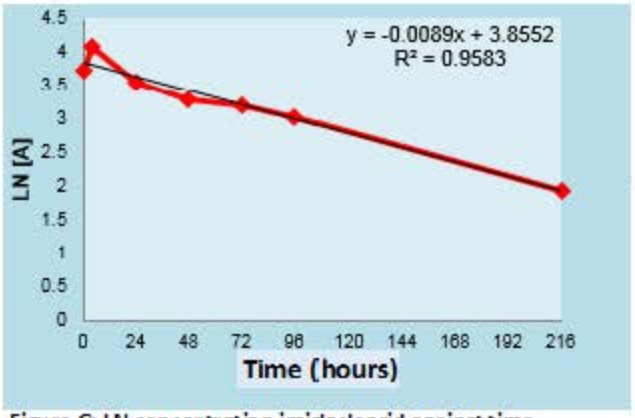

Figure G: LN concentration imidacloprid against time, mesocosm 7, batch 2

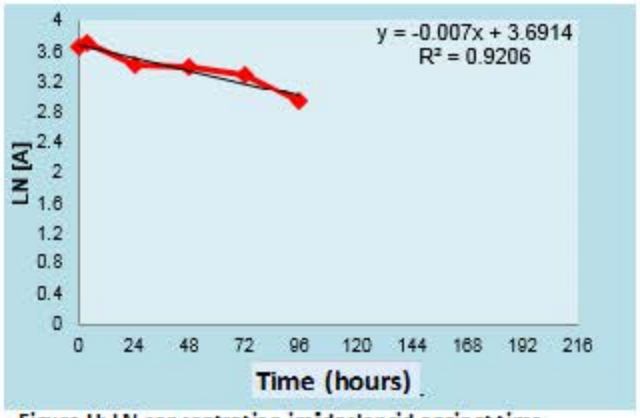

Figure H: LN concentration imidacloprid against time, mesocosm 8, batch 2

lineair regression of LN concentrations versus time for chapter 5

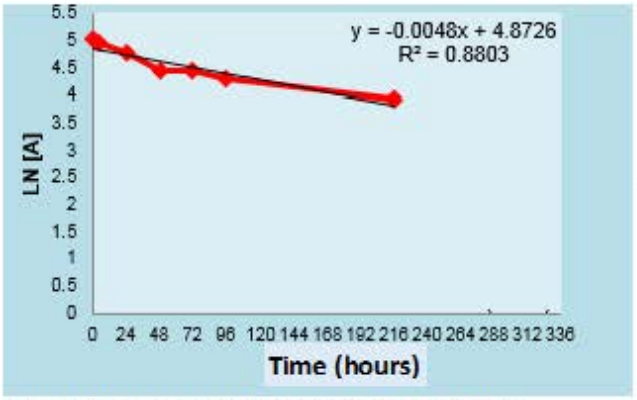

Figure I: LN concentration imidacloprid against time, mesocosm 3, batch 3

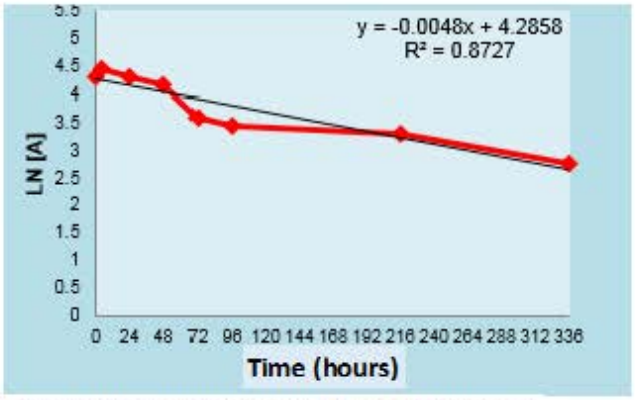

Figure J: LN concentration imidacloprid against time, mesocosm 4, batch 3

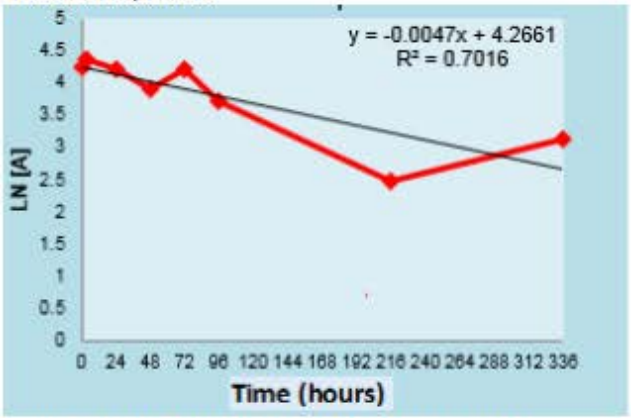

Figure K: LN concentration imidacloprid against time, mesocosm 7, batch 3

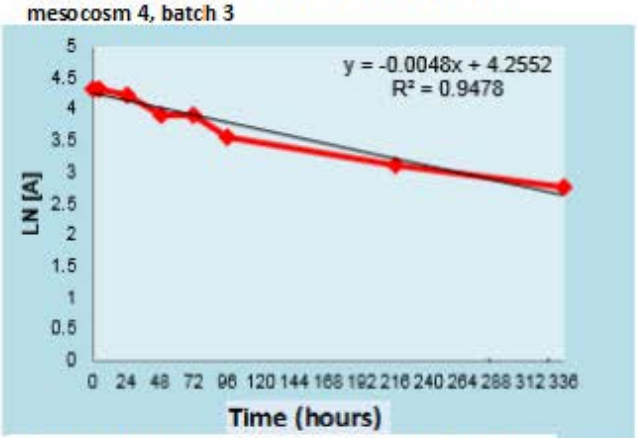

Figure L: LN concentration imidacloprid against time, mesocosm 8, batch 3

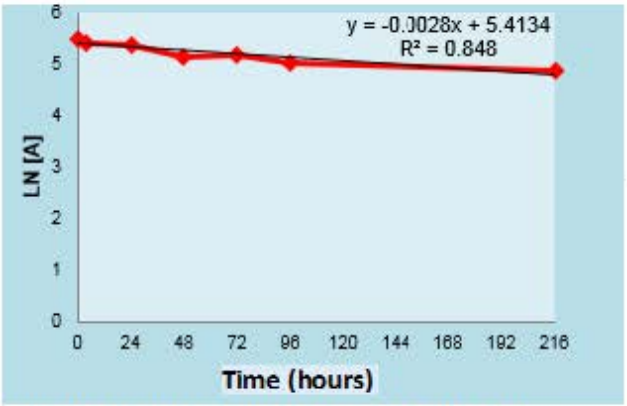

Figure M: LN concentration imidacloprid against time, mesocosm 3, batch 4

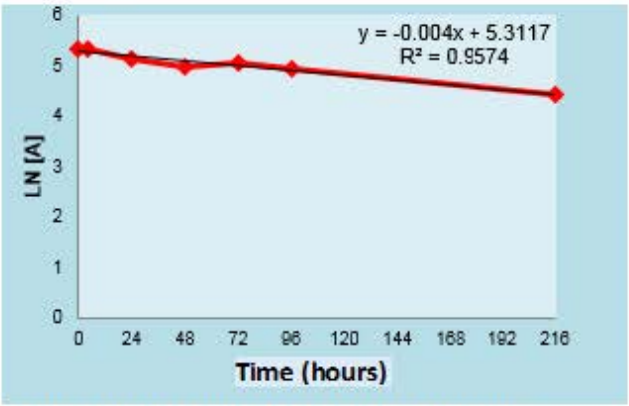

Figure N: LN concentration imidacloprid against time, mesocosm 4, batch 4

lineair regression of LN concentrations versus time for chapter 5

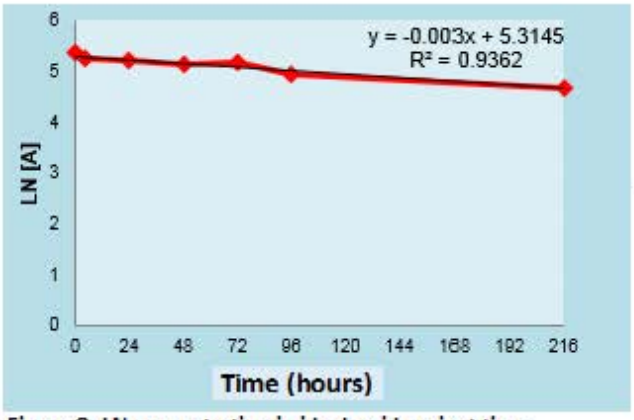

Figure O: LN concentration imidacloprid against time, mesocosm 7, batch 4

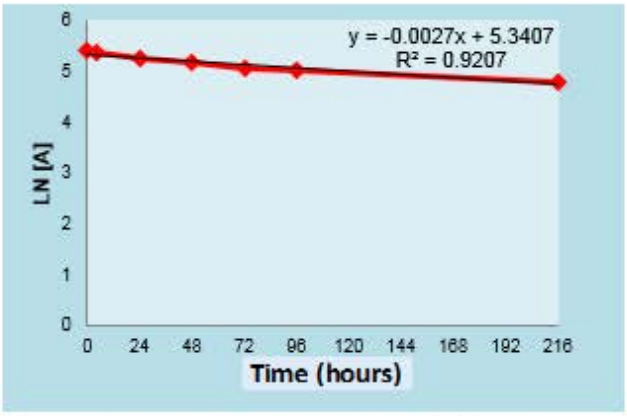

Figure P: LN concentration imidacloprid against time, mesocosm 8, batch 4

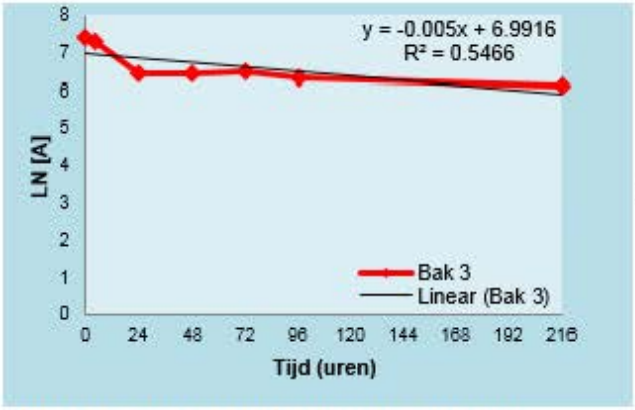

Figure Q: LN concentration imidacloprid against time, mesocosm 3, batch 5

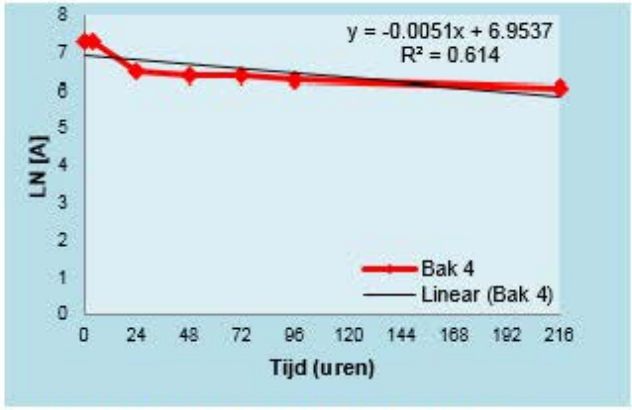

Figure R: LN concentration imidacloprid against time, mesocosm 4, batch 5

lineair regression of LN concentrations versus time for chapter 5

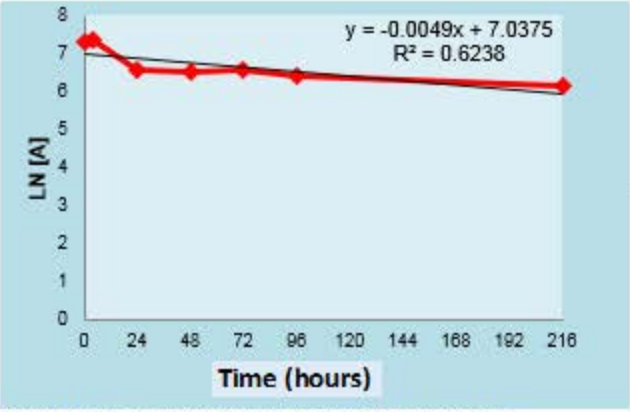

Figure S: LN concentration imidacloprid against time, mesocosm 7, batch 5

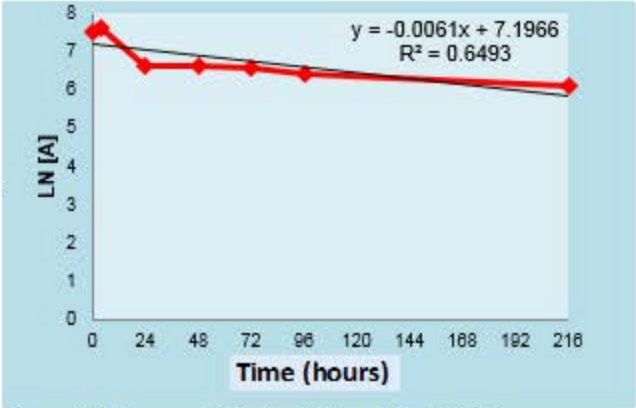

Figure T: LN concentration imidacloprid against time, mesocosm 8, batch 5

Appendix 4: Statistical outputs of different tests performed

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

Data of chapter 3 (G.A.P. Survey), used for different statistical tests

Age Class code[*1]	age class (years)	gloves	boots	Protective cloth	storage of cloth	washing of cloth	type of Education[*2]
a	< 25	2	3	4	4	4	p
a	< 25	1	1	3	1	1	s
b	25 - 40	2	4	4	2	1	s
b	25 - 40	4	4	2	4	4	s
b	25 - 40	2	4	4	4	4	p
b	25 - 40	4	4	2	1	2	n
b	25 - 40	4	4	4	4	4	s
b	25 - 40	4	4	4	3	4	p
b	25 - 40	4	4	4	2	4	t
b	25 - 40	4	4	4	4	4	p
b	25 - 40	4	4	4	4	4	p
b	25-40	2	4	4	4	4	s
b	25-40	4	4	3	4	4	s
b	25-40	3	3	3	4	4	s
b	25-40	2	4	4	4	4	s
b	25-40	4	4	4	4	4	p
b	25-40	2	4	4	4	4	s
c	41 - 60	2	4	4	4	4	s
c	41 - 60	2	4	4	4	1	p
c	41 - 60	4	4	4	4	1	s
c	41 - 60	2	4	4	4	2	n
c	41 - 60	2	3	4	4	4	s
c	41 - 60	4	4	4	4	4	s
c	41 - 60	3	4	4	4	4	s
c	41 - 60	2	4	4	4	4	p
c	41 - 60	2	4	4	4	4	p
c	41 - 60	2	4	4	4	4	p
c	41 - 60	3	4	4	4	3	p
c	41 - 60	4	4	4	4	4	s
c	41 - 60	2	4	4	4	3	p
c	41 - 60	1	1	0	1	1	s
c	41 - 60	2	4	4	4	4	s

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

Data of chapter 3 (G.A.P. Survey), used for different statistical tests							
Age Class code[*1]	age class (years)	gloves	boots	Protective cloth	storage of cloth	washing of cloth	type of Education[*2]
c	41 - 60	4	4	4	4	4	p
c	41 - 60	2	4	4	4	4	p
c	41 - 60	4	4	4	4	4	s
c	41 - 60	4	4	4	2	4	p
c	41 - 60	2	4	4	4	4	p
c	41 - 60	2	4	4	4	4	s
c	41 - 60	2	4	4	4	4	s
c	41 - 60	2	4	4	4	4	n
c	41 - 60	2	4	4	4	4	n
c	41 - 60	2	4	4	4	4	p
c	41 - 60	3	4	4	4	4	s
c	41 - 60	4	4	4	4	4	p
c	41 - 60	4	4	4	4	4	s
c	41 - 60	2	4	4	4	4	p
c	41 - 60	4	4	4	4	4	p
c	41 - 60	2	4	4	4	3	s
c	41 - 60	4	4	4	4	4	p
c	41 - 60	4	4	4	4	3	s
c	41 - 60	4	4	4	4	3	p
c	41 - 60	4	4	4	4	4	p
c	41 - 60	2	2	4	2	4	s
c	41 - 60	1	4	4	3	4	p
c	41 - 60	1	4	1	4	4	p
c	41 -60	4	4	4	4	4	p
c	41-60	2	4	4	4	4	p
c	41-60	2	4	4	4	4	p
c	41-60	2	4	4	4	4	s
c	41 - 60	4	4	4	4	4	s
c	41 -60	4	4	4	4	4	p
c	41-60	4	4	4	4	4	s
c	41-60	2	4	4	4	4	p
c	41-60	2	4	4	4	4	n
c	41-60	2	4	4	1	1	s

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

Data of chapter 3 (G.A.P. Survey), used for different statistical tests							
Age Class code[*1]	age class (years)	gloves	boots	Protective cloth	storage of cloth	washing of cloth	type of Education[*2]
c	41-60	2	4	3	2	2	p
d	> 60	4	4	4	4	4	n
d	> 60	4	4	4	3	2	p
d	> 60	3	4	4	4	4	p
d	> 60	2	4	4	4	4	p
d	> 60	4	4	4	4	4	p
d	> 60	4	4	4	4	4	p
d	>60	4	4	3	4	4	n
d	> 60	1	4	4	4	4	s
d	> 60	4	4	4	4	4	p
d	> 60	4	2	4	4	4	p
d	> 60	2	4	4	2	2	p

***[1]: a; age class < 25 years; b: 25-40 years; c: 41-60 years and d: >60 years**

***[2]: p: primary level or grades 5-12 years; s: secondary level or grades 12-19/20 years; n: no education and t: tertiary level of education**

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:37:38 PM

Data source: Data 1 in Notebook8

Dependent Variable: gloves

Group	N	Missing	Median	25%	75%
a	2	0	1.500	1.000	2
b	15	0	4.000	2	4.000
c	50	0	2	2	4.000
d	11	0	4.000	2.250	4.000

H = 8.662 with 3 degrees of freedom. (P = 0.034)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = 0.034)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

All Pairwise Multiple Comparison Procedures (Dunn's Method):

Comparison	Diff of Ranks	Q	P<0.05
d vs a	34.818	1.999	No
d vs c	11.558	1.532	Do Not Test
d vs b	0.0848	0.00943	Do Not Test
b vs a	34.733	2.036	Do Not Test
b vs c	11.473	1.720	Do Not Test
c vs a	23.260	1.423	Do Not Test

Note: The multiple comparisons on ranks do not include an adjustment for ties.

