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# The use of plant protection products in Flanders and risks for humans and the environment

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

### Dutch translation of the title:

Het gebruik van gewasbeschermingsmiddelen in Vlaanderen en de risico's voor mens en milieu.

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"That's what all we are. Amateurs. We don't live long enough to be anything else." Charlie Chaplin (1889-1977)

"If you can't explain it simply, you don't understand it well enough."

Albert Einstein (1879-1955)

"There is no magic to achievement. It is really hard work, choices and persistence." Michele Obama

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**Hoofdstuk 1** geeft een algemene inleiding over het gebruik van GBM en de geldende wetgeving. Daarnaast onthult **Hoofdstuk 1** ook het gebrek aan kennis over GBM gebruikt voor de behandeling van zaaizaden en niet-landbouwkundige doeleinden. In de volgende hoofdstukken wordt het gebruik van GBM in al zijn aspecten verder onderzocht, om zo het totale gebruik van GBM nauwkeuriger te kunnen schatten.

De verkoop en het gebruik van GBM in Vlaanderen worden toegelicht in **Hoofdstuk 2**. Daarnaast wordt ook de druk van het gebruik van GBM op het waterleven geïllustreerd in **Hoofdstuk 2**. In Vlaanderen wordt een risico-indicator op basis van spreidingsequivalenten ( $\sum$ Seq) toegepast in het milieubeleid. Deze Seq-indicator wordt bepaald door het gebruik van werkzame stoffen te wegen op hun toxiciteit voor waterorganismen en hun verblijftijd in het milieu. Een accurate schatting van het totale gebruik van GBM per regio is essentieel in dergelijke berekeningen. Het doel van dit onderzoek was dan ook om de huidige gebruikte Seq-indicator te verfijnen om zo een verbeterde weergave van de toestand van het milieu te bekomen. Een schatting van het totaal gebruik van GBM, in dit geval schattingen op basis van verkoopcijfers, werd met gebruikscijfers op basis van boekhoudkundige gegevens, zoals het Landbouwmonitoringsnetwerk vergeleken. De resultaten van dit hoofdstuk toonden hoofdzakelijk het verschil tussen gebruiks- en verkoopcijfers van GBM aan. Het geschatte gebruik van GBM op basis van boekhoudkundige gegevens is meer geschikt in vergelijking met verkoopcijfers om risicoberekeningen uit te voeren. Deze aanpak heeft geleid tot een betere kijk op het gebruik van GBM en de daarmee gepaarde milieudruk in Vlaanderen.

Volgens Richtlijn 2009/128/EG moet het bewustzijn en het gedrag van gebruikers van GBM op het niveau van het landbouwbedrijf worden verbeterd. Op basis van huidige beschikbare gegevens is het echter moeilijk om aangewende maatregelen op het landbouwbedrijf in te schatten. Daarom werden Vlaamse landbouwers bevraagd over hoe ze precies omgaan met GBM. Deze inzichten in hun gewasbeschermingspraktijken werden vervolgens gebruikt om risicobeoordelingen op het niveau van het landbouwbedrijf te verbeteren. Bestaande enquêtes over het gebruik van GBM in de hele Europese Unie verschaffen weinig informatie over de wijze waarop producten door landbouwers toegepast worden, details van maatregelen die aangewend worden om het risico te reduceren, het aantal gewerkte uren, het specifieke ogenblik van toepassing en andere werkzaamheden uitgevoerd door de landbouwer die kunnen bijdragen aan de blootstelling. **Hoofdstuk 3** begint met een

#### Samenvatting

beschrijving van de verzameling van data betreffende het toepassen van GBM in Vlaanderen, uitgevoerd in het kader van een project gefinancierd door EFSA (European Food Safety Authority). Informatie over verschillende factoren werd verzameld, nl. het aantal gewerkte uren per dag voor de belangrijkste toepasser, alle andere uitgevoerde landbouwactiviteiten naast het toepassen van GBM, de gehanteerde persoonlijke bescherming en details van de spuittoestellen. Bovendien werd ook specifieke informatie over de verschillende GBM toepassingen op 20 milieuvelden van maïs, aardappel en suikerbiet in 2013 verzameld. Deze gegevens werden vergeleken met milieuvelden van andere landen die soortgelijke gewassen verbouwen. Het tweede deel van Hoofdstuk 3 beschrijft hoe bekomen informatie (door middel van enquêtes) werd gehanteerd om de berekeningen van POCER (Pesticide OCcupational and Environmental Risk indicator) te optimaliseren. Eerst werd een casestudy van fruitteelt in het kader van DISCUSS (Dual Indicator Set for Sustainable Crop protection Sustainability Surveys) uitgevoerd. Simulaties met persoonlijke bescherming en driftmaatregelen illustreerden hoe DISCUSS kan worden toegepast om beslissingen van de landbouwers omtrent duurzame gewasbescherming te ondersteunen. Ten tweede werd ook een casestudy van aardappelen, aan de hand van informatie bekomen door face-to-face interviews (EFSA project), uitgevoerd. Dit met als beoogde doel om POCER berekeningen verder te verfijnen. Beide casestudies toonden aan hoe weloverwogen wijzigingen in gewasbeschermingspraktijken risico's kunnen verminderen op mens en milieu.