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:50:20 PM

Data source: Data 1 in Notebook8

Dependent Variable: boots

Group	**N**	**Missing**	**Median**	**25%**	**75%**
a	2	0	2	1.000	3.000
b	15	0	4.000	4.000	4.000
c	50	0	4.000	4.000	4.000
d	11	0	4.000	4.000	4.000

H = 20.899 with 3 degrees of freedom. (P = <0.001)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = <0.001)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

All Pairwise Multiple Comparison Procedures (Dunn's Method):

Comparison	**Diff of Ranks**	**Q**	**P<0.05**
c vs a	36.890	2.258	No
c vs d	1.231	0.163	Do Not Test
c vs b	0.107	0.0160	Do Not Test
b vs a	36.783	2.156	Do Not Test
b vs d	1.124	0.125	Do Not Test
d vs a	35.659	2.047	Do Not Test

Note: The multiple comparisons on ranks do not include an adjustment for ties.

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:51:50 PM

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

Data source: Data 1 in Notebook8

Dependent Variable: protcloth

Group	N	Missing	Median	25%	75%
a	2	0	3.500	3.000	4.000
b	15	0	4.000	3.250	4.000
c	50	0	4.000	4.000	4.000
d	11	0	4.000	4.000	4.000

H = 7.148 with 3 degrees of freedom. (P = 0.067)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.067)

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:52:38 PM

Data source: Data 1 in Notebook8

Dependent Variable: storagecloth

Group	N	Missing	Median	25%	75%
a	2	0	2.500	1.000	4.000
b	15	0	4.000	3.250	4.000
c	50	0	4.000	4.000	4.000
d	11	0	4.000	4.000	4.000

H = 3.881 with 3 degrees of freedom. (P = 0.275)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.275)

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:53:49 PM

Data source: Data 1 in Notebook8

Dependent Variable: washingcloth

Group	N	Missing	Median	25%	75%
a	2	0	2.500	1.000	4.000
b	15	0	4.000	4.000	4.000

c	50	0	4.000	4.000	4.000
d	11	0	4.000	4.000	4.000

H = 1.980 with 3 degrees of freedom. (P = 0.577)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.577)

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:54:33 PM

Data source: Data 1 in Notebook8

Dependent Variable: gloves

Group	N	Missing	Median	25%	75%
p	40	0	2.500	2	4.000
s	30	0	2	2	4.000
n	7	0	2	2	4.000
t	1	0	4.000	4.000	4.000

H = 1.626 with 3 degrees of freedom. (P = 0.654)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.654)

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:55:12 PM

Data source: Data 1 in Notebook8

Dependent Variable: boots

Group	N	Missing	Median	25%	75%
p	40	0	4.000	4.000	4.000
s	30	0	4.000	4.000	4.000
n	7	0	4.000	4.000	4.000
t	1	0	4.000	4.000	4.000

H = 3.787 with 3 degrees of freedom. (P = 0.285)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.285)

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:55:47 PM

Data source: Data 1 in Notebook8

Dependent Variable: protcloth

Group	N	Missing	Median	25%	75%
p	40	0	4.000	4.000	4.000
s	30	0	4.000	4.000	4.000
n	7	0	4.000	3.250	4.000
t	1	0	4.000	4.000	4.000

H = 4.405 with 3 degrees of freedom. (P = 0.221)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.221)

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:56:26 PM

Data source: Data 1 in Notebook8

Dependent Variable: storagecloth

Group	N	Missing	Median	25%	75%
p	40	0	4.000	4.000	4.000
s	30	0	4.000	4.000	4.000
n	7	0	4.000	4.000	4.000
t	1	0	2	2	2

H = 4.976 with 3 degrees of freedom. (P = 0.174)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.174)

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 06, 2013, 3:57:16 PM

Data source: Data 1 in Notebook8

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

Dependent Variable: washingcloth

Group	N	Missing	Median	25%	75%
p	40	0	4.000	4.000	4.000
s	30	0	4.000	4.000	4.000
n	7	0	4.000	2.500	4.000
t	1	0	4.000	4.000	4.000

H = 1.139 with 3 degrees of freedom. (P = 0.768)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.768)

Summary table for all statistical tests performed for result of the G.A.P. survey (Chapter 3)

Gloves

Group	N	Median	25%	75%
a	2	1.500	1.000	2
b	15	4.000	2	4.000
c	50	2 2	4.000	
d	11	4.000	2.250	4.000

H = 8.662 with 3 degrees of freedom. (P = 0.034): no significant differences (Dunn's Method).

Group	N	Median	25%	75%
p	40	2.500	2	4.000
s	30	2	2	4.000
n	7	2	2	4.000
t	1	4.000	4.000	4.000

H = 1.626 with 3 degrees of freedom. (P = 0.654)

Boots

Group	N	Median	25%	75%
a	2	2	1.000	3.000
b	15	4.000	4.000	4.000
c	50	4.000	4.000	4.000
d	11	4.000	4.000	4.000

H = 20.899 with 3 degrees of freedom.
(P = <0.001): no significant differences (Dunn's Method)

Group	N	Median	25%	75%
p	40	4.000	4.000	4.000
s	30	4.000	4.000	4.000
n	7	4.000	4.000	4.000
t	1	4.000	4.000	4.000

H = 3.787 with 3 degrees of freedom. (P = 0.285)

Prot. cloth

Dependent Variable: prot.cloth

Group	N	Median	25%	75%
a	2	3.500	3.000	4.000
b	15	4.000	3.250	4.000
c	50	4.000	4.000	4.000
d	11	4.000	4.000	4.000

H = 7.148 with 3 degrees of freedom. (P = 0.067)

Group	N	Median	25%	75%
p	40	4.000	4.000	4.000
s	30	4.000	4.000	4.000
n	7	4.000	3.250	4.000
t	1	4.000	4.000	4.000

H = 4.405 with 3 degrees of freedom. (P = 0.221)

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 3

Summary table for all statistical tests performed for result of the G.A.P. survey (Chapter 3)

Stor.cloth

Dependent Variable: stor.cloth

Group	N	Median	25%	75%
a	2	2.500	1.000	4.000
b	15	4.000	3.250	4.000
c	50	4.000	4.000	4.000
d	11	4.000	4.000	4.000

H = 3.881 with 3 degrees of freedom. (P = 0.275)

Group	N	Median	25%	75%
p	40	4.000	4.000	4.000
s	30	4.000	4.000	4.000
n	7	4.000	4.000	4.000
t	1	2	2	2

H = 4.976 with 3 degrees of freedom. (P = 0.174)

Wash.cloth

Dependent Variable: wash.cloth

Group	N	Median	25%	75%
a	2	2.500	1.000	4.000
b	15	4.000	4.000	4.000
c	50	4.000	4.000	4.000
d	11	4.000	4.000	4.000

H = 1.980 with 3 degrees of freedom. (P = 0.577)

Group	N	Median	25%	75%
p	40	4.000	4.000	4.000
s	30	4.000	4.000	4.000
n	7	4.000	2.500	4.000
t	1	4.000	4.000	4.000

H = 1.139 with 3 degrees of freedom. (P = 0.768)

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

TEST PERFORMED: TWO-WAY ANALYSIS OF VARIANCE PERFORMED FOR THE DEPENDENT VARIABLE CONCENTRATION/REMOVAL EFFICIENCY AND FACTORS BATCHES (DOSE APPLIED) AND TIME (CHAPTER 5)

Lambda-cyhalothrin	Batches	p value at α level of 0.05	Statistical output
***N. amazonum* mesocosms**	Batch1 *compared with* Batch2	0.089	No significant difference in concentration profile
		0.475	No significant difference in removal efficiency.
	Batch1 *compared with* Batch3	0.035	Significant difference in concentration profile
		0.088	No significant difference in removal efficiency.
	Batch1 *compared with* Batch4	0.697	No significant difference in concentration profile
		0.233	No significant difference in removal efficiency.
	Batch2 *compared with* Batch3	0.035	Significant difference in concentration profile
		0.210	No significant difference in removal efficiency.
	Batch2 *compared with* Batch4	0.100	No significant difference in concentration profile
		0.228	No significant difference in removal efficiency.
	Batch3 *compared with* Batch4	0.039	Significant difference in concentration profile
		0.299	No significant difference in removal efficiency.
***E. mutata* mesocosms**	Batch1 *compared with* Batch2	0.453	No significant difference in concentration profile
		0.258	No significant difference in removal efficiency.
	Batch1 *compared with* Batch3	0.136	No significant difference in concentration profile
		0.119	No significant difference in removal efficiency.
	Batch1 *compared with* Batch4	0.256	No significant difference in concentration profile
		0.649	No significant difference in removal efficiency.
	Batch2 *compared with* Batch3	0.040	Significant difference in concentration profile
		0.490	No significant difference in removal efficiency.
	Batch2 *compared with* Batch4	0.949	No significant difference in concentration profile
		0.160	No significant difference in removal efficiency.
	Batch3 *compared with* Batch4	0.062	No significant difference in concentration profile
		0.337	No significant difference in removal efficiency.
Target concentration batch1 and 2 is 10 µg/l **Target concentration van batch3 and 4 is 30 µg/l**			

TEST PERFORMED: TWO-WAY ANALYSIS OF VARIANCE PERFORMED FOR THE DEPENDENT VARIABLE REMOVAL EFFICIENCY AND FACTORS PLANT TYPE AND TIME (CHAPTER 5)

Lambda-cyhalothrin	**Batches**	**p value at α level of 0.05**	**Statistical output**
N. amazonum* compared to *E. mutata	Batch1	0.679	No significant difference in concentration profile.
		0.100	No significant difference in removal efficiency
	Batch2	0.518	No significant difference in concentration profile.
		0.342	No significant difference in removal efficiency
	Batch3	0.199	No significant difference in concentration profile.
		0.081	No significant difference in removal efficiency

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

Lambda-cyhalothrin	Batches	p value at α level of 0.05	Statistical output
	Batch4	0.488	No significant difference in concentration profile.
		0.177	No significant difference in removal efficiency
Target concentration of batch1 and batch2 is 10 µg/l **Target concentration of batch3 and 4 is 30 µg/l**			

TEST PERFORMED: TWO-WAY ANALYSIS OF VARIANCE PERFORMED FOR THE DEPENDENT VARIABLE CONCENTRATION/REMOVAL EFFICIENCY AND FACTORS BATCHES (DOSE APPLIED) AND TIME (CHAPTER 5)

Imidacloprid	Batches	p value at α level of 0.05	Statistical output
***N. amazonum* mesocosms**	Batch1 *compared with* Batch2	< 0.001	Significant difference in concentration profile.
		0.008	Significant difference in removal efficiency.
	Batch1 *compared with* Batch3	< 0.001	Significant difference in concentration profile.
		0.001	Significant difference in removal efficiency.
	Batch1 *compared with* Batch4	< 0.001	Significant difference in concentration profile.
		0.006	Significant difference in removal efficiency.
	Batch1 *compared with* Batch5	0.003	Significant difference in concentration profile.
		0.239	No Significant difference in removal efficiency.
	Batch2 *compared with* Batch3	0.001	Significant difference in concentration profile.
		0.896	No Significant difference in removal efficiency.
	Batch2 *compared with* Batch4	< 0.001	Significant difference in concentration profile.
		0.121	No significant difference in removal efficiency.
	Batch2 *compared with* Batch5	0.003	Significant difference in concentration profile.
		0.065	No significant difference in removal efficiency.
	Batch3 *compared with* Batch4	< 0.001	Significant difference in concentration profile.
		0.127	No significant difference in removal efficiency.
	Batch3 *compared with* Batch5	0.004	Significant difference in concentration profile.
		0.046	Significant difference in removal efficiency.
	Batch4 *compared with* Batch5	0.006	Significant difference in concentration profile.
		0.011	Significant difference in removal efficiency.
E. mutatameme soc	Batch1 *compared with*	< 0.001	Significant difference in concentration profile.
		0.006	Significant difference in removal efficiency.

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

Imidacloprid	Batches	p value at α level of 0.05	Statistical output
	Batch2		
	Batch1 *compared with* Batch3	< 0.001	Significant difference in concentration profile.
		0.003	Significant difference in removal efficiency.
	Batch1 *compared with* Batch4	< 0.001	Significant difference in concentration profile.
		0.02	Significant difference in removal efficiency.
	Batch1 *compared with* Batch5	0.003	Significant difference in concentration profile.
		0.328	No significant difference in removal efficiency.
	Batch2 *compared with* Batch3	< 0.001	Significant difference in concentration profile.
		0.658	No significant difference in removal efficiency.
	Batch2 *compared with* Batch4	< 0.001	Significant difference in concentration profile.
		0.138	No significant difference in removal efficiency.
	Batch2 *compared with* Batch5	0.004	Significant difference in concentration profile.
		0.019	Significant difference in removal efficiency.
	Batch3 *compared with* Batch4	< 0.001	Significant difference in concentration profile.
		0.315	No significant difference in removal efficiency.
	Batch3 *compared with* Batch5	0.004	Significant difference in concentration profile.
		0.066	No significant difference in removal efficiency.
	Batch4 *compared with* Batch5	0.007	Significant difference in concentration profile.
		0.022	Significant difference in removal efficiency.
Target concentration of batch1and batch2 is 60 µg/l			
Target concentration of batch3 and 4 is 180 µg/l			
Target concentration of batch5 is 1000 µg/l			

Test performed: Two-Way Analysis of Variance performed for the dependent variable removal efficiency and factors plant type and time

imidacloprid	Batches	p value at α level of 0.05	Statistical output
Nymphaea amazonum* Compared to *Eleocharis mutata	Batch1	0.007	Significant difference in concentration profile.
		0.549	No significant difference in removal efficiency.
	Batch2	0.948	No significant difference in concentration orofile.
		0.586	No significant difference in removal efficiency.