In Hoofdstuk 4 wordt achtergrondinformatie over zaaizaadbehandeling met GBM beschreven. Zaaizaadbehandeling is een wijdverspreide en effectieve manier om verschillende ziekten en plagen te bestrijden aan de hand van lagere dosissen met potentieel minder schadelijke nevenwerkingen. De belangrijkste nadelen van deze techniek zijn allemaal geassocieerd met de coating van het zaaizaad. Residuen van systemische GBM kunnen aanwezig zijn in de guttatievloeistof, stuifmeel en nectar van planten waarvan het zaaizaad behandeld werd. In de afgelopen jaren lijkt deze emissie te hebben geleid tot onder andere het verlies van bijen in verschillende landen en de verontreiniging van het oppervlaktewater. Om na te gaan of het gebruik van zaadbehandelingsproducten – anders dan neonicotinoïden – ook een druk op bestuivers uitoefenen, werden verschillende experimenten met behandeld zaad uitgevoerd. De opname, translocatie en persistentie in de plant variëren sterk tussen verschillende GBM. In dit hoofdstuk werden deze drie factoren onderzocht op verschillende gewassen met speciale aandacht voor het onderzoek naar residuen van methiocarb in maïs. Daarnaast werd ook een residuanalyse van bijenwas en bijenbrood uit verschillende bijenkorven gevestigd in Vlaanderen uitgevoerd. De resultaten van dit hoofdstuk illustreren dat verschillende werkzame stoffen, gebruikt als zaaizaadbehandelingsproduct, verplaatst worden doorheen de plant. Geen enkel spoor van neonicotinoïden werd gevonden en geen enkele van alle gedetecteerde stoffen in bijenbrood en bijenwas zijn erkend voor de behandeling van zaaizaden in België. Op basis van de resultaten uit dit hoofdstuk is het risico van blootstelling aan bestuivers via zaaizaadbehandeling zeer laag voor de gewassen en werkzame stoffen onderzocht in deze studie.

Hoofdstuk 5 start met een identificatie en toewijzing van het niet-landbouwkundig gebruik van GBM in België, uitgevoerd in het kader van een pilotstudie van Eurostat, wegens het gebrek aan kennis over het gebruik van GBM voor niet-landbouwkundige doeleinden. Private bedrijven voor tuinonderhoud, private bedrijven en amateurgebruikers zijn de belangrijkste niet-landbouwkundige gebruikers van GBM waarvoor een combinatie van administratieve data (voor amateurgebruikers) met enquêtes vereist is om gegevens te verzamelen betreffende het niet-landbouwkundig GBM gebruik. Vervolgens werd één van de belangrijkste niet-landbouwkundige gebruikers verder onderzocht, nl. niet-professionele gebruikers en meer bepaald amateurgebruikers die geïdentificeerd werden in het eerste deel van dit hoofdstuk. De resultaten van het eerste deel van dit hoofdstuk suggereren om verkoopcijfers van niet-professionele GBM te hanteren om het niet-professioneel GBM gebruik te bepalen. Op basis van deze resultaten werd in het tweede deel van dit hoofdstuk een schatting van de druk van het niet-professioneel gebruik van GBM op toepassers, bijen waterorganismen in België vastgesteld. Zowel verkoopcijfers als drie en blootstellingsmodellen werden gebruikt en een indeling in niet-professioneel gebruik werd gemaakt op basis van type GBM, toepassingsmethode en intensiteit van het nietprofessioneel gebruik. Algemeen werd vastgesteld dat het totale gebruik en de druk van GBM daalde voor de bestudeerde periode als gevolg van inspanningen van zowel de overheid als de industrie. Bovendien is de combinatie van niet-professionele GBM verkoopcijfers met meer gedetailleerde informatie over hoe niet-professionele gebruikers omgaan met GBM aanbevolen. Kennis over huis- en tuingebruik van GBM door amateurgebruikers in België is beperkt. Hierdoor werd het gebruik van GBM door amateurgebruikers in Vlaanderen onderzocht met behulp van een enquête. De vragenlijst werd beknopt gehouden om de hoogst mogelijke respons te bekomen en werd door middel van drie verschillende onderzoeksmethoden uitgevoerd. Uit de resultaten van de enquête blijkt dat de kennis over de verschillende gevaarsymbolen op het etiket van GBM nog onvoldoende is. Een 100% bescherming van de gebruikers tijdens de aanmaak en toepassing van GBM moet verder nagestreefd worden. Amateurgebruikers kunnen de druk op het milieu verminderen door bewust om te gaan met spuitresten en spoelwater van het gebruikte GBM. Het blijft een uitdaging om de amateurgebruiker beter te informeren door middel van eenvoudigere etiketten op de GBM en advies te verstrekken hoe ze correct moeten omgaan met GBM op vlak van veiligheid voor zowel mens als milieu. De resultaten van dit hoofdstuk illustreren ook het belang van het verzamelen van een combinatie van administratieve data met enquêtes om het niet-landbouwkundig gebruik van GBM nauwkeuriger te kunnen schatten.

Om af te sluiten werden de belangrijkste resultaten van dit doctoraat samengevat en besproken in **Hoofdstuk 6** en werden er enkele suggesties naar verder onderzoek voorgesteld.

This doctoral dissertation presents the work that has been done to improve the estimation of plant protection product (PPP) use in Flanders in order to respect the requirements stipulated in Directive 2009/128/EC. These usage estimates are used to perform risk assessments to indicate the pressure of PPPs on human health and the environment in the framework of policy decisions.

**Chapter 1** serves as general introduction to PPP use and its prevailing legislation. This chapter also reveals the lack of knowledge on PPPs used for seed treatment and non-agricultural purposes. The following chapters further investigate PPP use in all its aspects in order to collect accurate usage estimates which are essential to all PPP risk indicator calculations.

In **Chapter 2**, background information about the sales and use of PPPs in Flanders is presented. The pressure exerted by PPP use on aquatic life was calculated as well. Pressure of PPPs on the environment is quantified by means of indicators, which typically require weighting environmental exposure by a no effect concentration. In Flanders, an indicator based on spread equivalents ( $\Sigma$ Seq) is used in environmental policy. This Seq-indicator is estimated by weighting the use of each active substance by the ratio of their toxicity to aquatic organisms and their residence time in the environment. Accurate PPP use estimates in the region are essential in such calculations. The main goal of this study was to modify the currently used Seq-indicator in order to better reflect the state of the environment. Total PPP use estimates, in this case estimates based on PPP sales, were compared to estimates based on usage registration like Farm Accountancy Data Network. In general, this chapter showed the difference between use estimates and sales figures of PPPs. The estimated use of PPPs based on accountancy data is more appropiate compared to sales figures in order to perform risk calculations. This approach resulted in a better view on PPP use and its respective environmental pressure in Flanders.