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

imidacloprid	Batches	p value at α level of 0.05	Statistical output
	Batch3	0.008	Significant difference in concentration profile.
		0.314	No significant difference in removal efficiency.
	Batch4	0.705	No significant difference in concentration profile.
		0.452	No significant difference in removal efficiency.
	Batch5	0.022	Significant difference in concentration profile.
		0.048	Significant difference in removal efficiency.
Target concentration of batch1and batch2 is 60 µg/l **Target concentration of batch3 and 4 is 180 µg/l** **Target concentration of batch5 is 1000 µg/l**			

Appendix 4: Statistical analysis for DT_{50} comparison of pesticides in the water phase of different mesocosms (Chapter 5)

Lambda-cyhalothrin DT_{50} values in N. amazonum and E. mutata mesocosms (see Chapter 5). Values in red are outliers

lambda-cyhalothrin (target concentration)	*N. amazonum* mesocosms (M1, M2)			*E. mutata* mesocosms (M5, M6)		
	$k\ (h)^{-1}$	R^2	DT_{50} (h)	$k\ (h)^{-1}$	R^2	DT_{50} (h)
Batch1, M1, M5 (10 µg/l)	0.042	0.9067	16.50	0.019	0.9149	36.48
Replicates, M2, M6 (10 µg/l)	0.018	0.8730	38.51	0.014	0.9536	N.D.[d]
Batch2, M1, M5 (10 µg/l)	0.031	0.9912	22.36	0.034	0.9778	20.39
Replicates, M2, M6 (10 µg/l)	0.066	0.9993	10.50	0.036	0.9906	19.25
Batch3, M1, M5 (30 µg/l)	0.028	0.8704	24.76	0.027	0.8314	25.67
Replicates, M2, M6 (30 µg/l)	0.030	0.8687	23.10	0.031	0.9661	22.36
Batch4, M1, M5 (30 µg/l)	0.032	0.9934	21.66	0.022	0.9692	18.24
Replicates, M2, M6 (30 µg/l)	0.035	0.9775	19.80	0.038	0.9886	18.24
DT_{50} ± SD (h), n=4, Batches 1 and 2	22.0 ± 12.0			25.4 ± 9.6		
DT_{50} ± SD (h), n=4, Batches 3 and 4	22.3 ± 2.1			24.4 ± 5.6		
Average DT_{50} (h)	**23.0**			**22.2**		
Median (h)	**22.05**			**20.39**		
Acceptable DT_{50} range (h)	**12.00-30.35**			**10.84-31.92**		

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

Raw data used for statistical tests of DT_{50} values for lambda-cyhalothrin in the water phase of different mesocosms (Chapter 5)

Batch lambda-cyhalothrin	DT_{50} (hours)	Exposure concentration
1nympha	16.5	con1
1nympha	38.51	con1
2nympha	22.36	con1
2nympha	10.50	con1
3nympha	24.76	con2
3nympha	23.1	con2
4nympha	21.66	con2
4nympha	19.8	con2
1E. mutata	36.48	con1
2E. mutata	20.39	con1
2E. mutata	19.25	con1
3E. mutata	25.67	con2
3E. mutata	22.36	con2
4E. mutata	18.24	con2
4E. mutata	18.24	con2

T-test: Significant differences in DT_{50} values of lambda-cyhalothrin between two types of mesocosms

Saturday, August 13, 2016, 4:39:22 PM

Data source: Data 1 in statisticaloanalysischapter5thesisB

Normality Test: Passed (P = 0.055)

Equal Variance Test: Passed (P = 0.941)

Group Name	**N**	**Missing**	**Mean**	**Std Dev**	**SEM**	
DT50lambdaN.amazonum		8	0	22.149	8.007	2.831
DT50lambdaE. mutata		7	0	22.947	6.525	2.466

Difference -0.798

t = -0.210 with 13 degrees of freedom. (P = 0.837)

95 percent confidence interval for difference of means: -9.028 to 7.431

The difference in the mean values of the two groups is not great enough to reject the possibility that the difference is due to random sampling variability. There is not a statistically significant difference between the input groups (P = 0.837).

Power of performed test with alpha = 0.050: 0.050

The power of the performed test (0.050) is below the desired power of 0.800.

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

Less than desired power indicates you are less likely to detect a difference when one actually exists. Negative results should be interpreted cautiously.

General Linear Model

Dependent Variable: DT50lambda

Normality Test: Passed (P = 0.221)

Equal Variance Test: Passed (P = 0.262)

Source of Variation	DF	SS	MS	F	P
plant-type	1	4.481	4.481	0.0732	0.792
concentration	1	13.920	13.920	0.227	0.643
plant-type x concentration	1	19.603	19.603	0.320	0.583
Residual	11	673.069	61.188		
Total	14	706.615	50.472		

The difference in the mean values among the different levels of plant-type is not great enough to exclude the possibility that the difference is just due to random sampling variability after allowing for the effects of differences in concentration. There is not a statistically significant difference (P = 0.792).

The difference in the mean values among the different levels of concentration is not great enough to exclude the possibility that the difference is just due to random sampling variability after allowing for the effects of differences in plant-type. There is not a statistically significant difference **(P = 0.643).**

The effect of different levels of plant-type does not depend on what level of concentration is present. There is not a statistically significant interaction between plant-type and concentration (p = 0.583)

.

Power of performed test with alpha = 0.0500: for plant-type : 0.0500
Power of performed test with alpha = 0.0500: for concentration: 0.0500
Power of performed test with alpha = 0.0500: for plant-type x concentration: 0.0500

Least square means for plant-type:

Group	Mean	SEM
NYM	22.149	2.766
MUT	23.250	2.987

Least square means for concentration:

Group	Mean	SEM
con1	23.670	2.987
con2	21.729	2.766

Least square means for plant-type x concentration:

Group	Mean	SEM
NYM x con1	21.968	3.911

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

NYM x con2 22.330 3.911
MUT x con1 25.373 4.516
MUT x con2 21.128 3.911

Imidacloprid's DT_{50} values in N. amazonum and E. mutata mesocosms (see Chapter 5). Values in red are outliers

Imidacloprid (target concentration)	***N. amazonum* mesocosms**			***E. mutata* mesocosms**		
	k $(h)^{-1}$	**R^2**	**DT_{50} (h)**	**k $(h)^{-1}$**	**R^2**	**DT_{50} (h)**
Batch1 (60 µg/l)	0.013	0.9836	53.3	0.013	0.9348	53.3
	0.028	0.9092	24.7	0.026	0.831	26.7
Batch2 (60 µg/l)	0.005	0.8978	139	0.009	0.9583	77.0
	0.010	0.9115	69.3	0.007	0.9206	99.0
Batch3 (180 µg/l)	0.005	0.8803	139	0.005	0.7016	139
	0.005	0.8727	139	0.005	0.9478	139
Batch4 (180 µg/l)	0.003	0.848	231	0.003	0.9362	231
	0.004	0.9574	173	0.003	0.9207	231
Batch5 (1000 µg/l)	0.005	0.5466	139	0.005	0.6238	139
	0.005	0.614	139	0.006	0.6493	116
DT_{50} ± SD (h), n=4, Batches 1 and 2	71.5 ± 48.4			64.0 ± 31.1		
DT_{50} ± SD (h), n=4, Batches 3 and 4	170.4 ± 43.6			184.8 ± 53.4		
DT_{50}, n=2, Batch5	139			127		
Average DT_{50} (h)	**125**			**125**		
Median (h)	**139**			**127.5**		
Acceptable DT_{50} range (h)	**8.3-217**			**0-224**		

Raw data used for statistical tests of DT_{50} values for imidacloprid in the water phase of different mesocosms (Chapter 5)

Batch	**DT_{50}**	**plant Type**	**conc imi**	**Batch(batchA =batch1, batchB=batch2 etcetera).**
1.0000	53.3000	NYM	con1	A
1.0000	24.7000	NYM	con1	A
1.0000	53.3000	MUT	con1	A
1.0000	26.7000	MUT	con1	A
2.0000	139.0000	NYM	con1	B
2.0000	69.3000	NYM	con1	B
2.0000	77.0000	MUT	con1	B
2.0000	99.0000	MUT	con1	B
3.0000	139.0000	NYM	con2	C
3.0000	139.0000	NYM	con2	C
3.0000	139.0000	MUT	con2	C
3.0000	139.0000	MUT	con2	C
4.0000	173.0000	NYM	con2	D
5.0000	139.0000	NYM	con3	E
5.0000	139.0000	NYM	con3	E
5.0000	139.0000	MUT	con3	E

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5
5.0000 116.0000 MUT con3 E

Two Way Analysis of Variance Friday, December 02, 2016, 10:29:15 PM

Data source: Data 1 in statisticaloanalysischapter5thesis (data without outliers)

General Linear Model

Dependent Variable: DT50

Normality Test: Passed (P = 0.054)

Equal Variance Test: Passed (P = 0.274)

Source of Variation	DF	SS	MS	F	P
type of plant	1	396.286	396.286	0.396	0.542
conc ug/l	2	21821.011	10910.505	10.893	0.002
type of plant x conc ug/l	2	15.415	7.708	0.00770	0.992
Residual	11	11017.494	1001.590		
Total	16	33990.339	2124.396		

The difference in the mean values among the different levels of type of plant is not great enough to exclude the possibility that the difference is just due to random sampling variability after allowing for the effects of differences in conc ug/l. There is not a statistically significant difference (P = 0.542).

The difference in the mean values among the different levels of conc ug/l is greater than would be expected by chance after allowing for effects of differences in type of plant. There is a statistically significant difference (P = 0.002). To isolate which group(s) differ from the others use a multiple comparison procedure.

The effect of different levels of type of plant does not depend on what level of conc ug/l is present. There is not a statistically significant interaction between type of plant and conc ug/l. (P = 0.992)

Power of performed test with alpha = 0.0500: for type of plant : 0.0500
Power of performed test with alpha = 0.0500: for conc ug/l : 0.948
Power of performed test with alpha = 0.0500: for type of plant x conc ug/l : 0.0500

Least square means for type of plant :

Group	Mean	SEM
NYM	120.303	10.980
MUT	110.167	11.794

Least square means for conc ug/l :

Group	Mean	SEM
con1	67.787	11.189
con2	144.667	14.445
con3	133.250	15.824

Least square means for type of plant x conc ug/l :

Group	Mean	SEM
NYM x con1	71.575	15.824
NYM x con2	150.333	18.272

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

NYM x con3	139.000	22.378
MUT x con1	64.000	15.824
MUT x con2	139.000	22.378
MUT x con3	127.500	22.378

All Pairwise Multiple Comparison Procedures (Tukey Test):

Comparisons for factor: **conc ug/l**

Comparison	Diff of Means	p	q	P	P<0.050
con2 vs. con1	76.879	3	5.950	0.004	Yes
con2 vs. con3	11.417	3	0.754	0.857	No
con3 vs. con1	65.463	3	4.777	0.016	Yes

Two Way Analysis of Variance Friday, December 02, 2016, 10:32:44 PM

Data source: Data 1 in statisticaloanalysischapter5thesisB

General Linear Model (No Interactions)

Dependent Variable: DT50

Normality Test: Passed (P = 0.197)

Equal Variance Test: Failed (P < 0.050)

Source of Variation	DF	SS	MS	F	P
type of plant	1	177.556	177.556	0.499	0.495
Batch	4	29223.182	7305.796	20.528	<0.001
Residual	11	3914.822	355.893		
Total	16	33990.339	2124.396		

The difference in the mean values among the different levels of type of plant is not great enough to exclude the possibility that the difference is just due to random sampling variability after allowing for the effects of differences in Batch. There is not a statistically significant difference (P = 0.495).

The difference in the mean values among the different levels of Batch is greater than would be expected by chance after allowing for effects of differences in type of plant. There is a statistically significant difference (P = <0.001). To isolate which group(s) differ from the others use a multiple comparison procedure.

Power of performed test with alpha = 0.0500: for type of plant : 0.0500
Power of performed test with alpha = 0.0500: for Batch : 1.000

Least square means for type of plant :

Group	Mean	SEM
NYM	118.830	6.535
MUT	112.167	7.778

Least square means for Batch :

Group	Mean	SEM
A	39.500	9.433
B	96.075	9.433
C	139.000	9.433
D	169.669	19.446

E 133.250 9.433

All Pairwise Multiple Comparison Procedures (Tukey Test):

Comparisons for factor: **Batch**

Comparison	Diff of Means	p	q	P	P<0.050
D vs. A	130.169	5	8.518	<0.001	Yes
D vs. B	73.594	5	4.816	0.038	Yes
D vs. E	36.419	5	2.383	0.480	No
D vs. C	30.669	5	2.007	0.629	Do Not Test
C vs. A	99.500	5	10.549	<0.001	Yes
C vs. B	42.925	5	4.551	0.051	No
C vs. E	5.750	5	0.610	0.992	Do Not Test
E vs. A	93.750	5	9.939	<0.001	Yes
E vs. B	37.175	5	3.941	0.103	Do Not Test
B vs. A	56.575	5	5.998	0.010	Yes

A result of "Do Not Test" occurs for a comparison when no significant difference is found between two means that enclose that comparison. For example, if you had four means sorted in order, and found no difference between means 4 vs. 2, then you would not test 4 vs. 3 and 3 vs. 2, but still test 4 vs. 1 and 3 vs. 1 (4 vs. 3 and 3 vs. 2 are enclosed by 4 vs. 2: 4 3 2 1). Note that not testing the enclosed means is a procedural rule, and a result of Do Not Test should be treated as if there is no significant difference between the means, even though one may appear to exist.

Because assumptions for ANOVA are not met, an ANOVA on ranks was performed

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 02, 2016, 11:04:57 PM

Data source: Data 1 in statisticaloanalysischapter5thesisB

Dependent Variable: DT50nymphaea amazonum

Group	N	Missing	Median	25%	75%
A	2	0	39.000	24.700	53.300
B	2	0	104.150	69.300	139.000
C	2	0	139.000	139.000	139.000
D	1	0	173.000	173.000	173.000
E	2	0	139.000	139.000	139.000

H = 7.200 with 4 degrees of freedom. (P = 0.126)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.126)

Kruskal-Wallis One Way Analysis of Variance on Ranks Friday, December 02, 2016, 11:06:41 PM

Data source: Data 1 in statisticaloanalysischapter5thesis (Data without outliers)

Dependent Variable: DT50eleocharis mutata

Equal Variance Test: Failed (P < 0.050)

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

Group	N	Missing	Median	25%	75%
A	2	0	40.000	26.700	53.300
B	2	0	88.000	77.000	99.000
C	2	0	139.000	139.000	139.000
E	2	0	127.500	116.000	139.000

H = 6.475 with 3 degrees of freedom. P(est.)= 0.091 P(exact)= 0.038

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = 0.038)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

All Pairwise Multiple Comparison Procedures (Tukey Test):

Comparison	Diff of Ranks	q	P<0.05
C vs A	11.000	3.175	No
C vs B	7.000	2.021	Do Not Test
C vs E	2.000	0.577	Do Not Test
E vs A	9.000	2.598	Do Not Test
E vs B	5.000	1.443	Do Not Test
B vs A	4.000	1.155	Do Not Test

Note: The multiple comparisons on ranks do not include an adjustment for ties.

A result of "Do Not Test" occurs for a comparison when no significant difference is found between the two rank sums that enclose that comparison. For example, if you had four rank sums sorted in order, and found no significant difference between rank sums 4 vs. 2, then you would not test 4 vs. 3 and 3 vs. 2, but still test 4 vs. 1 and 3 vs. 1 (4 vs. 3 and 3 vs. 2 are enclosed by 4 vs. 2: 4 3 2 1). Note that not testing the enclosed rank sums is a procedural rule, and a result of Do Not Test should be treated as if there is no significant difference between the rank sums, even though one may appear to exist.

t-test to compare DT_{50} values of different mesocosms Friday, December 02, 2016, 10:42:46 PM

Data source: Data 1 in statisticaloanalysischapter5thesisB

Normality Test: Failed (P < 0.050)

Test execution ended by user request, Rank Sum Test begun

Mann-Whitney Rank Sum Test Friday, December 02, 2016, 10:42:46 PM

Data source: Data 1 in statisticaloanalysischapter5thesis (data without outliers)

Group	N	Missing	Median	25%	75%
DT50nymphaea amazonum	9	0	139.000	65.300	139.000
DT50eleocharis mutata	8	0	107.500	65.150	139.000

Mann-Whitney U Statistic= 28.000

T = 64.000 n(small)= 8 n(big)= 9 (P = 0.446)
The difference in the median values between the two groups is not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.446)

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 5

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

Two Way Analysis of Variance Tuesday, January 28, 2014, 7:14:36 AM

Data source: Data 1 in Notebook1Chlorothalonil removal chapter 6

Balanced Design

Dependent Variable: Removal efficiency

Normality Test: Failed (P < 0.050)

Equal Variance Test: Passed (P = 0.908)

Source of Variation	DF	SS	MS	F	P
Vegetation	1	4063.061	4063.061	2.713	0.102
Batch	4	6840.088	1710.022	1.142	0.339
Vegetation x Batch	4	1420.467	355.117	0.237	0.917
Residual	150	224635.462	1497.570		
Total	159	236959.078	1490.309		

The difference in the mean values among the different levels of Vegetation is not great enough to exclude the possibility that the difference is just due to random sampling variability after allowing for the effects of differences in Batch. There is not a statistically significant difference (P = 0.102).