According to Directive 2009/128/EC, the awareness and behaviour of PPP users at farm level has to be improved. However, it is difficult to directly assess on-farm response measures from any data currently available. Therefore, Flemish farmers were questioned about their behaviour related to PPP handling and application. These insights in their crop protection practices were then used to improve risk assessments at farm level. Some countries in the European Union already perform PPP usage surveys. However, these surveys provide little information on how products are applied by operators nor do they give details of the mitigation measures used to reduce exposure, hours worked, specific times of application or other working activities performed by the operator that may contribute to the exposure. **Chapter 3** starts off with a description of the PPP application data collection in Flanders performed in the context of a project funded by the European Food Safety Authority (EFSA).

#### Summary

Information was collected in 2013 on a wide range of factors for operators such as the number of hours worked each day for the specific principle operator, personal protective equipment used, other worker activities and details of sprayers. Furthermore, specific information on the multiple PPP applications to twenty environmental fields of maize, potato and sugar beet was collected as well. This information was compared with the environmental fields of other countries cultivating similar crops. The second part of Chapter 3 describes how collected information (by means of questionnaires) was used to refine calculations of the Pesticide OCcupational and Environmental Risk indicator (POCER). It showed how the trump of having additional response information improved POCER as a risk indicator in the framework of sustainable PPP use. To start off, a case study of fruit production was performed in the context of DISCUSS, the Dual Indicator Set for Sustainable Crop protection Sustainability Surveys. Simulations with drift mitigation measures and personal protection illustrated how DISCUSS can be used to support the farmers' sustainable crop protection decision-making process. Subsequently, information collected by means of face-to-face interviews conducted in the context of the EFSA project was used to perform a case study of potatoes in order to upgrade POCER as well. Both case studies illustrated how responsive modifications in crop protection practices can reduce human and environmental risk.

In Chapter 4, background information about seed treatment with PPPs is presented. Seed treatment is a widespread and effective way to control various pests and diseases using smaller doses with potentially less harmful side-effects. Main disadvantages of this technique are all related to the coating of the seed. Residues of systemic active substances can be present in the guttation fluid, plant pollen and nectar of seed treated plants. Several incidents of bee losses have recently occurred that seemed to be caused by this emission. In order to verify if the use of seed treatment products - other than neonicotinoids - also expert pressure on pollinators, several experiments with treated seed were performed. The uptake, translocation and persistence in the plant vary among various seed treatment products. These three factors were examined for various crops, with focus on the investigation of methiocarb residues in maize. Residue analysis of beeswax and bee bread from different hives located in Flanders was performed as well. The results of this chapter illustrate that several active substances, used as seed treatment products, are translocated through the plant. No traces of neonicotinoids were found and not one of all detected substances in bee bread and beeswax is authorised in the treatment of seeds in Belgium. Based on the results of this chapter, the risk of exposure to pollinators by means of seed treatment is supposed to be very low for the crops and active substances investigated in this study.

**Chapter 5** starts off with an identification and allocation of non-agricultural PPP use in Belgium performed in the framework of a pilot study financed by Eurostat, since a lack of knowledge on PPPs used for non-agricultural purposes still exists. Key players in the non-agricultural use of PPPs were put forward, i.e. private companies of parks and gardens,

private companies and amateur gardeners. These interlocutors require a combination of administrative data (for amateur gardeners) with inquiries for all kind of private companies in order to collect data on the non-agricultural use of PPPs. Subsequently, one of the key players in the non-agricultural PPP use was further investigated, i.e. non-professional users and more precisely amateur gardeners which were identified in the first part. The results of the first part of this chapter suggest to use sales figures of non-professional PPPs to estimate non-professional PPP use. Based on these results, an estimation of the pressure of nonprofessional use of PPPs on operators, aquatic organisms and bees in Belgium was conducted in the second part of this chapter. Both sales figures and three exposure models were used and a classification in non-professional use was made based on type of PPP, application method and on intensity of non-professional use. Both total usage and pressure of PPPs decreased for the period studied here due to efforts made by the government and industry. Furthermore, the combination of non-professional sales figures with more detailed information about the handling behaviour of amateur gardeners is recommended in order to provide an indication of total non-professional PPP use. The levels of knowledge, usage and awareness of amateur gardeners in Flanders were investigated by means of a questionnaire. The questionnaire was kept concise in order to have the highest response rate possible and was conducted by means of three different survey methods. Survey results illustrated that knowledge of the different hazard symbols on the PPP label is still insufficient. 100% protection of the operators during preparation and application of PPPs should definitely be pursued. The amateur gardener can reduce the pressure on the environment by consciously dealing with surpluses and rinse water of the used PPPs. It still remains a challenge to better inform the amateur gardener by providing easier to understand PPP labels and to provide advice on how to deal with PPPs both in terms of safety for humans and the environment. The results of this chapter also illustrate the importance of collecting additional data by a combination of administrative data with inquiries in order to be able to estimate nonagricultural PPP use more precisely.

Finally, the general discussion in **Chapter 6** summarises and discusses the main results obtained in this doctoral dissertation and provides some suggestions for further research.