The difference in the mean values among the different levels of batch is not great enough to exclude the possibility that the difference is just due to random sampling variability after allowing for the effects of differences in Vegetation. There is not a statistically significant difference (P = 0.339).

The effect of different levels of Vegetation does not depend on what level of batch is present. There is not a statistically significant interaction between Vegetation and Batch. (P = 0.917)

Power of performed test with alpha = 0.0500: for Vegetation: 0.239
Power of performed test with alpha = 0.0500: for Batch: 0.0778
Power of performed test with alpha = 0.0500: for Vegetation x Batch: 0.0500

Least square means for Vegetation:

Group	Mean
Vegetated	66.386
Non-vegetated	56.308

Standard Error of LS Mean = 4.327

Least square means for Batch:

Group	Mean
A	61.290
B	63.656
C	52.376

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

D 57.449
E 71.965
Standard Error of LS Mean = 6.841

Least square means for Vegetation x Batch:

Group	Mean
Vegetated x A	61.927
Vegetated x B	68.232
Vegetated x C	59.226
Vegetated x D	66.917
Vegetated x E	75.630
Non-vegetated x A	60.654
Non-vegetated x B	59.081
Non-vegetated x C	45.525
Non-vegetated x D	47.980
Non-vegetated x E	68.299

Standard Error of LS Mean = 9.675

Note: Because the assumption for Normality failed, an ANOVA ON RANKS was performed.

Kruskal-Wallis One Way Analysis of Variance on Ranks Tuesday, January 28, 2014, 6:18:56 PM

Data source: Data 1 in CHLOROTHALONIL_28JAN14

Dependent Variable: Removal Efficiency

Group	N	Missing	Median	25%	75%
vegA0hour	2	0	0.000	0.000	0.000
vegA2hour	2	0	34.322	15.385	53.260
vegA4hour	2	0	65.936	55.077	76.796
vegA22hour	2	0	43.484	38.122	48.846
vegA48hour	2	0	60.261	51.462	69.061
vegA72hour	2	0	91.410	86.519	96.300
vegA97hour	2	0	100.000	100.000	100.000
vegA334hour	2	0	100.000	100.000	100.000
unvegA0hour	2	0	0.000	0.000	0.000
unvegA2hour	2	0	28.400	16.143	40.656
unvegA4hour	2	0	51.203	27.787	74.619
unvegA22hour	2	0	38.834	37.668	40.000
unvegA48hour	2	0	81.584	70.164	93.004
unvegA72hour	2	0	87.015	86.721	87.309
unvegA97hour	2	0	98.197	96.393	100.000
unveg334hour	2	0	100.000	100.000	100.000
vegB0hour	2	0	0.000	0.000	0.000
vegB2hour	2	0	6.020	0.000	12.041
vegB4hour	2	0	60.071	45.815	74.327

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

vegB22hour	2	0	82.516	75.429	89.604
vegB48hour	2	0	97.245	94.490	100.000
vegB72hour	2	0	100.000	100.000	100.000
vegB97hour	2	0	100.000	100.000	100.000
vegB334hour	2	0	100.000	100.000	100.000
unvegB0hour	2	0	0.000	0.000	0.000
unvegB2hour	2	0	19.411	0.000	38.821
unvegB4hour	2	0	24.995	22.358	27.632
unvegB22hour	2	0	52.791	40.947	64.634
unvegB48hour	2	0	79.902	69.593	90.211
unvegB72hour	2	0	97.785	95.569	100.000
unvegB97hour	2	0	97.764	95.528	100.000
unvegB334hour	2	0	100.000	100.000	100.000
vegC0hour	2	0	0.000	0.000	0.000
vegC2hour	2	0	43.371	42.246	44.496
vegC4hour	2	0	24.557	1.159	47.954
vegC22hour	2	0	41.079	30.202	51.957
vegC48hour	2	0	74.207	71.239	77.174
vegC72hour	2	0	93.116	88.768	97.464
vegC97hour	2	0	97.482	96.232	98.732
vegC334hour	2	0	100.000	100.000	100.000
unvegC0hour	2	0	0.000	0.000	0.000
unvegC2hour	2	0	29.519	20.281	38.758
unvegC4hour	2	0	2.934	0.000	5.867
unvegC22hour	2	0	9.255	0.000	18.509
unvegC48hour	2	0	36.595	15.179	58.012
unvegC72hour	2	0	91.415	89.413	93.416
unvegC97hour	2	0	94.483	92.919	96.046
unvegC334hour	2	0	100.000	100.000	100.000
vegD0hour	2	0	0.000	0.000	0.000
vegD2hour	2	0	6.540	0.000	13.080
vegD4hour	2	0	78.882	70.253	87.511
vegD22hour	2	0	77.657	73.710	81.603
vegD48hour	2	0	83.579	80.407	86.751
vegD72hour	2	0	93.111	90.928	95.294
vegD97hour	2	0	96.414	94.389	98.439
vegD334hour	2	0	99.156	98.312	100.000
unvegD0hour	2	0	0.000	0.000	0.000
unvegD2hour	2	0	0.000	0.000	0.000
unvegD4hour	2	0	37.570	7.563	67.578
unvegD22hour	2	0	38.257	34.161	42.353
unvegD48hour	2	0	37.368	19.580	55.155
unvegD72hour	2	0	83.697	78.824	88.571
unvegD97hour	2	0	97.510	97.205	97.815
unvegD334hour	2	0	89.439	81.988	96.891
vegE0hour	2	0	0.000	0.000	0.000

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

vegE2hour	2	0	66.915	44.505	89.325
vegE4hour	2	0	51.731	29.289	74.174
vegE22hour	2	0	89.734	85.813	93.655
vegE48hour	2	0	97.233	95.331	99.135
vegE72hour	2	0	99.429	99.056	99.801
vegE97hour	2	0	100.000	100.000	100.000
vegE334hour	2	0	100.000	100.000	100.000
unvegE0hour	2	0	0.000	0.000	0.000
unvegE2hour	2	0	55.123	42.857	67.389
unvegE4hour	2	0	31.288	0.000	62.575
unvegE22hour	2	0	72.079	67.504	76.655
unvegE48hour	2	0	89.292	84.236	94.349
unvegE72hour	2	0	98.613	98.069	99.158
unvegE97hour	2	0	100.000	100.000	100.000
unvegE334hour	2	0	100.000	100.000	100.000

H = 152.364 with 79 degrees of freedom. (P = <0.001)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = <0.001)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

All Pairwise Multiple Comparison Procedures (Tukey Test): Do not test: no significant differences found.

Lambda-cyhalothrin
Two Way Analysis of Variance Tuesday, January 28, 2014, 11:01:41 AM

Data source: Data 1 in Notebook1lambda-cyhalothrin

General Linear Model

Dependent Variable: Removal Efficiency

Normality Test: Failed (P < 0.050)

Equal Variance Test: Failed (P < 0.050)

Source of Variation	DF	SS	MS	F	P
Vegetation	1	187130.201	187130.201	1.572	0.212
Batch	4	7833018.636	1958254.659	16.454	<0.001
Vegetation x Batch	4	822122.065	205530.516	1.727	0.147
Residual	138	16423641.205	119011.893		
Total	147	25303056.588	172129.637		

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

Note: Both assumptions failed for performing a TWO-WAY ANOVA and an ANOVA on Ranks was executed.

Kruskal-Wallis One Way Analysis of Variance on Ranks Tuesday, January 28, 2014, 8:53:35 PM

Data source: Data 1 in LAMBDACYHALOTHRIN_28JAN

Dependent Variable: removal efficiency

Group	N	Missing	Median	25%	75%
vegA0hour	2	0	0.000	0.000	0.000
vegA2hour	2	0	28.363	7.059	49.667
vegA4hour	2	0	34.333	0.000	68.667
vegA22hour	2	0	58.999	21.765	96.233
vegA48hour	2	0	44.300	0.000	88.600
vegA72hour	2	0	54.790	37.647	71.933
vegA97hour	2	0	73.265	63.529	83.000
unvegA0hour	2	0	0.000	0.000	0.000
unvegA2hour	2	0	25.385	0.000	50.769
unvegA4hour	2	0	68.077	63.077	73.077
unvegA22hour	2	0	56.154	42.692	69.615
unvegA48hour	2	0	75.596	68.269	82.923
unvegA72hour	2	0	49.231	35.769	62.692
unvegA97hour	2	0	69.135	61.731	76.538
vegB0hour	2	0	0.000	0.000	0.000
vegB2hour	2	0	16.721	0.000	33.443
vegB4hour	2	0	44.688	15.508	73.869
vegB22hour	2	0	43.051	20.856	65.246
vegB47hour	2	0	42.753	20.588	64.918
vegB72hour	2	0	51.763	33.690	69.836
vegB93hour	2	0	67.631	54.278	80.984
vegB334hour	2	0	79.679	59.358	100.000
unvegB0hour	2	0	0.000	0.000	0.000
unvegB2hour	2	0	0.000	0.000	0.000
unvegB4hour	2	0	0.000	0.000	0.000
unvegB22hour	2	0	0.000	0.000	0.000
unvegB47hour	2	0	70.386	60.930	79.843
unvegB72hour	2	0	82.674	80.465	84.882
unvegB93hour	2	0	74.759	65.581	83.937
unvegB334hour	2	0	100.000	100.000	100.000
vegC0hour	2	0	0.000	0.000	0.000
vegC2hour	2	0	0.000	0.000	0.000
vegC4hour	2	0	0.000	0.000	0.000
vegC22hour	2	0	0.000	0.000	0.000
vegC47hour	2	0	2.796	0.000	5.593
vegC72hour	2	0	5.928	0.000	11.857

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

vegC93hour	2	0	7.494	0.000	14.989
vegC334hour	2	0	43.812	43.624	44.000
unvegC0hour	2	0	0.000	0.000	0.000
unvegC2hour	2	0	0.000	0.000	0.000
unvegC4hour	2	0	0.000	0.000	0.000
unvegC22hour	2	0	0.000	0.000	0.000
unvegC47hour	2	0	0.000	0.000	0.000
unvegC72hour	2	0	0.000	0.000	0.000
unvegC93hour	2	0	27.616	0.000	55.231
unvegC334hour	2	0	64.171	55.763	72.578
vegD0hour	2	0	0.000	0.000	0.000
vegD2hour	2	0	-428.363	-463.393	-393.333
vegD4hour	2	0	-2443.024	-4060.714	-825.333
vegD22hour	2	0	-349.851	-366.667	-333.036
vegD47hour	2	0	-768.423	-1446.667	-90.179
vegD72hour	2	0	-895.369	-1174.667	-616.071
vegD93hour	2	0	-464.554	-749.107	-180.000
vegD334hour	2	0	-265.304	-444.000	-86.607
unvegD0hour	2	0	0.000	0.000	0.000
unvegD2hour	2	0	-33.563	-41.176	-25.949
unvegD4hour	2	0	-1015.748	-1096.203	-935.294
unvegD22hour	2	0	-402.007	-465.190	-338.824
unvegD47hour	2	0	-442.945	-741.772	-144.118
unvegD72hour	2	0	-494.985	-532.911	-457.059
unvegD93hour	2	0	-129.724	-208.861	-50.588
unveg334hour	2	0	-204.579	-229.412	-179.747
vegE0hour	2	0	0.000	0.000	0.000
vegE2hour	2	0	64.319	47.200	81.438
vegE4hour	2	0	52.180	32	72.360
vegE22hour	2	0	90.418	89.600	91.236
vegE47hour	2	0	74.985	62.667	87.303
vegE72hour	2	0	80.677	77.820	83.533
unvegE0hour	2	0	0.000	0.000	0.000
unvegE2hour	2	0	28.967	1.176	56.757
unvegE4hour	2	0	31.787	13.235	50.338
unvegE22hour	2	0	87.779	83.294	92.264
unvegE47hour	2	0	65.605	58.000	73.209
unvegE72hour	2	0	74.946	66.412	83.48

H = 135.393 with 73 degrees of freedom. (P = <0.001)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = <0.001)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

Note: results for Tukey test are omitted, but no significant differences were found.

Imidacloprid

Two Way Analysis of Variance Tuesday, February 04, 2014, 12:59:07 PM

Data source: Data 1 in IMIDACLOPRID_28JAN13

Balanced Design

Dependent Variable: Removal Efficiency

Normality Test: Failed (P < 0.050)

Equal Variance Test: Failed (P < 0.050)

Source of Variation	DF	SS	MS	F	P
Vegetation	1	170.064	170.064	0.466	0.496
Time	7	167419.653	23917.093	65.590	<0.001
Vegetation x Time	7	176.790	25.256	0.0693	0.999
Residual	144	52509.153	364.647		
Total	159	220275.660	1385.382		

Note: the assumptions for execution of a two-way ANOVA were not met and an ANOVA ON RANKS was performed.

Kruskal-Wallis One Way Analysis of Variance on Ranks Tuesday, February 04, 2014, 12:59:57 PM

Data source: Data 1 in IMIDACLOPRID_28JAN13

Dependent Variable: Removal Efficiency

Group	N	Missing	Median	25%	75%
vegA0hour	3	0	0.000	0.000	0.000
vegA2hour	2	0	1.288	0.000	2.575
vegA4hour	2	0	14.592	0.000	29.185
vegA6hour	2	0	13.090	0.000	26.180
vegA23hour	2	0	54.399	50.000	58.798
vegA76hour	2	0	51.077	44.643	57.511
vegA146hour	2	0	76.618	70.833	82.403
Group	**N**	**Missing**	**Median**	**25%**	**75%**
vegA240hour	2	0	94.020	93.991	94.048
unvegA0hour	2	0	0.000	0.000	0.000
unvegA2hour	2	0	3.165	0.000	6.329
unvegA4hour	2	0	21.454	13.793	29.114
unvegA6hour	2	0	30.780	23.629	37.931
unvegA23hour	2	0	71.560	48.101	95.019

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

unvegA76hour	2	0	66.718	55.274	78.161
unvegA146hour	2	0	71.536	70.498	72.574
unvegA240hour	2	0	92.199	91.571	92.827
vegB0hour	2	0	0.000	0.000	0.000
vegB2hour	2	0	3.630	0.000	7.261
vegB4hour	2	0	9.406	0.000	18.812
vegB6hour	2	0	66.773	60.726	72.821
vegB23hour	2	0	64.930	56.923	72.937
vegB76hour	2	0	85.671	82.564	88.779
vegB146hour	2	0	95.291	94.872	95.710
vegB240hour	2	0	97.124	95.897	98.350
unvegB0hour	2	0	0.000	0.000	0.000
unvegB2hour	2	0	14.821	0.000	29.643
unvegB4hour	2	0	25.171	23.913	26.429
unvegB6hour	2	0	55.303	53.106	57.500
unvegB23hour	2	0	63.579	61.801	65.357
unvegB76hour	2	0	84.783	83.851	85.714
unvegB146hour	2	0	91.988	91.429	92.547
unvegB240hour	2	0	97.057	95.357	98.758
vegC0hour	1	0	0.000	0.000	0.000
vegC2hour	2	0	13.345	0.000	26.690
vegC4hour	2	0	14.173	0.000	28.346
vegC6hour	2	0	14.173	0.000	28.346
vegC23hour	2	0	78.270	73.622	82.918
vegC76hour	2	0	95.018	90.036	100.000
vegC146hour	2	0	86.155	79.004	93.307
vegC240hour	2	0	86.678	83.986	89.370
unvegC0hour	2	0	0.000	0.000	0.000
unvegC2hour	2	0	0.000	0.000	0.000
unvegC4hour	2	0	0.000	0.000	0.000
unvegC6hour	2	0	0.000	0.000	0.000
unvegC23hour	2	0	25.248	20.792	29.703
unvegC76hour	2	0	55.941	41.089	70.792
unvegC146hour	2	0	79.703	74.257	85.149
unvegC240hour	2	0	75.495	75.248	75.743
vegD0hour	2	0	0.000	0.000	0.000
vegD2hour	2	0	8.915	0.000	17.829
vegD4hour	2	0	15.078	10.000	20.155
vegD6hour	2	0	15.078	10.000	20.155
vegD23hour	2	0	27.031	19.565	34.496
vegD76hour	2	0	83.217	80.000	86.434
vegD146hour	2	0	23.883	18.696	29.070
vegD240hour	2	0	96.739	93.478	100.000
unvegD0hour	2	0	0.000	0.000	0.000
unvegD2hour	2	0	0.000	0.000	0.000
unvegD4hour	2	0	0.000	0.000	0.000

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

unvegD6hour	2	0	0.000	0.000	0.000
unvegD23hour	2	0	37.572	34.906	40.239
unvegD76hour	2	0	84.265	82.075	86.454
unvegD146hour	2	0	36.325	35.857	36.792
unvegD240hour	2	0	97.809	95.618	100.000
vegE0hour	2	0	0.000	0.000	0.000
vegE2hour	2	0	0.000	0.000	0.000
vegE4hour	2	0	0.000	0.000	0.000
vegE6hour	2	0	0.000	0.000	0.000
vegE23hour	2	0	10.505	5.625	15.385
vegE76hour	2	0	38.612	26.875	50.350
vegE146hour	2	0	27.445	20.625	34.266
vegE240hour	2	0	100.000	100.000	100.000
unvegE0hour	2	0	0.000	0.000	0.000
unvegE2hour	2	0	2.232	0.000	4.464
unvegE4hour	2	0	0.000	0.000	0.000
unvegE6hour	2	0	0.000	0.000	0.000
unvegE23hour	2	0	17.068	11.607	22.530
unvegE76hour	2	0	39.706	38.340	41.071
unvegE146hour	2	0	38.271	36.364	40.179
unvegE240hour	2	0	100.000	100.000	100.000

H = 151.465 with 79 degrees of freedom. (P = <0.001)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = <0.001)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

All Pairwise Multiple Comparison Procedures (Dunn's Method) showed no significant differences.

Appendix 4: Statistical analysis DT_{50} values Chapter 6

Dataset used to compare significant differences by means of a t-test between $DT_{50\ values}$ of vegetated and non-vegetated systems

	CHLOVEG	**LAMVEG**	**IMIVEG**	**CHLO nonveg**	**LAM nonveg**	**IMI nonveg**
Batch1	20.7	61.1	63	21.6	68	115.5
Batch2	9.3	82.5	41.9	14.4	31.9	44.2
Batch3	18.7	189.9	55.7	17.3	149.1	55.7
Batch4	20.4	outlier	55.2	20.4	outlier	55.2
Batch5	10.2	43.3	70.7	12.8	31.5	86.6

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

T-test

Data source: Data 1 in DT50lamvegnonveghoofdstuk6zonderoutlier

Normality Test: Passed (P = 0.094)

Equal Variance Test: Passed (P = 0.449)

Group Name	N	Missing	Mean	Std Dev	SEM
DT50chloVEG	5	0	15.860	5.639	2.522
DT50chlononveg	5	0	17.300	3.767	1.685

Difference -1.440

t = -0.475 with 8 degrees of freedom. (P = 0.648)

95 percent confidence interval for difference of means: -8.433 to 5.553

The difference in the mean values of the two groups is not great enough to reject the possibility that the difference is due to random sampling variability. There is not a statistically significant difference between the input groups (P = 0.648).

Power of performed test with alpha = 0.050: 0.050

The power of the performed test (0.050) is below the desired power of 0.800.
Less than desired power indicates you are less likely to detect a difference when one actually exists. Negative results should be interpreted cautiously.

T-test Sunday, August 21, 2016, 11:46:26 AM

Data source: Data 1 in DT50lamvegnonveghoofdstuk6zonderoutlier

Normality Test: Failed (P < 0.050)

Test execution ended by user request, Rank Sum Test begun

Mann-Whitney Rank Sum Test Sunday, August 21, 2016, 11:46:26 AM

Data source: Data 1 in DT50lamvegnonveghoofdstuk6zonderoutlier

Group	N	Missing	Median	25%	75%
DT50lamVEG	5	1	71.800	52.200	136.200
DT50lamnonveg	5	1	49.950	31.700	108.550

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6
Mann-Whitney U Statistic= 5.000

T = 21.000 n(small)= 4 n(big)= 4 P(est.)= 0.470 P(exact)= 0.486

The difference in the median values between the two groups is not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.486)

T-test Sunday, August 21, 2016, 11:49:04 AM

Data source: Data 1 in DT50lamvegnonveghoofdstuk6zonderoutlier

Normality Test: Passed (P = 0.453)

Equal Variance Test: Passed (P = 0.264)

Group Name	N	Missing	Mean	Std Dev	SEM
DT_{50} imiVEG	5	0	57.300	10.679	4.776
DT_{50}iminonveg	5	0	71.440	29.260	13.086

Difference -14.140

t = -1.015 with 8 degrees of freedom. (P = 0.340)

95 percent confidence interval for difference of means: -46.262 to 17.982

The difference in the mean values of the two groups is not great enough to reject the possibility that the difference is due to random sampling variability. There is not a statistically significant difference between the input groups (P = 0.340).

Power of performed test with alpha = 0.050: 0.051

The power of the performed test (0.051) is below the desired power of 0.800.
Less than desired power indicates you are less likely to detect a difference when one actually exists. Negative results should be interpreted cautiously.

Note: For a two way ANOVA data was not normally distributed and an ANOVA on ranks was performed. To determine if significant differences exists between pesticides in veg and nonveg systems.

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

Dataset used to compare significant differences by means of a Kruskal-Wallis One Way Analysis of Variance on Ranks between DT $_{50}$ values of pesticides in vegetated and non-vegetated systems

Pesticide	**DT_{50}**
CHLOVEG	20.7
CHLOVEG	9.3
CHLOVEG	18.7
CHLOVEG	20.4
CHLOVEG	10.2
LAMVEG	61.1
LAMVEG	82.5
LAMVEG	189.9
LAMVEG	43.3
IMIVEG	63
IMIVEG	41.9
IMIVEG	55.7
IMIVEG	55.2
IMIVEG	70.7
CHLONONVEG	21.6
CHLONONVEG	14.4
CHLONONVEG	17.3
CHLONONVEG	20.4
CHLONONVEG	12.8
LAMNONVEG	68
LAMNONVEG	31.9
LAMNONVEG	149.1
LAMNONVEG	31.5
IMINONVEG	115.5
IMINONVEG	44.2
IMINONVEG	55.7
IMINONVEG	55.2
IMINONVEG	86.6

Two Way Analysis of Variance Sunday, August 21, 2016, 11:31:23 AM

Data source: Data 1 in DT50lamvegnonveghoofdstuk6zonderoutlier

General Linear Model (No Interactions)

Dependent Variable: DT50chloVEG

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 6

Normality Test: Failed (P < 0.050)

Equal Variance Test: Passed (P = 1.000)

Kruskal-Wallis One Way Analysis of Variance on Ranks Sunday, August 21, 2016, 11:41:09 AM

Data source: Data 1 in DT50lamvegnonveghoofdstuk6zonderoutlier

Dependent Variable: DT50chloVEG

Group	N	Missing	Median	25%	75%
CHLOVEG	5	0	18.700	9.975	20.475
LAMVEG	4	0	71.800	52.200	136.200
IMIVEG	5	0	55.700	51.875	64.925
CHLONONVEG	5	0	17.300	14.000	20.700
LAMNONVEG	4	0	49.950	31.700	108.550
IMINONVEG	5	0	55.700	52.450	93.825

H = 19.155 with 5 degrees of freedom. (P = 0.002)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = 0.002)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

All Pairwise Multiple Comparison Procedures (Dunn's Method):

Comparison	Diff of Ranks	Q	P<0.05
LAMVEG vs CHLOVEG	16.400	2.972	Yes
LAMVEG vs CHLONONVEG	15.600	2.827	No
LAMVEG vs LAMNONVEG	3.500	0.602	Do Not Test
LAMVEG vs IMIVEG	3.100	0.562	Do Not Test
LAMVEG vs IMINONVEG	1.300	0.236	Do Not Test
IMINONVEG vs CHLOVEG	15.100	2.902	No
IMINONVEG vs CHLONONVEG	14.300	2.749	Do Not Test
IMINONVEG vs LAMNONVEG	2.200	0.399	Do Not Test
Comparison	**Diff of Ranks**	**Q**	**P<0.05**
IMINONVEG vs IMIVEG	1.800	0.346	Do Not Test
IMIVEG vs CHLOVEG	13.300	2.556	Do Not Test
IMIVEG vs CHLONONVEG	12.500	2.403	Do Not Test
IMIVEG vs LAMNONVEG	0.400	0.0725	Do Not Test
LAMNONVEG vs CHLOVEG	12.900	2.338	Do Not Test
LAMNONVEG vs CHLONONVEG	12.100	2.193	Do Not Test
CHLONONVEG vs CHLOVEG	0.800	0.154	Do Not Test

Note: The multiple comparisons on ranks do not include an adjustment for ties.

APPENDIX 4

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 7

APPENDIX 4: STATISTICAL OUTPUTS OF DIFFERENT TESTS PERFORMED (CHAPTER 7)

NOTE: FOR LAMBDA-CYHALOTHRIN'S COMPARISON IN PLANTS/ROOTS, DATA WAS NOT NORMALLY DISTRIBUTED AND AN ANOVA ON RANKS WAS PERFORMED.

Kruskal-Wallis One Way Analysis of Variance on Ranks Saturday, September 03, 2016, 2:15:50 AM

Data source: Data 1 in Notebook2

Dependent Variable: lamda Nym+Polyt/gDW ph1ph2

Group	N	Missing	Median	25%	75%
plantnympha1	18	0	0.000	0.000	0.000
rootsnympha1	18	0	0.000	0.000	0.1000
plantnympha2	18	0	0.000	0.000	0.370
rootsnympha2	18	0	0.000	0.000	0.340
plantpolysta	18	0	3.110	0.0600	9.170
rootspolysta	18	0	0.165	0.01000	0.830

H = 26.300 with 5 degrees of freedom. (P = <0.001)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference (P = <0.001)

To isolate the group or groups that differ from the others use a multiple comparison procedure.

All Pairwise Multiple Comparison Procedures (Tukey Test): Values in red show a significant difference

Comparison	Diff of Ranks	q	P<0.05
plantpolysta vs rootsnympha1	737.500	5.550	Yes
plantpolysta vs plantnympha1	732.500	5.512	Yes
plantpolysta vs rootsnympha2	577.000	4.342	Yes
plantpolysta vs plantnympha2	555.500	4.180	Yes
plantpolysta vs rootspolysta	313.500	2.359	No
rootspolysta vs rootsnympha1	424.000	3.191	No
rootspolysta vs plantnympha1	419.000	3.153	Do Not Test
rootspolysta vs rootsnympha2	263.500	1.983	Do Not Test
rootspolysta vs plantnympha2	242	1.821	Do Not Test
plantnympha2 vs rootsnympha1	182	1.370	Do Not Test
plantnympha2 vs plantnympha1	177.000	1.332	Do Not Test
plantnympha2 vs rootsnympha2	21.500	0.162	Do Not Test
rootsnympha2 vs rootsnympha1	160.500	1.208	Do Not Test
rootsnympha2 vs plantnympha1	155.500	1.170	Do Not Test
plantnympha1 vs rootsnympha1	5.000	0.0376	Do Not Test

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 7

Note: The multiple comparisons on ranks do not include an adjustment for ties.

A result of "Do Not Test" occurs for a comparison when no significant difference is found between the two rank sums that enclose that comparison. For example, if you had four rank sums sorted in order, and found no significant difference between rank sums 4 vs. 2, then you would not test 4 vs. 3 and 3 vs. 2, but still test 4 vs. 1 and 3 vs. 1 (4 vs. 3 and 3 vs. 2 are enclosed by 4 vs. 2: 4 3 2 1). Note that not testing the enclosed rank sums is a procedural rule, and a result of Do Not Test should be treated as if there is no significant difference between the rank sums, even though one may appear to exist.

Two Way Analysis of Variance woensdag, November 05, 2014, 16:57:44

Data source: Data 1 in Stat difference for removal imidacloprid phase 1 and phase 2
General Linear Model (No Interactions)

Dependent Variable: Concentrations

Normality Test: Passed (P = 0,295)

Equal Variance Test: Passed (P = 1,000)

Source of Variation	DF	SS	MS	F	P
Time	9	5,450	0,606	1,232	0,380
Phase	1	4,518	4,518	9,193	0,014
Residual	9	4,423	0,491		
Total	19	14,391	0,757		

The difference in the mean values among the different levels of Time is not great enough to exclude the possibility that the difference is just due to random sampling variability after allowing for the effects of differences in phase. There is not a statistically significant difference (P = 0,380).

The difference in the mean values among the different levels of phase is greater than would be expected by chance after allowing for effects of differences in Time. There is a statistically significant difference (P = 0,014). To isolate which group(s) differ from the others use a multiple comparison procedure.

Power of performed test with alpha = 0,0500: for Time: 0,0863
Power of performed test with alpha = 0,0500: for Phase: 0,728

Least square means for Time:

Group	Mean
0,000	1,180E-016
2,000	1,316
24,000	1,400
48,000	0,852
72,000	0,566

96,000	0,963
144,000	0,111

Least square means for Time:

Group	**Mean**
192,000	0,105
240,000	0,00350
312,000	0,147

Standard Error of LS Mean = 0,496

Least square means for phase:

Group	**Mean**
Phase 1: 0,0710	
Phase 2: 1,022	

Standard Error of LS Mean = 0,222

All Pairwise Multiple Comparison Procedures (Holm-Sidak method):

Overall significance level = 0,05

Comparison	**Diff of Means**	**t**	**Unadjusted P**	**Critical Level**	**Significant?**
48 vs. 2	273.273	5.012	<0.001	0.003	Yes
96 vs. 2	257.743	4.727	<0.001	0.004	Yes
312 vs. 2	238.283	4.370	<0.001	0.004	Yes
144 vs. 2	214.986	3.943	<0.001	0.004	Yes

Comparisons for factor: **Time**

Comparison	**Diff of Means**	**t**	**Unadjusted P**	**Critical Level**	**Significant?**
24,000 vs. 0,000	1,400	1,997	0,077	0,001	No
24,000 vs. 240,000	1,397	1,992	0,078	0,001	No
2,000 vs. 0,000	1,316	1,877	0,093	0,001	No
2,000 vs. 240,000	1,312	1,872	0,094	0,001	No
24,000 vs. 192,000	1,295	1,848	0,098	0,001	No
24,000 vs. 144,000	1,289	1,839	0,099	0,001	No
24,000 vs. 312,000	1,253	1,787	0,108	0,001	No
2,000 vs. 192,000	1,211	1,727	0,118	0,001	No
2,000 vs. 144,000	1,205	1,719	0,120	0,001	No
2,000 vs. 312,000	1,169	1,667	0,130	0,001	No
96,000 vs. 0,000	0,963	1,374	0,203	0,001	No
96,000 vs. 240,000	0,960	1,369	0,204	0,002	No
96,000 vs. 192,000	0,858	1,224	0,252	0,002	No
96,000 vs. 144,000	0,852	1,215	0,255	0,002	No
48,000 vs. 0,000	0,852	1,215	0,255	0,002	No
48,000 vs. 240,000	0,848	1,210	0,257	0,002	No