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Abbreviation	Description
Ac	acaricide
Adobe Coldfusion	a web application and server software running on commodity hardware and
	Linux
AM	average mean
AMPA	aminomethylphosphonic acid
AP	application
aq. org.	aquatic organisms
a.s.	active substance
ASTA	Amercian Seed Trade Association
BBCH	Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie
	(BBCH- scale)
BCPC	British Crop Production Council
BE	Belgium
body	body protection
CAP	Common Agricultural Policy
Capex2	database developed in the context of the EFSA project
Cat.	category
CCD	colony collapse disorder
CF	conversion factor
CIEH	Chartered Institute of Environmental Health
CLE	Centre for Agricultural Economics
CTGB	College voor de toelating van gewasbeschermingsmiddelen en biociden
DEA	diethanolamine
DGSEI	Statistics Belgium
DISCUSS	Dual Indicator Set for Sustainable Crop protection Sustainability Surveys
DPSIR	Driving forces, Pressure, State, Impact, Response
DR	drench (soil drench)
EC	emulsion concentrate
EC	European Commission
ECPA	European Crop Protection Association
EFSA	European Food Safety Authority
EPPO	European and Mediterranean Plant Protection Organization
ERA(s)	Environmental Risk Assessment(s)
ERA field	environmental field
ES	emulsion for seed treatment
ESA	European Seed Association
ESTA	European Seed Treatment Assurance
EU	European Union
Eurostat	Directorate-General of the European Commission located in Luxembourg
FADN	Farm Accountancy Data Network
FAO	Food and Agriculture Organization of the United Nations
Fera	Food Environment Research Agency

Abbreviation	Description
FI	arable field
FIS	Seed Treatment and Environment Committee of International Seed Trade
	Federation
FPRP	Federal Pesticide Reduction Program
FPS	Federal Public Service Health, Food Chain Safety and Environment
FS	suspension concentrate for seed treatment
FTF	face-to-face survey
Fu	fungicide
Fytoweb	the official Belgian website displaying authorised PPPs
GB	granules broadcast (vehicle mounted)
GDs	Guidance Documents
Gr	growth regulator
GUS	Ground Ubiquity Score
HA	hydraulic boom with air assistance (downward)
hand	hand protection
HD	hedgerow
HDB	hydraulic boom (downward)
Не	herbicide
HM	herbaceous margin
IC	in-crop margin
ICPPR	International Commission for Plant-Pollinator Relations
ILVO	Institute for Agricultural and Fisheries Research
In	insecticide
IOBC	International Organisation for Biological and Integrated Control
IPM	Integrated Pest Management
IT	Italy
КВО	Central Enterprise Databank
KN	pressurised knapsack
LCMS-MS	Liquid chromatography-tandem mass spectrometry
LERAP	Local Environmental Risk Assessment for Pesticides
LI	liquid
LOD	limit of detection
LOQ	limit of quantification
LS	local systemic
LT	Lithuania
mask	facial and respiratory protection
max	maximum
MB	molluscicides broadcast
MI	molluscicides incorporated
min	minimum
MINA	Environmental Policy and Nature Development
MIRA	Report on the Environment and Nature
MK	motorised knapsack
ML	mixing and loading
MM	several (mixed) margin types combined
MNBA	4-methylsulfonyl-2-nitrobenzoic acid

Abbreviation	Description
MRL	Maximum Residue Level
MS	Member States
MySQL	open-source relational database running on Debian Linux
n	number of measurements/data points
NAFTA	North American Free Trade Agreement
NAK	Dutch General Inspection Service
NAPAN(s)	National Action Plan(s)
NATO	North Atlantic Treaty Organization
NL	Netherlands
NM	no margin
NR	natural regenerating margin
NS	non-systemic
NTF	NAPAN Task Force
OECD	Organisation for Economic Co-operation and Development
OF	other field
OR	orchard
OS	online survey
PA	pasture
Phytofar	Phytofar group manufacturers and formulators of PPPs
PIASs	Pesticide Impact Assessment Systems
PL	Poland
POCER	Pesticide OCcupational and Environmental Risk indicator
PPE	personal protective equipment
PPP(s)	plant protection product(s)
PPR	Panel on Plant protection products and their Residues
PSA	primary-secondary amine
QuEChERS	Quick, Easy, Cheap, Effective, Rugged and Safe
RE	re-entry
RI(s)	Risk Indicator(s); Risk Index
RO	roads and other artificial structures
RPE	respiratory protective equipment
RQ	research question
S	systemic
SC	suspension concentrate
SD	standard deviation
SDr	seed drum
SEPTWA	System for the Evaluation of Pesticide Transport to WAters
SM	sown or planted margin
ST	seed treatment
Syn	synergist
TR	track
TRR	total radioactive residues
UCL	Catholic University of Louvain
UGent	Ghent University; Laboratory of Crop Protection Chemistry
UK	United Kingdom
UK-POEM	UK-Predictive Operator Exposure Model

Abbreviation	Description
UN	unknown
UPJ	Union des entreprises pour la Protection des Jardins et Espaces verts
US	United States
US EPA	United States Environmental Protection Agency
UV-VIS	Ultraviolet-visible
VC	vertebrate control/repellent
VMM	Flemish Environment Agency
WB	wind break
wf	weighting factor
WG	water dispersible granules
WHO	World Health Organization
WO	woodland, spinneys, copses, forests, etc.
WP	wettable powder

Symbol	Description	Unit
∑Seq	sum of annual spread equivalents	-
AOEL	Acceptable Operator Exposure Level	mg/kg BW/day
BFI	Base Flow Index, fraction of water not directly linked to rainfall	0.5
BW	body weight	kg
Conc <sub>PPP</sub>	PPP concentration	g/kg
DT <sub>50</sub>	degradation time of 50% of the a.s. in the soil	years
E	annual sales of PPPs	kg a.s./year
EC <sub>50</sub>	Effect Concentration at which 50% of the test species cause a	mg/l
	desired effect (not necessarily mortality)	
f <sub>condm</sub>	fraction of applied dose carried with condensation in method m	-
f <sub>der</sub>	fraction of applied dose that is responsible for drift	-
f <sub>dir</sub>	fraction of applied dose that is responsible for direct losses	-
f <sub>dirsup</sub>	fraction of the water that can reach the surface water	-
f <sub>m</sub>	frequency of use in method m	-
f <sub>sup</sub>	fraction of land area covered by surface water	-
HRD	highest registered dose	g/m²
LC <sub>50</sub>	Lethal Concentration at which 50% of the test species cause	mg/l
	mortality in a single dose	
LD <sub>50</sub>	Lethal Dose at which 50% of the test species cause mortality in a	mg/kg or
	single dose	µg/bee
MAC	Maximum Allowable Concentration	mg/l
NOEC	No Observable Effect Concentration	mg/l
PCOW	Predicted Concentration in Outflowing Water	mg/l
PEC	Predicted Environmental Concentration	mg/l

### 1.1 Problem context: changing pesticide use, impacts and roles

Pesticide use changes over time and its impact on human health and the environment is dependent on newly introduced pesticide products, climatic conditions, new resistant crop varieties and new scientific developments, such as formulation and spraying techniques. Pesticides are considered valuable and necessary to provide sufficient quantity of quality foods and to offer humans protection from vector-borne diseases. The annual use in the last decade in Flanders (Belgium) is about 4000 tonnes of pesticides for agricultural and nonagricultural use (public services, gardens, hard surfaces, non-professional use, etc.) (MIRA, 2015). The use of pesticides can however give rise to a range of side-effects such as health effects of the operator, contamination of the water cycle, residues on agricultural products, toxicity for honey bees, birds, beneficial arthropods, etc. (Schulz et al., 2002; US EPA, 2014). Due to the non-specificity of pesticides and losses during application, a portion of the applied pesticide ends up in non-target areas and organisms (Darvas and Polgár, 1998; Huber et al., 1998; De Smet et al., 2005; US EPA, 2014). Today, however, agriculture should ensure the production of sufficient food to feed the entire population with respect for humans, animals and the environment. The responsible use of pesticides should be in conjunction with sustainable development (Tyvaert et al., 1999; De Smet et al., 2005).

Given this dynamic setting of changing pesticide use and its impacts, the role of public authorities is indispensable. Before a pesticide can be placed on the market, it has to undergo a strict authorisation procedure aimed at proving that the product is beneficial for crop production and achieves a high level of protection for humans, animals, and the environment. This procedure was first harmonised within the European Union (EU) by Directive 91/414/EEC, which has now been replaced by the more recent Regulation (EC) No 1107/2009. In Europe, highly polluting pesticides are prohibited since the 1970s (e.g. DDT). Other pesticides (e.g. lindane and parathion) are recently taken off the market under the review program of the EU due to their ecotoxicity, endocrine disrupting effects or possible bioaccumulating properties (Directive 91/414/EEC; Regulation (EC) No 1107/2009; Peeters et al., 2010). Adjustments and improvements made the current generation of pesticides less harmful than its predecessors. However, excessive use of pesticides remains strongly not recommended. Application of pesticides may give rise to the presence of residues in the food intended for consumption. Also, the operator of pesticides can be directly exposed to pesticides during treatment.

The objectives and principles of the Flemish environmental policy are stipulated in the Environmental Policy and Nature Development Plan (MINA-plan) of the Flemish Government. This MINA-plan provides the legal basis for a long-term policy on how to deal with the environment in a sustainable way. Pesticides are of special interest for the environment and the public health due to their possible side-effects. This has led to the decision by the Flemish authorities to propose a 50% reduction of the pressure exerted by pesticide use on aquatic organisms (expressed as spread equivalents) between 1990 and 2010. In order to determine the pressure exerted by the use of pesticides, an easy-to-use risk indicator was adopted in the Flemish environmental policy plan. The state of the environment is presented annually in the Report on the Environment and Nature (MIRA) according to the DPSIR-framework, a widespread analysis framework within international environmental reporting. It is within this framework that pesticide use and its related pressure on aquatic organisms are reported on an annual basis in Flanders (De Smet et al., 2005). The DPSIR (Driving forces, Pressure, State, Impact, Response) conceptual framework represents a systems analysis view (Figure 1-1), i.e. economic and social developments (D) exert pressure (P) on the environment and, as a consequence, the state (S) of the environment changes. This leads to impacts (I) on e.g. ecosystems, materials or human health that may elicit a societal response (R) that feeds back on the driving forces, on the pressures or on the state or impacts directly, through adaptation or curative action (Smeets and Weterings, 1999).



Figure 1-1 The DPSIR (Driving forces, Pressure, State, Impact, Response) framework.

Furthermore, laws on commercialisation and use of pesticides are strictly defined at both Belgian and European level. Directive 2009/128/EC (Article 4) of the European Parliament and of the Council of 21 October 2009 established a framework for Community action to achieve a sustainable use of pesticides. It imposes the Member States of the EU to introduce National Action Plans while setting quantitative objectives, measures, timelines and indicators to reduce risks of pesticide use for human health and the environment. Member States should monitor the use of pesticides containing active substances (a.s.) of particular concern and establish timetables and targets for the reduction of their use, in particular when it is an appropriate means to achieve risk reduction targets. National Action Plans should be coordinated with implementation plans under other relevant Community legislation and could be used for grouping together objectives to be achieved under other Community legislation related to pesticides. In Belgium, the National Action Plan (NAPAN) consists of the federal plan (FPRP), the Flemish regional plan, the Brussels regional plan and the Walloon regional plan. Each of these plans contains specific and joint actions that are set up in cooperation with the other authorities with the support of a NAPAN Task Force (NTF). The Flemish Action Plan is Flanders' contribution to this NAPAN. It specifies objectives and measures that are in line with the Flemish competences and Flemish policy. This plan is created with the participation of the representatives of the Flemish authorities, including amongst others, the Department of Environment, Nature and Energy, the Department of Agriculture and Fisheries and the Department of Welfare, Public Health and Family. The Flemish Environment Agency is represented here as well. In preparing its action plan other Flemish action plans are taken into account, i.e. the MINA-plan and the River basin management plans. The health, social, economic and environmental impacts of the planned actions and the views of stakeholders are considered as well (NTF, 2014).