24,000 vs. 72,000	0,834	1,190	0,264	0,002	No
96,000 vs. 312,000	0,816	1,164	0,274	0,002	No
2,000 vs. 72,000	0,750	1,070	0,313	0,002	No
48,000 vs. 192,000	0,747	1,066	0,314	0,002	No
48,000 vs. 144,000	0,741	1,057	0,318	0,002	No
48,000 vs. 312,000	0,705	1,005	0,341	0,002	No
72,000 vs. 0,000	0,566	0,807	0,440	0,002	No
72,000 vs. 240,000	0,562	0,802	0,443	0,002	No
24,000 vs. 48,000	0,548	0,782	0,454	0,002	No
2,000 vs. 48,000	0,464	0,662	0,525	0,003	No
72,000 vs. 192,000	0,461	0,658	0,527	0,003	No
72,000 vs. 144,000	0,455	0,649	0,533	0,003	No
24,000 vs. 96,000	0,437	0,623	0,549	0,003	No
72,000 vs. 312,000	0,419	0,597	0,565	0,003	No
96,000 vs. 72,000	0,397	0,567	0,585	0,003	No
2,000 vs. 96,000	0,353	0,503	0,627	0,004	No
48,000 vs. 72,000	0,286	0,408	0,693	0,004	No
312,000 vs. 0,000	0,147	0,210	0,838	0,004	No
312,000 vs. 240,000	0,144	0,205	0,842	0,005	No
96,000 vs. 48,000	0,111	0,159	0,877	0,005	No
144,000 vs. 0,000	0,111	0,158	0,878	0,006	No
144,000 vs. 240,000	0,108	0,153	0,882	0,006	No
192,000 vs. 0,000	0,105	0,149	0,885	0,007	No
192,000 vs. 240,000	0,101	0,144	0,888	0,009	No
24,000 vs. 2,000	0,0842	0,120	0,907	0,010	No
312,000 vs. 192,000	0,0425	0,0606	0,953	0,013	No
312,000 vs. 144,000	0,0362	0,0517	0,960	0,017	No
144,000 vs. 192,000	0,00625	0,00892	0,993	0,025	No
240,000 vs. 0,000	0,00350	0,00499	0,996	0,050	No

Comparisons for factor: ph**ase**

Comparison	**Diff of Means**	**t**	**Unadjusted P**	**Critical Level**	**Significant?**
Phase 2 vs. phase 1	0,951	3,032	0,014	0,050	Yes

ANALYSIS FOR PHASE 1, BECAUSE ASSUMPTIONS WERE NOT MET FOR NORMALITY, A RANK SUM WAS PERFORMED

Tuesday, September 01, 2015, 4:28:42 PM

Data source: Data 1 in statisticalanalysisimiinplantswetlandexperiment

Normality Test: Failed (P < 0.050)

Test execution ended by user request, Rank Sum Test begun

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 7

Mann-Whitney Rank Sum Test Tuesday, September 01, 2015, 4:28:42 PM

Data source: Data 1 in statisticalanalysisimiinplantswetlandexperiment

Group	N	Missing	Median	25%	75%
Plantsug/g n.amaph1	18	0	177.625	76.330	400.540
Rootsug/g n.amaph1	18	0	182.535	68.080	468.730

Plantsug/g n.amaph1: plant part of *N. amazonum* phase 1
Mann-Whitney U Statistic= 159.000

T = 336.000 n (small) = 18 n (big) = 18 (P = 0.937)

The difference in the median values between the two groups is not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference (P = 0.937)

Three Way Analysis of Variance Tuesday, September 01, 2015, 6:54:44 PM

Data source: Data 1 in statisticalanalysisimidaclopridnplantswetlandexp.

Balanced Design (No Interactions)

Dependent Variable: ug/g imi phase2 diff. plants

Normality Test: Passed (P = 0.109)

Equal Variance Test: Passed (P = 1.000)

Source of Variation	DF	SS	MS	F	P
Time (hours)	5	714306.652	142861.330	8.010	<0.001
Sampling point	2	181156.023	90578.011	5.078	0.013
Plant part	3	4411773.529	1470591.176	82.451	<0.001
Residual	30	535081.165	17836.039		
Total	71	8176868.777	115167.166		

The difference in the mean values among the different levels of time (hours) are greater than would be expected by chance after allowing for the effects of differences in sampling point and plant part. There is a statistically significant difference (P = <0.001). To isolate which group(s) differ from the others use a multiple comparison procedure.

The difference in the mean values among the different levels of sampling point are greater than would be expected by chance after allowing for the effects of differences in time (hours) and plant part. There

is a statistically significant difference (P = 0.013). To isolate which group(s) differ from the others use a multiple comparison procedure.

The difference in the mean values among the different levels of plant part are greater than would be expected by chance after allowing for the effects of differences in time (hours) and sampling point. There is a statistically significant difference (P = <0.001). To isolate which group(s) differ from the others use a multiple comparison procedure.

All Pairwise Multiple Comparison Procedures (Holm-Sidak method):
Overall significance level = 0.05

Comparisons for factor: **time (hours)**

Comparison	Diff of Means	t	Unadjusted P	Critical Level	Significant?
48 vs. 2	273.273	5.012	<0.001	0.003	Yes
96 vs. 2	257.743	4.727	<0.001	0.004	Yes
312 vs. 2	238.283	4.370	<0.001	0.004	Yes
144 vs. 2	214.986	3.943	<0.001	0.004	Yes
48 vs. 192	181.382	3.327	0.002	0.005	Yes
96 vs. 192	165.852	3.042	0.005	0.005	Yes
312 vs. 192	146.393	2.685	0.012	0.006	No
144 vs. 192	123.095	2.258	0.031	0.006	No
192 vs. 2	91.891	1.685	0.102	0.007	No
48 vs. 144	58.287	1.069	0.294	0.009	No
96 vs. 144	42.757	0.784	0.439	0.010	No
48 vs. 312	34.990	0.642	0.526	0.013	No
312 vs. 144	23.297	0.427	0.672	0.017	No
96 vs. 312	19.459	0.357	0.724	0.025	No
48 vs. 96	15.531	0.285	0.778	0.050	No

Comparisons for factor: sampling point

Comparison	Diff of Means	t	Unadjusted P	Critical Level	Significant?
V1 vs. V2	121.680	3.156	0.004	0.017	Yes
V1 vs. V3	75.596	1.961	0.059	0.025	No
V3 vs. V2	46.084	1.195	0.241	0.050	No

Comparisons for factor: plant part indicates shoots and leaves
Note: Plantpoly: plant part of the *E.polystachya* plant
Rootsnympha: root part of the *N. amazonum* plant

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 7

Comparison	Diff of Means	t	Unadjusted P	Critical Level
plantpolysta vs. rootsnympha	657.471	14.769	<0.001	0.009
plantpolysta vs. rootspolysta	535.889	12.038	<0.001	0.010
plantpolysta vs. plantnympha	375.989	8.446	<0.001	0.013
plantnympha vs. rootsnympha	281.482	6.323	<0.001	0.017
plantnympha vs. rootspolysta	159.900	3.592	0.001	0.025
rootspolysta vs. rootsnympha	121.582	2.731	0.010	0.050

Comparison	Significant?
plantpolysta vs. rootsnympha	Yes
plantpolysta vs. rootspolysta	Yes
plantpolysta vs. plantnympha	Yes
plantnympha vs. rootsnympha	Yes
plantnympha vs. rootspolysta	Yes
rootspolysta vs. rootsnympha	Yes

Power of performed test with alpha = 0.0500: for time (hours): 0.996
Power of performed test with alpha = 0.0500: for sampling point: 0.680
Power of performed test with alpha = 0.0500: for plant part: 1.000

Least square means for time (hours):

Group	Mean
2	91.839
48	365.112
96.000	349.582
144.000	306.825
192	183.730
312	330.123

Standard Error of LS Mean = 38.553

Least square means for sampling point:

Group	Mean
V1	336.960
V2	215.280
V3	261.365

Standard Error of LS Mean = 27.261

Least square means for plant part:

Group	Mean
plantnympha	287.550
rootsnympha	6.068
plantpolysta	663.539
rootspolysta	127.650

Standard Error of LS Mean = 31.478

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 7

Raw data used for statistical tests (phase 2, chapter 7)		
Time (hours)	Planttype plant part	ug/gimiphase2diffplants
2	plantnympha	4.9000
2	plantnympha	33.7000
2	plantnympha	101.1000
48	plantnympha	700.3000
48	plantnympha	225.2000
48	plantnympha	1071.9000
96	plantnympha	584.0000
96	plantnympha	58.7000
96	plantnympha	726.7000
144	plantnympha	419.8000
144	plantnympha	183.0000
144	plantnympha	204.6000
192	plantnympha	159.6000
192	plantnympha	22.5000
192	plantnympha	263.4000
312	plantnympha	370.0000
312	plantnympha	29.7000
312	plantnympha	16.8000
2	rootsnympha	0.0000
2	rootsnympha	0.6500
2	rootsnympha	6.1200
48	rootsnympha	0.0000
48	rootsnympha	20.3500
48	rootsnympha	8.4000
96	rootsnympha	49.0200
96	rootsnympha	0.0000
96	rootsnympha	0.4600
144	rootsnympha	2.6700
144	rootsnympha	0.0000
144	rootsnympha	6.6300
192	rootsnympha	7.3800
192	rootsnympha	2.5400
192	rootsnympha	0.7400
312	rootsnympha	3.2800
312	rootsnympha	0.9900
312	rootsnympha	0.0000
2	plantpolysta	568.5000
2	plantpolysta	165.2000
2	plantpolysta	181.1000
48	plantpolysta	352.2000

APPENDIX 4: STATISTICAL OUTPUTS FOR CHAPTER 7

Raw data used for statistical tests (phase 2, chapter 7)		
Time (hours)	Planttype plant part	ug/gimiphase2diffplants
48	plantpolysta	363.0000
48	plantpolysta	417.8000
96	plantpolysta	889.1000
96	plantpolysta	660.0000
96	plantpolysta	740.7000
144	plantpolysta	834.1000
144	plantpolysta	880.2000
144	plantpolysta	850.5000
192	plantpolysta	669.2000
192	plantpolysta	651.5000
192	plantpolysta	331.7000
312	plantpolysta	1293.0000
312	plantpolysta	1101.4000
312	plantpolysta	994.5000
2	rootspolysta	5.5000
2	rootspolysta	320
2	rootspolysta	3.3000
48	rootspolysta	681.0000
48	rootspolysta	324.7000
48	rootspolysta	216.5000
96	rootspolysta	265.9000
96	rootspolysta	183.0000
96	rootspolysta	37.4000
144	rootspolysta	144.4000
144	rootspolysta	99.6000
144	rootspolysta	56.4000
192	rootspolysta	58.3000
192	rootspolysta	33.9000
192	rootspolysta	4.0000
312	rootspolysta	24.9000
312	rootspolysta	94.9000
312	rootspolysta	320

APPENDIX 5: QUESTIONNAIRE USED TO ASSESS G.A.P. OF PESTICIDE USE

Appendix 5: Questionnaire used to assess G.A.P. of pesticide use

APPENDIX 5 CONTINUED

Questionnaire used to assess G.A.P. of pesticide use

SURVEY: ONDERZOEK NAAR G.A.P.-RICHTLIJNEN BIJ LANDBOUWERS VAN HET COMMEWIJNE GEBIED (TAMANREDJO EN ALKMAAR)
Periode maart - april 2010
Begeleider: S. Mahabali (docent MW, F.Te.W, ADEK)
Datum survey: Naam LVV-begeleider:

1. DEMOGRAFISCHE -/ ALGEMENE GEGEVENS

A) Naam:
B) Adres:
C) Telefoon:
D) Leeftijd: □< 25 jr □25-40 jr □41-60 □> 60 jr
E) Geslacht: Man □ Vrouw □
F) Beroep: landbouwer Voltijds□ deeltijds□ anders□, noteer bij deeltijds, nevenberoep
G) Hoeveel dagen/ uren besteedt u in de landbouw?
H) Scholing:
I) Areaal grootte: Beplant areaal:
J) Gewas(sen) die u teelt:
K) Bodemsoort:
□ Zand □ klei □ schelp □ scherpzand
Opgebracht grondsoort:
L) Grondbewerking
□ Mechanisch □ Handbewerking □geen, reden?
M) Grondontsmetting
□ Kwekerij: ***inzaai in cups/bloembak et cetera***.

- □ Pesticiden/ chemicaliën anders dan pesticiden
- □ Geen pesticiden, reden?

□ Directe inzaai: zonder overplanten vanuit cups/bak etc.

- □ Pesticiden/ chemicaliën anders dan pesticiden
- □ Geen pesticiden, reden?

Pesticide/ chemicaliën	Reden van gebruik; tegen welke ziekte/ plaag?/ anders	Dosering ml of cup of dop/x liters per oppervlakte

APPENDIX 5 CONTINUED

Questionnaire used to assess G.A.P. of pesticide use

RASSEN EN UITGANGSMATERIAAL

1) Welke gewassen teelt u?

Gewas	**oppervlakte**	**Periode (zaaien tot oogst)**

2a) Waarom uw keus op deze gewassen (export, economisch voordeel, keuze consument, bewaarduur, impact op het milieu, weinig pesticiden gebruik etc.).

2b) Wat doet u met het geoogst product?

□ eigen gebruik □ verkoop aan □ anders:

3) Hoe komt u aan het uitgangsmateriaal?

□ zelf, hoe precies

□ gekocht, waar

4) Indien gekocht, hebt u info erbij gehad omtrent

a) Ras naam
b) Ras echtheid
c) Partijnummer
d) Herkomst

5) Is het uitgangsmateriaal (zaden/ plantjes) resistent/tolerant tegen ziekten/plagen?

□ Ja, hoe weet u dat?

□ Nee, hoe weet u dat?

6) Worden gewasbeschermingsmiddelen gebruikt gedurende de opkweek?

□ Indien ja, welke

□ Indien nee, waarom niet

7) Weet u dat er zaadjes/ plantjes verkocht worden die sneller bloeien, groetere vruchten geven, en geen vlekken krijgen et cetera?

□ Zo ja, vanwaar de info?

8) Hebt u gehoord van aanaarden? N.B. dit is het proces bij maken van bedden dat o.a. ervoor zorgt dat b.v. nutriënten niet weggewassen worden.

□ zo ja, wat is dat?

EVENTUELE OPM.

PERCEELGESCHIEDENIS EN BEHEER

9) Is het perceel uw eigendom?

□ Ja

□ Nee, grondhuur, erfpacht, anders:

10) Is de bodem geschikt/ vruchtbaar?

□ Ja, hoe weet u dat?

□ Ja, er is bodemonderzoek verricht (vermeld de instantie)

□ Nee, hoe weet u dat?

Questionnaire used to assess G.A.P. of pesticide use

11) Welke gewassen werden eerder (afgelopen 5 jaar) geteeld? Idem voor gebruikte pesticiden.

Gewas	Pesticide

12) Welke zijn de verschillende waterbronnen?

Bestemming waterbron	Soort: regen, put, grond, zwamp, loosleiding
drinkwater	
waswater	
irrigatiewater	
Spoelen toilet	

13) Waar liggen de waterbronnen (irrigatie, huishoudelijk gebruik, grondwater) t.o.v. landbouwareaal? Geef de afstand en diepte steeds aan.

Afstand drinkwaterbron tot landbouw areaal	Afstand landbouw areaal tot irrigatiebron	Diepte drinkwaterbron	Afmeting irrigatiebron (oppervlakte)	Ligt irrigatiebron lager dan areaal, ja/nee?

14) Wordt gedaan aan vruchtwisseling of wisseling van gewas?
□ indien ja, reden?
□ indien nee, waarom niet?
15) Vindt grondontsmetting plaats, zo ja, waarmee?
16) Wie geeft u advies omtrent het gebruik van meststoffen?
17) Vindt registratie van het gebruik van meststoffen plaats? Zo ja, hoe dan?

18) Kunt u een overzicht geven van de dosering + samenstelling meststof?

Questionnaire used to assess G.A.P. of pesticide use

Geef steeds de leeftijd van de plant aan alsook na hoeveel tijd weer meststof wordt toegediend.

Tijdstip	Stalmest Dosering *1) /samenstelling	Kippenmest Dosering *1) /samenstelling	Kunstmest, blauwkorrel Dosering *1) /samenstelling	Andere, b.v. ureum Dosering *1) /samenstelling
a) zaaien tot overplanten *leeftijd plant***?**				
b) overplanten tot oogsten *leeftijd plant?*				
c)directe inzaai *leeftijd plant?*				

1)** ***Met dosering wordt bedoeld, hoeveel kg per oppervlakte beplant areaal

EVENTUELE OPM.ONKRUIDBESTRIJDING

19) Doet u aan onkruidbestrijding?

□ Ja, hoe ☼) mechanisch (machine, houwer etc.)

☼) chemisch, wat precies?

□ Nee, niet nodig?

20) Welke bestrijdingsmiddelen gebruikt u tegen onkruid? Vermeld steeds de leeftijd van de plant.

Tijdstip	Bestrijdings-middel	Frequentie *)	Tegen welk onkruid (gras)	Dosering *)
a)van zaaien tot overplanten *leeftijd plant***?**				
b) van overplanten tot oogst *leeftijd plant***?**				
c)directe inzaai *leeftijd plant***?**				

*** frequentie: om de zoveel dagen/weken.**

*** dosering: met dosering wordt bedoeld hoeveel cup/dop/ml onverdunde pesticide/tankvolume (liters) per oppervlakte beplant areaal**

21) Hoe vaak (frequentie) is onkruidbestrijding nodig?