In order to respect the requirements stipulated in Directive 2009/128/EC, the challenge is now to improve the pesticide risk index currently used in environmental policy in Flanders. This especially includes an improvement of the estimation of pesticide use in Flanders. Accurate estimates of total pesticide usage are essential to all pesticide risk indicator calculations which provide an important source of information for policy makers and help to guide decision-making as well as monitoring and evaluation. According to the European Commission (EC), pesticides include plant protection products and biocides (Directive 2009/128/EC, Article 3). Pesticides evaluated in this thesis, however, only include all substances described as plant protection products (PPPs) in European legislation (Regulation (EC) No 1107/2009). These products are applied on plants as crop protection products. The term plant protection products (PPPs) will be used throughout this thesis.

# 1.2 Problem analysis: from challenge to research objectives

The determination of the exact amount of PPP usage in Flanders is difficult because sales figures are registered at a federal level (Belgium). Hence, the use of each product in the various crops and/or application areas is not sufficiently known. Depending on local growing

conditions, cropping systems (indoor and outdoor) and recommendations of agronomists, the use of PPPs may vary significantly from area to area. Furthermore, the distribution of crops across land areas varies, even in time (Peeters et al., 2010). Based on Belgian PPP sales figures provided by the Federal Government, the use of PPPs in Flanders was calculated, taking into account the ratio of crop areas for agriculture and the population number for non-agricultural use. This method is described in De Smet and Steurbaut (2002). For all products that were made available on the market after 2002, a method was developed to divide the quantities sold across all crops based on data from the Farm Accountancy Data Network (FADN), information provided by the official Belgian website displaying authorised PPPs (Fytoweb), the percentage area of crops relative to the total crop area and per crop the ratio of the area in Flanders relative to Belgium (Peeters et al., 2010).

Up to now, the use of PPPs (kg/year) in Flanders which is divided into groups (e.g. insecticides, fungicides and herbicides), target (agriculture, horticulture, non-agriculture) or crop group, is estimated based on sales figures from the Federal Public Service Health, Food Chain Safety and Environment (FPS) (De Smet and Steurbaut, 2002). These data include the amount of active substances and not the commercial formulations, which contain all sorts of additives (including solvents, surfactants, and fillers). However, by stock processing, export and import, the actual use can be deviated from the sales figures (Peeters et al., 2010). This thesis wants to address this difference in the framework of making policy decisions.

Agricultural PPP use also includes seed treatment with PPPs. Seed treatment is defined as the process of applying fungicidal and/or insecticidal seed treatment products onto various types of seed as a protective coating to create a 'protective zone' of active substance in the soil against soil-borne pathogens and insects. Depending on the market requirements, a combination of different seed treatment products (fungicides and insecticides) is normally applied at varying application rates (Taylor et al., 2001; Nuyttens et al., 2013). Treated seed entails less emission to surface waters (Gray et al., 1996; Nault et al., 2004), but they also pose certain risks, such as contamination of the environment by the emission of abraded seed particles during sowing (Nuyttens et al., 2013). Birds may also eat treated seed, which poses a risk to their health (Vercruysse and Steurbaut, 2002). The amount of PPPs used in the treatment of seeds is not included in the current total PPP use estimates. Due to its disadvantages related to the coating of the seed and its link to the overall decline of pollinators, seed treatment has to be taken into account in the PPP risk indicator calculations.

The area of non-agricultural use is also not well researched. Unnecessary or abundant use of non-agricultural PPPs and also accidental release can put the health of humans, other organisms, plants and the environment at risk. Simple but essential safety steps can be applied in order to ensure that non-agricultural PPPs are stored and used safely. However, the actors responsible for non-agricultural PPP use (= interlocutors) are still not sufficiently known. Non-agricultural use of PPPs in Flanders so far was calculated on the basis of

population ratio Flanders/Belgium but in order to allocate the effective use of nonagricultural PPPs to the actual users, an identification of all non-agricultural PPP users in Flanders has to be made. Directive 2009/128/EC splits authorisations into two groups of PPP users, i.e. professional and non-professional users (Figure 1-2). A professional user is defined as any person who uses PPPs in the course of their professional activities, including operators, technicians, employers and self-employed people, both in farming and other sectors (i.e. agricultural and non-agricultural users). A non-professional user, on the other hand, does not meet the definition of a professional user. Non-professional users include all operators using PPPs only in private households, e.g. amateur gardeners. Only ready-to-use products – like aerosols and triggers – and products to be diluted or dissolved in water can be authorised for non-professional use. Products that should be combined with any other product, can only be authorised for professional use (Grey et al., 2006; FOD, 2009).



Figure 1-2 Users of PPPs as described in Directive 2009/128/EC (Picture references: Lievens et al., 2014).

Interpretation of DPSIR has been variable and different approaches to its definition can be found in literature. In order to avoid misinterpretations, the structure of this thesis follows the enhanced DPSIR (eDPSIR) causal network for crop protection constructed by Wustenberghs et al. (2012). Chemical PPP use forms the root node for most environmental and human health impacts. The use itself is determined by the choice of which PPP is applied, the application rate and the precautions taken during application. The volume of active substance used or frequency (defined as use indicators) are good indicators to express the driving forces (D). However, it is widely acknowledged that use indicators are not adequate proxies for assessing the risk (P) exerted by PPP use (Barnard et al., 1997; Tzilivakis

et al., 2004; Stenrod et al., 2008). Emissions of PPPs into the environment are at the very centre of the causal network. These emissions caused by single PPP applications are of interest because they in turn cause the PPP concentrations (S) in the different environmental compartments and thus pose a risk to the biota therein. Assessing the risk (P) imposed by PPP emissions typically requires estimation of the human or environmental exposure they can cause, as based on the application rate or residues. This exposure is then weighted by the concentration that is considered as having no unacceptable impact, i.e. the acute or chronic health and environmental toxicity coefficient (Barnard et al., 1997). Many such pesticide impact assessment systems (PIASs, defined as risk indicators) have been developed that take various amounts of environmental compartments or human and ecosystem effects into account (Reus et al., 2002; Labite et al., 2011). Directive 2009/128/EC also refers to using this type of indicators to evaluate progress towards sustainable PPP use. The effects caused by PPPs on human health and biota populations cause societal concerns (R) about the environment and human health. These concerns feed back to the way crop protection is carried out. This thesis especially covers the parts of D, P, S and R (Figure 1-3).

# 1.3 Thesis outline and research questions

This doctoral dissertation presents the work that has been done to improve the estimation of PPP use in Flanders in order to respect the requirements stipulated in Directive 2009/128/EC. These usage estimates are used to perform risk assessments to indicate the pressure of PPPs on human health and the environment in the framework of policy decisions. To obtain this research objective, various research questions (RQ) were formulated, investigated and discussed throughout this thesis. This **introductory chapter** has already provided some background information on PPP use and its prevailing legislation. This **Chapter 1** also reveals the lack of knowledge on PPPs used for seed treatment and non-agricultural purposes. The following chapters describe and investigate the use of PPPs in all its aspects in order to make accurate usage estimates which are essential to all PPP risk indicator calculations.

**Chapter 2** provides information about the sales and use of PPPs in Flanders. The pressure exerted by PPP use on aquatic life is described as well to answer following research questions:

- **RQ1** Do total PPP use estimates (D) based on sales figures differ from estimates based on usage registration?
- **RQ2** Does the approach based on usage registration narrow the gap between risk indicators (P) and reality (S)?

In contrast to former legislation, which focused on PPP authorisation and residues, Directive 2009/128/EC focuses on the use of PPPs at farm level. It aims to improve the awareness and behaviour of PPP users, to promote good practices, to improve low-input farming and to protect the aquatic environment. On-farm response measures or good management

practices cannot be evaluated by means of risk indicators. Furthermore, it is difficult to directly assess on-farm response measures from any data currently available. Therefore, farmers were questioned about their crop protection management practices. **Chapter 3** consists of two parts in order to answer following research questions:

**RQ3** What is currently the behaviour of farmers in Flanders related to PPP handling and application (D-R)?

Existing PPP usage surveys throughout the EU provide little information on how the products are applied by operators nor do they give details of the mitigation measures used to reduce exposure, hours worked, specific times of application or other working activities performed by the operator that may contribute to the exposure. The first part of **Chapter 3** describes the collection of PPP application data in Belgium and more precisely in Flanders performed in the context of a project funded by the European Food Safety Authority (EFSA). A wide range of factors for operators was investigated, i.e. personal protective equipment used, number of hours worked each day, etc.

RQ4 Can risk assessments (P) be improved by having extra insights in the practice (D-R)?

The second part of **Chapter 3** describes how collected information was used to refine calculations of the Pesticide OCcupational and Environmental Risk indicator (POCER). The trump of having additional response information to improve POCER as a risk indicator in the framework of sustainable PPP use is illustrated. First, a case study of fruit production was performed in the context of DISCUSS, the Dual Indicator Set for Sustainable Crop protection Sustainability Surveys. Second, information collected by means of face-to-face interviews conducted in the context of the EFSA project was used to perform a case study of potatoes in order to upgrade POCER as well.

Seed treatment is considered to be a more environment-friendly form of chemical crop protection that substitutes for other, less environment-friendly, practices. The use of treated seeds also has some disadvantages, e.g. the potential presence of systemic active substance residues in the guttation fluid, plant pollen and nectar of seed treated plants. In the last few years, this emission seemed to have resulted in bee losses in several countries. The uptake, translocation and persistence in the plant vary greatly among various products. Up to now, the amount of PPPs used in the treatment of seeds is not included in the total PPP use estimates. Agricultural PPP use only contained the use of PPPs on farms. The first part of **Chapter 4** provides background information about seed treatment with PPPs to answer following research questions:

RQ5 Seed treatments: a guide to sustainable use?

RQ5.1 What are the purpose and role of seed treatment? RQ5.2 What are the different seed treatment types and technology? RQ5.3 Which seed treatment issues do exist? Chapter 1

This information about seed treatment with PPPs has led to new research questions.

**RQ6** Does the use of seed treatment exert pressure (P) on pollinators (e.g. honey bees)? *RQ6.1 Can residue levels of PPPs be found in the plant (S) after seed treatment? RQ6.2 Can residue levels of PPPs be found in the hive (S)?* 

In order to provide an answer on these questions, a system analysis of seed treatment was conducted. Various studies already illustrated the issue of neonicotinoids and translocation of these substances through the plant. The use of neonicotinoids is limited in Belgium since 2013. Therefore, the uptake, translocation and persistence of other seed treatment products were examined for various crops, with special focus on the investigation of methiocarb residues in maize. Residue analysis of beeswax and bee bread from different hives located in Flanders was performed as well.

A lack of knowledge on PPPs used for non-agricultural purposes still exists. Nowadays, non-agricultural PPP use in Flanders only takes the population number into account. Research questions raised about this topic were investigated in **Chapter 5**.

**RQ7** Which actors are responsible for the non-agricultural use of PPPs (D) in Flanders/Belgium?

Identification and allocation of non-agricultural use of PPPs in Belgium were performed in the framework of a pilot study financed by Eurostat (Directorate-General of the European Commission located in Luxembourg). Subsequently, one of the key players in the nonagricultural PPP use (identified in the first part of this chapter) was further investigated.

RQ8 Does the use of non-professional PPPs (D) exert pressure (P)

RQ8.1 on operators? RQ8.2 on aquatic organisms? RQ8.3 on bees?