Questionnaire used to assess G.A.P. of pesticide use

□ regentijd
□ droge tijd
EVENTUELE OPM.

BEHEERSING ZIEKTEN EN PLAGEN
22) Noteer welke pesticiden gebruikt worden tegen welke ziekte/ plaag.
Code: Z1: bladkrul Z4: verwelken
Z2: groeistagnatie Z5: vlekken
Z3: smelten

Voor plagen, zie G.A.P.-boekje LVV

Tijdstip	Ziekte Code/ plaag	middel	dosering	Oppervlakte areaal	Tankinhoud (volume + totaal volume nodig per areaal)
a)van zaaien tot overplanten *leeftijd plant***?**					
b) van overplanten tot oogst *leeftijd plant***?**					
c) directe inzaai *leeftijd plant***?**					

23) Waar koopt u deze middelen?
24) Kunt u zeggen als de middelen die u gebruikt milieuvriendelijk zijn of slecht voor de gezondheid van de mens?
25) Hebt u behoefte aan info/ trainingen?
26) Weet u welke pesticiden verboden zijn in
□ Suriname:
□ Nederland
EVENTUELE OPM.

27) Worden de instructies op het etiket opgevolgd voor wat betreft dosering, frequentie van toediening en veiligheidstermijn?:

Questionnaire used to assess G.A.P. of pesticide use

Noteer voor elk bestrijdingsmiddel wat de boer opgeeft als te zijn overgenomen van het etiket voor:

Pesticide	**dosering**	**Frequentie**	**Wachttijd/ veiligheidstermijn**
1			
2			
3			

28) Weet de boer wat de gevolgen kunnen zijn voor het niet volgen van de voorschriften m.b.t. dosering, frequentie, wachttijd?

29) Hoe vindt toepassing van het bestrijdingsmiddel plaatst?

Handmatig, spuit?

30) Vindt registratie van deze applicatie plaatst?

31) Gebruikt u bij toediening van pesticiden:

□ neus/mondkapje
□ handschoenen
□ laarzen
□ lange mouw/ lange broek
□ dichte schoenen (anders dan laarzen, specificeer)
□ veiligheidsbril

32) Hoe vindt opslag van kleding + apparatuur plaats (gescheiden van chemicaliën)?

33) Wordt de kleding apart van andere was gewassen?

34)· Kan de opslag van pesticiden getoond worden(noteer nogmaals alle pesticiden). Geef een duidelijke beschrijving van de situatie, pesticide opslag, kast op slot, buiten bereik van kinderen, ventilatie goed, niet dichtbij waterbronnen et cetera.

EVENTUELE OPM.

35) Zijn er noodvoorzieningen bij een eventuele Spill? Bijvoorbeeld een emmer met zand, oogdouche?

36) Wie heeft toegang tot de pesticide opslagruimte?

37) Wat doet de landbouwer met resten pesticide en lege verpakkingen?

- Resten:
- Lege verpakkingen
- Oude flessen met of zonder pesticiden

Questionnaire used to assess G.A.P. of pesticide use

IRRIGATIE

38) Geef het tijdstip, de methode en bron voor irrigatie aan, vermeld nota bene steeds de dosering van irrigatiewater per beplant oppervlakte

Tijdstip	METHODE *)	HERKOMST WATER*)	Dosering/AREAAL, indien moeilijk, noteer hoeveel duim de pomp is en hoe lang (uren) per dag het werkt
Van zaaien tot overplanten *leeftijd plant***?**			
Overplanten tot oogst *leeftijd plant***?**			
Directe inzaai *leeftijd plant***?**			

)Methode: sproei, handmatig, pomp, drip irrigatie, anders (emmer)
)Herkomst water: put, kreek, loosleiding, zwamp, anders)
EVENTUELE OPM.

39) In geval de herkomst van het water een loosleiding is, wat is de reden?
40) Wanneer vindt irrigatie plaats in relatie tot:
a) Pesticiden gebruik:
- Voor pesticiden gebruik, reden?
- Na pesticiden gebruik, reden?

b) Regenval: vindt bv. geen irrigatie plaats of minder?
- Is er sprake van wateroverlast? Hoe vaak?

c) droge tijd: is er bv. watertekort?

41) Maak een schets van het drainagesysteem op het areaal.
- ❖ Is er b.v. oppervlakteafvoer (geen greppels)?
- ❖ of wordt het water geleid naar een bufferende vijver of sloot of
- ❖ rechtstreeks naar oppervlaktewater?

42) Doet u aan naoogst behandeling met pesticiden, zo ja waartegen?
43) Houdt u rekening met de dosering en wachttijd?
44) Vindt er registratie plaats van deze naoogstbehandelingen?
45) Waarom hebt u zich als G.A.P.-boer geregistreerd?
46) Wat ziet u graag veranderd in de landbouw?

Appendix 6: Data used to construct boxplots showing the distribution of DT_{50} values in chapter 5 and 6

Data used to construct boxplots showing the distribution of DT50 values in chapter 5 and chapter 6

Table f-1: DT_{50} values obtained for lambda-cyhalothrin and imidacloprid during 4 batches in the water phase of N. amazonum and E. mutata mesocosms

Batch no.	**DT_{50} Lambda-cyhalothrin**		**DT_{50} Imidacloprid**	
	N. amazonum	E. mutata	N. amazonum	E. mutata
B1	16.5	36.5	53.3	53.3
B1DUPLO	38.5	20.39	24.7	26.7
b2	22.4	19.25	139	77
b2DUPLO	10.5	25.67	69.3	99
B3	24.8	22.36	139	139
B3duplo	23.1	18.24	139	139
B4	21.7	18.24	231	231
B4duplo	19.8		173	231
			139	139
			139	116
Average ± SD	22.2 ± 7.5	23 ± 6.5	125.0 ± 60.4	125.1 ± 67.4

Table f-2: Raw data used for construction of a boxplot showing the distribution of DT_{50} values of lambda-cyhalothrin in the water phase of N. amazonum and E. mutata mesocosms

lambda-cyhalothrin					
	N. amazonum	**N. amazonum**		**E. mutata**	**E. mutata**
		diff			**diff**
MIN	10.5	**10.5**	**MIN**	18.24	**18.24**
Q1	18.975	**8.475**	**Q1**	18.745	**0.505**
MED	22.05	**3.075**	**MED**	20.39	**1.645**
Q3	23.525	**1.475**	**Q3**	24.015	**3.625**
MAX	38.5	**14.975**	**MAX**	36.5	**12.485**

Data used to construct boxplots showing the distribution of DT50 values in chapter 5 and chapter 6

Table f-3: The Inter quartile range (IQR) used for calculation of the acceptable DT_{50} range for lambda-cyhalothrin and imidacloprid

	lam-nympha	
IQR	**4.55**	
1.5IQR	**6.825**	
Q1-1.5IQR (lower limit of acceptable DT_{50} interval)		**12.15**
Q3+1.5IQR (upper limit of the acceptable DT_{50} interval)		**30.35**
	lam-E. mutata	
IQR	**5.27**	
1.5IQR	**7.905**	
Q1-1.5IQR (lower limit of acceptable DT_{50} interval)		10.84
Q3+1.5IQR (upper limit of the acceptable DT_{50} interval)		**31.92**
	imi-nympha	
IQR	**52.275**	
1.5IQR	**78.4125**	
Q1-1.5IQR		**8.31**
Q3+1.5IQR		**217.41**
	imi-E. mutata	
IQR	**56.5**	
1.5IQR	**84.75**	
Q1-1.5IQR		**-2.25**
Q3+1.5IQR		**223.75**
*** note: all values outside the red interval are outliers**		

Table f-4: Raw data used for construction of a boxplot showing the distribution of DT_{50} values of imidacloprid in the water phase of N. amazonum and E. mutata mesocosms

	imidacloprid				
	N. amazonum	**N. amazonum**		**E. mutata**	**E. mutata**
		diff			**diff**
MIN	24.7	**24.7**	**MIN**	26.7	**26.7**
Q1	86.73	**62.1**	**Q1**	82.5	**55.8**
MED	139	**52.28**	**MED**	127.5	**45**
Q3	139	**0**	**Q3**	139	**11.5**
MAX	231	**92**	**MAX**	231	**92**

Data used to construct boxplots showing the distribution of DT50 values in chapter 5 and chapter 6

Data for Chapter 6

Table f-5: DT_{50} values (h) obtained for chlorothalonil, lambda-cyhalothrin and imidacloprid during 5 batches in vegetated (P. australis) mesocosms

DT_{50} values (h) for vegetated mesocosms in chapter 6			
veg	chlo	lam	imi
B1	20.7	61.1	63
B2	9.3	68	115.5
B3	18.7	189.9	55.7
B4	20.4	43.3	55.2
B5	10.2		70.7

Table f-5: DT_{50} values (h) obtained for chlorothalonil, lambda-cyhalothrin and imidacloprid during 5 batches in non-vegetated mesocosms

DT_{50} values (h) for non-vegetated mesocosms in chapter 6			
nonveg	chlo	lam	imi
B1	21.6	68	115.5
B2	14.4	31.9	44.2
B3	17.3	149.1	55.7
B4	34	31.5	38.3
B5	12.8		86.6

Table f-6: Raw data used for construction of a boxplot showing the distribution of DT_{50} values of chlorothalonil, lambda-cyhalothrin and imidacloprid in the water phase of P. australis mesocosms

veg	**chlo**	**diff**	**lam**	**diff**	**imiveg**	**diff**
MIN	9.3	9.3	43.3	43.3	55.2	55.2
Q1	10.2	0.9	56.65	13.35	55.7	0.5
MED	18.7	8.5	64.55	7.9	63	7.3
Q3	20.4	1.7	98.48	33.93	70.7	7.7
MAX	20.7	0.3	189.9	91.43	115.5	44.8

Data used to construct boxplots showing the distribution of DT50 values in chapter 5 and chapter 6

Table f-7: Raw data used for construction of a boxplot showing the distribution of DT_{50} values of chlorothalonil, lambda-cyhalothrin and imidacloprid in the water phase of non-vegetated mesocosms

nonveg	chlo	diff	lam	diff	iminonveg	diff
MIN	12.8	12.8	31.5	31.5	38.3	38.3
Q1	14.4	1.6	31.8	0.3	44.2	5.9
MED	17.3	2.9	49.95	18.15	55.7	11.5
Q3	21.6	4.3	88.275	38.33	86.6	30.9
MAX	34	12.4	149.1	60.82	115.5	28.9

Table f-8: The Inter quartile range (IQR) used for calculation of the acceptable DT_{50} range for chlorothalonil, lambda-cyhalothrin and imidacloprid in Chapter 6

	Chlo veg	lam veg	imiveg		Chlo nonveg	lam nonveg	iminonveg
IQR	**11.10**	41.83	**15.00**	**IQR**	**7.20**	**56.48**	**42.40**
1.5IQR	**16.65**	62.74	22.50	**1.5IQR**	**10.80**	**84.71**	63.60
Q1-1.5IQR	**-6.45**	**-6.09**	**33.20**	**Q1-1.5IQR**	**3.60**	**-52.91**	**-19.40**
Q3+1.5IQR	**37.05**	**161.21**	**93.20**	**Q3+1.5IQR**	**32.40**	**172.99**	**150.20**

Appendix 7: Accumulation of pesticides in shoots, leaves, roots and sediment of a field scale SFCW

Accumulation of pesticides in shoots, leaves, roots and sediment of a field scale SFCW

Table g-1: Results for the amount of lambda-cyhalothrin (abbreviated as lam) measured in shoots and leaves and root samples (g DW) during phase 1 and phase 2

Time (h)	**Sampling point**	**phase 1**		**phase 2**			
		Nymphaea amazonum		***Nymphaea amazonum***		***Echinocloa polystachya***	
		shoots and leaves µg lam/g DW	**roots uglam/g DW**	**shoots and leaves µg lam/g DW**	**roots µg lam/g DW**	**shoots and leaves µg lam/g DW**	**roots µg lam/g DW**
2	V1	ND	0.10	ND	0.34	3.12	ND
	V2	5.16	ND	ND	ND	6.80	ND
	V3	ND	ND	ND	ND	9.17	0.99
48	V1	0.18	ND	6.85	ND	3.10	7.85
	V2	ND	ND	0.37	ND	0.03	1.12
	V3	1.22	ND	0.15	1.27	ND	0.06
96	V1	1.64	ND	2.58	0.17	5.89	0.01
	V2	ND	ND	1.22	ND	17.5	ND
	V3	ND	ND	ND	ND	9.47	ND
144	V1	ND	ND	ND	0.32	15.3	0.83
	V2	ND	ND	ND	ND	ND	0.14
	V3	ND	0.45	ND	ND	26.4	0.72
192	V1	ND	0.61	0.65	1.91	0.06	0.52
	V2	ND	ND	ND	ND	1.33	0.19
	V3	ND	ND	ND	ND	7.52	0.97
312	V1	ND	0.54	0.22	1.36	2.94	0.24
	V2	ND	ND	0.06	ND	0.09	0.11
	V3	ND	0.19	ND	5.61	ND	0.07

Accumulation of pesticides in shoots, leaves, roots and sediment of a field scale SFCW

Table g-2: Results for the amount of imidacloprid (abbreviated as imi) measured in shoots and leavess and root samples (g DW) during phase 1 and 2

time (h) after pesticide addition		**phase 1**		**phase 2**			
		Nymphaea amazonum		***Nymphaea amazonum***		***Echinochloa polystachya***	
	Sampling point	**shoots and leaves µg imi/g**	**roots µg imi/g**	**shoots and leaves µg imi/g**	**roots µg imi/g**	**shoots and leaves µg imi/g**	**roots µg imi/g**
2	V1	12.85	11.46	4.9	ND	568.5	5.5
	V2	19.04	22.28	33.7	0.65	165.2	32.0
	V3	10.97	9.74	101.1	6.12	181.1	3.3
48	V1	19.60	17.49	700.3	ND	352.2	681.0
	V2	173.22	202.71	225.2	20.35	363.0	324.7
	V3	101.23	89.93	1071.9	8.40	417.8	216.5
96	V1	76.33	68.08	584.0	49.02	889.1	265.9
144	V1	152.91	136.39	419.8	2.67	834.1	144.4
	V2	334.57	391.53	183.0	ND	880.2	99.6
	V3	599.05	532.17	204.6	6.63	850.5	56.4
192	V1	182.03	162.36	159.6	7.38	669.2	58.3
	V2	782.39	915.59	22.5	2.54	651.5	33.9
	V3	317.70	282.23	263.4	0.74	331.7	4.0
312	V1	277.01	247.07	370.0	3.28	1293.0	24.9
	V2	741.69	867.96	29.7	0.99	1101.4	94.9
	V3	143.82	127.76	16.8	ND	994.5	32.0

Table g-3: Amount of lambda-cyhalothrin and imidacloprid detected in the sediment of different sampling locations and expressed as µg lam/g sed and µg imi/g sed

time (h)	**sampling point**	**phase 1**	**phase 2**	**phase 1**	**phase 2**
		µg lam/g sed		**µg imi/g sed**	
2	B*	4.56	1.21	33.9	2.36
	ditch1	62.3	ND	6.54	1.32
	ditch2	297	ND	350	0.00
	inflow	10.5	ND	346	3.08
	V1	61.7	0.041	0.53	0.32
	V2	32.3	ND	4.38	0.42
	V3	0.70	2.47	ND	2.27
48	B	133	7.51	98.9	72.5
	ditch1	40.3	1.81	7.63	15.0

Accumulation of pesticides in shoots, leaves, roots and sediment of a field scale SFCW

Table g-3: Amount of lambda-cyhalothrin and imidacloprid detected in the sediment of different sampling locations and expressed as µg lam/g sed and µg imi/g sed

time (h)	sampling point	phase 1	phase 2	phase 1	phase 2
		µg lam/g sed		µg imi/g sed	
	ditch2	1.22	ND	2.26	12.7
	inflow	37.2	ND	32.3	0.17
	V1	41.1	0.36	3.29	23.7
	V2	20.8	23.0	0.65	2.97
	V3	0.014	0.64	0.83	6.98
96	B	123	36.1	84.0	135
	ditch1	45.8	3.87	11.6	ND
	ditch2	57.1	24.7	43.9	8.62
	inflow	155	3.12	23.0	1.35
	V1	52.2	37.9	29.6	7.35
	V2	27.2	3.95	23.1	0.03
	V3	33.3	55.7	2.50	96.3
144	B	63.9	4.09	70.0	259
	ditch1	46.1	0.09	34.4	16.4
	ditch2	3.23	0.80	38.7	2.63
	inflow	15.4	29.3	0.91	ND
	V1	47.3	1.31	2.62	7.01
	V2	41.6	96.7	0.57	11.6
	V3	71.3	3.99	0.48	0.00
192	B	10.2	3.14	26.9	33.5
	ditch1	0.27	7.47	2.67	0.66
	ditch2	0.11	6.89	3.47	14.28
	inflow	0.85	54.4	171	0.00
	V1	0.16	18.5	341	0.00
	V2	1.11	6.77	406	0.04
	V3	0.46	7.50	252	0.19
312	B	9.54	18.2	38.0	9.11
	ditch1	44.5	52.3	0.64	0.12
	ditch2	51.2	8.96	0.44	1.16
	inflow	31.5	15.6	14.9	0.09
	V1	1.30	0.94	0.15	0.02

Accumulation of pesticides in shoots, leaves, roots and sediment of a field scale SFCW

Table g-3: Amount of lambda-cyhalothrin and imidacloprid detected in the sediment of different sampling locations and expressed as µg lam/g sed and µg imi/g sed

time (h)	**sampling point**	**phase 1**	**phase 2**	**phase 1**	**phase 2**
		µg lam/g sed		**µg imi/g sed**	
	V2	0.39	18.9	1.44	0.04
	V3	2.92	8.07	19.0	0.09

Calculated amounts of pesticides in runoff (ditch 1 and ditch 2) in phase 1 and phase 2

Table g-4: Amounts of imidacloprid in water of ditch 1 and in runoff (phase 1)

Time (h)	**mg/l**	**mg in ditch 1[a]**	**g in ditch 1**	**% of applied amount in ditch water[b]**	**% of applied amount in runoff**
2	2.10	4529.52	4.53	4.12	0.74
24	0.45	980.64	0.98	0.89	0.16
48	0.46	999.00	1.00	0.91	0.16
72	0.15	331.56	0.33	0.30	0.05
96	0.50	1086.48	1.09	0.99	0.18
144	0.06	137.16	0.14	0.12	0.02
192	0.08	180.36	0.18	0.16	0.03
240	0.10	220.32	0.22	0.20	0.04

[a]*Calculated by multiplying the concentration with the total water volume (2160 liters) in ditch 1*
[b]*The applied amount is 110 grams*
[c]*The amount of runoff equals 392 liters. This value divided by the total ditch volume gives a conversion factor of 0.18 used to multiply the% of applied amount in ditch water. The output is the % of the applied amount in runoff*

Table g-5: Amounts of imidacloprid in water of ditch 2 and in runoff (phase 1)

Time (h)	**mg/l**	**mg in ditch 2[a]**	**g in ditch 2**	**% of applied amount in ditch water[b]**	**% of applied amount in runoff**
2	1.82	2753.49	2.75	2.50	0.65
24	0.17	258.21	0.26	0.23	0.06
48	0.21	317.1	0.32	0.29	0.07
72	0.25	372.97	0.37	0.34	0.09
96	0.25	371.46	0.37	0.34	0.09
144	0.11	163.84	0.16	0.15	0.04
192	0.11	172.14	0.17	0.16	0.04
240	0.07	98.15	0.10	0.09	0.02
312	0.03	40.02	0.04	0.04	0.01

[a]Calculated by multiplying the concentration with the total water volume (1510 liters) in ditch 2
[b]*The applied amount is 110 grams*
[c]*The amount of runoff equals 392 liters. This value divided by the total ditch volume gives a conversion factor of 0.26 used to multiply the % of applied amount in ditch water. The output is the % of the applied amount in runoff*

Calculated amounts of pesticides in runoff (ditch 1 and ditch 2) in phase 1 and phase 2

Table g-6: Total amount of imidacloprid in runoff in phase 1. This value is the sum of% of applied amount in runoff of ditch 1 + ditch 2

Time (h)	% of applied amount in runoff (ditch 1)	% of applied amount in runoff (ditch 2)	Total% in runoff
2	0.74	0.65	1.4
24	0.16	0.06	0.2
48	0.16	0.07	0.2
72	0.05	0.09	0.1
96	0.18	0.09	0.3
144	0.02	0.04	0.1
192	0.03	0.04	0.1
240	0.04	0.02	0.1

Table g-7: Amounts of lambda-cyhalothrin (lamda) in water of ditch 1 and in runoff (phase 2)

Time (h)	lambda mg/l ditch 1	amount in ditch 1 (mg)[a]	amount in ditch 1 (g)	% of applied amount in ditch water[b]	% of applied amount in runoff
20	0.0934	201.8088	0.2018	3.2289	0.5812
24	0.0163	35.1457	0.0351	0.5623	0.1012
48	0.0283	61.1828	0.0612	0.9789	0.1762
144	0.0000	0.0000	0.0000	0.0000	0.0000
1920	0.0000	0.0000	0.0000	0.0000	0.0000
3120	0.0000	0.0000	0.0000	0.0000	0.0000

[a]Calculated by multiplying the concentration with the total water volume (2160 liters) in ditch 1
[b]The applied amount is 6.25 grams/459 m^2
[c]The amount of runoff equals 392 liters. This value divided by the total ditch volume gives a conversion factor of 0.18 used to multiply the% of applied amount in ditch water. The output is the% of applied amount in runoff

Table g-8: Amounts of lambda-cyhalothrin in water of ditch 2 and in runoff (phase 2).

Time (h)	Lambda in mg/l ditch 1	Amount in ditch 2 (mg)[a]	Amount in ditch 2 (g)	% of applied amount in ditch water [b]	% of applied amount in runoff	Total amount in runoff (%)[d]
2	0.0000	0.0000	0.0000	0.0000	0.0000	**0.5812**
24	0.0069	10.4722	0.0105	0.1676	0.0436	**0.1448**
48	0.0156	23.5031	0.0235	0.3760	0.0978	**0.2740**
144	0.0000	0.0000	0.0000	0.0000	0.0000	**0.0000**
192	0.0000	0.0000	0.0000	0.0000	0.0000	**0.0000**
312	0.0000	0.0000	0.0000	0.0000	0.0000	**0.0000**

[a]Calculated by multiplying the concentration with the total water volume (1510 liters) in ditch 2

Calculated amounts of pesticides in runoff (ditch 1 and ditch 2) in phase 1 and phase 2

[b]The applied amount is 6.25 grams/459 m²
[c]The amount of runoff equals 392 liters. This value divided by the total ditch volume gives a conversion factor of 0.26 used to multiply the% of applied amount in ditch water. The output is the% of applied amount in runoff
[d]The total amount found in runoff is the sum of the% of the applied amount found in ditch 1 and ditch 2

Table g-9: Amounts of imidacloprid(imi) in water of ditch 1 and in runoff (phase 2)

time (h)	Imi in mg/l	Amount in ditch 1 (mg)[a]	Amount in ditch 1 (g)	% of applied amount in ditch water[b]	**% of applied amount in runoff[c]**
2	9.68	20898.00	20.90	19.00	**3.42**
24	2.52	5452.92	5.45	4.96	**0.89**
48	2.04	4397.76	4.40	4.00	**0.72**
72	2.16	4675.32	4.68	4.25	**0.77**
96	1.65	3570.48	3.57	3.25	**0.58**
144	0.16	346.68	0.35	0.32	**0.06**
192	0.11	234.36	0.23	0.21	**0.04**
240	0.86	1852.20	1.85	1.68	**0.30**
312	0.00	0.00	0.00	0.00	**0.00**

[a]Calculated by multiplying the concentration with the total water volume (2160 liters) in ditch 1
[b]The applied amount is 110 grams
[c]The amount of runoff equals 392 liters. This value divided by the total ditch volume gives a conversion factor of 0.18 used to multiply the% of applied amount in ditch water. The output is the% of applied amount in runoff

Table g-10: Amounts of imidacloprid(imi) in water of ditch 2 and in runoff (phase 2)

time (h)	Imi in mg/l	Amount in ditch 2 (mg)[a]	Amount in ditch 2 (g)	% of applied amount in ditch water[b]	% of applied amount in runoff[c]	Total% in runoff (sum of ditch 1 + 2)[d]
2	6.45	9743.28	9.74	8.86	2.30	**5.72**
24	2.81	4239.33	4.24	3.85	1.00	**1.89**
48	1.19	1790.86	1.79	1.63	0.42	**1.14**
72	1.86	2815.40	2.82	2.56	0.67	**1.43**
96	0.32	477.16	0.48	0.43	0.11	**0.70**
144	0.23	340.51	0.34	0.31	0.08	**0.14**
192	0.19	292.94	0.29	0.27	0.07	**0.11**
240	0.98	1481.31	1.48	1.35	0.35	**0.65**
312	0.61	919.59	0.92	0.84	0.22	**0.22**

[a]Calculated by multiplying the concentration with the total water volume (1510 liters) in ditch 1

Calculated amounts of pesticides in runoff (ditch 1 and ditch 2) in phase 1 and phase 2

[b]The applied amount is 110 grams/459 m^2
[c]The amount of runoff equals 392 liters. This value divided by the total ditch volume gives a conversion factor of 0.26 used to multiply the% of applied amount in ditch water. The output is the% of applied amount in runoff
[d]The total amount found in runoff is the sum of the% of the applied amount found in ditch 1 and ditch 2

Table g-11: Total amount of imidacloprid found in runoff during phase 1 and phase 2

Time (h)	**Total% imidacloprid in runoff (phase 1)**	**Total% imidacloprid in runoff (phase 2)**
2	**1.4**	**5.7**
24	**0.2**	**1.9**
48	**0.2**	**1.1**
72	**0.1**	**1.4**
96	**0.3**	**0.7**
144	**0.1**	**0.1**
192	**0.1**	**0.1**
240	**0.1**	**0.7**
312	**NM[a]**	**0.2**

[a]Not Measured

CURRICULUM VITAE

PERSONAL DETAILS

Surname:	Mahabali
First names	Shirley Sahina
Date of birth	1st June 1970
Place of birth	Suriname
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EDUCATION

1988-1994	Instituut voor de Opleiding van Leraren (I.O.L.) Chemistry course (LO-II and MO-A level)
1993	LO-II certificate in Chemistry Certificate "Pedagogisch Getuigschrift"
1994	MO-A certificate in Chemistry
1999-2001	Course MO-B in Chemistry
May 2001	MO-B certificate in Chemistry (according to NUFFIC (Netherlands University Foundation for International Cooperation) comparable to a MSc. in Chemistry)
2002-2004	MSc. Environmental Sanitation. Ghent University, Belgium
2009-Present	PhD studies. Doctoral Schools of Bioscience Engineering, Ghent University, Belgium.

SHORT COURSES AND SURVEYS

1996	Training Analytical Chemistry, V.U.B. (Belgium)
1997	Training pesticides residues in food by means of TLC, Anton de Kom University of Suriname
2001	Training atomic absorption and gas chromatography, Anton de Kom University of Suriname in collaboration with Hoge School Zeeland, Paramaribo, Suriname
2005	Survey: Pesticide use on cabbage (*Brassica oleracea* var. capitata) and tomato (*Lycopersicon esculentum*) in the Kwatta area, Suriname Survey: Investigation of different routes of pesticide contamination in the "Bigi Pan Wetland" in the Nickerie district Survey: The implementation of a Pesticide Residue Laboratory using "Good Laboratory Practices" Survey: Vegetables and the use of pesticides; conducted for three markets in Suriname
2007	Improving Municipal Wastewater Management in Suriname, collaboration between Unesco-IHE, UN/DOALOS, UNEP/GPA, ADEKUS and the Ministry of Labour, Paramaribo, Suriname
2008	Survey: Women and pesticide use in the Commewijne district. (Under supervision of Dr. H. van de Lande) Training sustainable sanitation alternatives, Costa Rica (Acepesa) and Nicaragua (Habitar)

	Training R-statistics, Ghent University, Belgium
2011	Course: Creative thinking, Ghent University, Belgium
	Hydrus training course: simulating soil water movement & chemical transport using Hydrus & the biogeochemical model HP1, Adelaide, Australia
	Course: Scientific writing and publishing, Institute for Graduate Studies (IGSR), Paramaribo, Suriname
2012	Course: Environmental impact of pesticides, Ghent University, Belgium
	Course: Dynamica en residu's of pesticides, Ghent University, Belgium
	Workshop model based geostatistics with epidemiological applications, Ghent University, Belgium

EMPLOYMENT RECORD

1991-1994	Full-time assistant to the Chemical laboratory of the Faculty of Technology of the Anton de Kom University of Suriname
	Part-time assistant to the practical chemistry courses of the I.O.L[*1], LOMBO[*2], and the preparatory year of the University
	Part-time teacher at the I.O.L., MO-A Biology course
1994-2002	Full-time laboratory manager of the Chemical laboratory of the Faculty of Technology of the Anton de Kom University of Suriname
	Part-time Chemistry teacher at AMTO[*3], Suriname
	Part-time teacher "instrumentenkennis" at NATIN[*4], Suriname
2004-2013	Part-time teacher Biochemistry at the IOL, Biology course
2009-2012	Examinator for the Chemistry exams of the secondary schools in Suriname
2004-up till now	Lecturer/researcher at the Environmental Sciences Department of the Anton de Kom University of Suriname. Lecturer Water Quality Management, Soil Pollution and Sanitation (co-teacher), Air Pollution and Sanitation, Environmental Chemistry, and Introduction to Environmental Ethics
2013-up till now	Lecturer Environmental Pollution and Sanitation for the Masters course "Sustainable Management of Natural Resources".

*1 *I.O.L: Instituur voor de Opleiding van Leraren (tertiary educational level)*

*2 *LOMBO: Leraren Opleiding Middelbaar Beroeps onderwijs (secondary educational level)*

*3 *AMTO: Avond Middelbaar Technisch Onderwijs (secondary educational level)*

*4 *NATIN: Natuur Technisch Instituut (secondary educational level)*

LIST OF PUBLICATIONS

Mahabali, S.S. and Spanoghe, P., 2015. Risk assessment of pesticide usage by farmers in Commewijne, Suriname, South America: a pilot study for the Alkmaar and Tamanredjo regions. Environmental monitoring and assessment, 187 (3): 153, 12 pages

Mahabali, S.S. and Spanoghe, P., 2014. Mitigation of two insecticides by wetland plants: feasibility study for the treatment of agricultural runoff in Suriname (South America). Water Air and Soil Pollution, 225: 1771, 12 pages. DOI10.1007/s11270-013-1771-2

Mahabali, S.S., 2004. Effects of low concentrations of odorous compounds on the functional and microbiological characteristics of a biofilter. Thesis MSc. Environmental Sanitation, Ghent University, Belgium

Mahabali, S.S., 2001. Mercury in the aquatic environment. Thesis MO-B Chemistry course, Instituut voor de Opleiding van Leraren, Paramaribo, Suriname

CONFERENCE PRESENTATIONS AND CONTRIBUTIONS

Mahabali, S.S. and Spanoghe, P., 2010. Constructed wetlands for the treatment of agricultural runoff in Suriname, Society of Wetland Scientists (SWS) 2010 annual meeting, June 27-July 2, Salt Lake City, UT, United States of America

Mahabali, S.S. and Spanoghe, P., 2011. Agricultural practices by farmers in Suriname: feasibility of constructed wetlands for the treatment of agricultural runoff, World Environmental and Water Resources Congress, May 22-26, Palm Springs, CA, United States of America

Mahabali, S.S. and Spanoghe, P., 2011. Feasibility of constructed wetland mesocosms to treat agricultural runoff in Suriname, Joint meeting of SWS, WETPOL and Wetland Biogeochemistry Symposium, July 3-8, Prague, Czech Republic

Mahabali, S.S. and Spanoghe, P., 2012. Mitigation of two insecticides by wetland plants. The 9th INTECOL International Wetlands Conference, June 3-8, Orlando, Florida, United States of America

Mahabali, S.S., 2012. Poster and oral presentation. Good agricultural practices of pesticides use under farmers in the Commewijne district. Annual National Congress VLIR-UOS-Adekus, IGSR, Paramaribo, Suriname

Moderator during day 1 of the Caribbean Food Crops Society 51st Annual Meeting. July 19-24, 2015, Paramaribo, Suriname