The results of the first part of this chapter also suggest to use sales figures of nonprofessional PPPs to estimate non-professional PPP use. Based on these results, an estimation of the pressure of non-professional PPP use on operators, aquatic organisms and bees in Belgium was conducted in the second part of this chapter. To provide an indication of total non-professional PPP use, non-professional sales figures could be supplemented with more detailed information about the handling behaviour of amateur gardeners.

**RQ9** What is the current PPP handling behaviour by non-professional users (D-R), e.g. amateur gardens?

**RQ10** Can the estimation of the pressure (P) exerted by non-professional PPP use be improved by having extra insights in the practice (D-R)?

The levels of knowledge, usage and awareness of amateur gardeners in Flanders were investigated by means of a questionnaire. The questionnaire was kept concise in order to

have the highest response rate possible and was conducted by means of three different survey methods, i.e. an online survey, face-to-face interviews and telephone interviews.

The last chapter – **Chapter 6** – discusses and draws conclusions about the main results of this dissertation. This chapter not only focuses on the results obtained in this dissertation, but also reflects on the challenges ahead and provides some suggestions for future research. A graphical overview of the different research chapters is presented in Figure 1-3.



Figure 1-3 Overview of different research chapters.
**Chapter 2** describes the sales and use of PPPs in Flanders in order to investigate if total PPP use estimates (D) based on sales figures differ from estimates based on usage registration (**RQ1**). The pressure of PPP use on aquatic life is presented as well to verify if the approach based on usage registration narrows the gap between risk indicators (P) and reality (S) (**RQ2**).

### This chapter has been compiled from:

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## Abstract

Indicators are used to quantify the pressure of PPPs on the environment. PPP risk indicators typically require weighting environmental exposure by a no effect concentration. An indicator based on spread equivalents ( $\Sigma$ Seq) is used in environmental policy in Flanders (Belgium). The PPP risk for aquatic life is estimated by weighting active substance usage by the ratio of their maximum allowable concentration and their soil half-life. Accurate estimates of total PPP usage in the region are essential in such calculations. Up to 2012, the environmental pressure of PPPs was estimated on sales figures provided by the Federal Government. Since 2013, PPP use is calculated based on results from the Farm Accountancy Data Network. The estimation of PPP use was supplemented with data for non-agricultural use based on sales figures of amateur use provided by industry and data obtained from public services. The Seq-indicator was modified to narrow the gap with reality. This method was applied for the period 2009-2012 and showed differences between estimated use and sales figures in order to perform risk calculations. This approach resulted in a better view on PPP use and its respective environmental pressure in Flanders.

# Introduction

The Environmental Policy and Nature Development Plan (MINA-plan) of the Flemish Government stipulates the objectives and principles of the Flemish environmental policy and provides the legal basis for a long-term policy on how to deal with the environment in a sustainable way (De Smet et al., 2005). An indicator based on spread equivalents is used in environmental policy in Flanders to quantify the pressure of PPPs on the environment. The sum of the annual spread equivalents per PPP ( $\Sigma$ Seq) expresses the pressure that is caused by the use of PPPs on aquatic life. The use of each PPP is weighted by differences in toxicity to aquatic organisms and residence time in the environment (De Smet et al., 2005). The  $\Sigma$ Seq, used since 1996, is included in the environmental policy of the Flemish Government for a regional evaluation of PPP use (De Smet and Steurbaut, 2002). In its 2003-2007 Environmental Policy Plan, the Flemish Government planned to reduce the pressure exerted by PPPs on aquatic organisms (expressed as  $\Sigma$ Seq) by 50% compared to the reference year 1990 (LNE, 2014). That goal was shifted to 2010 in MINA-plan 3+. The MINA-plan 4 (2010-2015) stipulated a further decline for that period (Peeters et al., 2010).

## 1.1 Sales and use of plant protection products

In 2010, sales figures that take into account the export of PPPs were provided for the first time by FPS. By using the corrected figures taking export exchange into account, a more accurate image of PPP usage in Belgium was obtained (Peeters et al., 2010). However, to better reflect reality, agricultural PPP use should be reconsidered. This PPP usage can be obtained by the Flemish FADN. The Farm Accountancy Data Network exists for over 50 years now and is an EU-wide instrument for evaluating the income of agricultural holdings and the impact of the Common Agricultural Policy (CAP). Each year, accountancy data are collected from a representative sample of agricultural holdings in the European Union (EC, 2013). Until approximately 2005, only monetary values were registered in the Flemish FADN by the Centre for Agricultural Economics (CLE), e.g. the costs of PPP purchase. When more knowledge of the amount of PPP usage was required, additional studies had to be performed. As these studies implied a lot of paperwork, they were only performed for a few crops per year (Van den Bossche and Van Lierde, 2002, 2003; Claeys and Van Lierde, 2005). After computerization of the FADN in 2006 (this coincided with the split of the CLE into the Social Sciences Unit, which was merged into Institute for Agricultural and Fisheries Research (ILVO), and the division for Policy Analysis, under the Department of Agriculture and Fisheries of the Flemish Government), the monetary accounts were extended with an environmental module. That is how it became possible to register not only the costs, but also the quantities of PPPs purchased and stored on a farm. Farm PPP use per year is calculated as purchase plus stock on the first of January minus stock at the end of December. The annual use of PPPs of some 700 farmers is since then monitored yearly (Peeters et al., 2010). The area of non-agricultural use is not well researched. Since 2004, public services in the Flemish region have been reducing their PPP usage with the aim of obtaining a zero-use of PPPs by 2015. Each municipality and several public services (transport services, universities, etc.) annually report their PPP use to the Flemish Government. Since 2012, information on non-agricultural use has become available due to the implementation of a separate registration of active substances for non-agricultural amateur use (KB 10/01/2010).

### 1.2 Risk indicators

An indicator is a tool that provides information on matters of wider significance than what is actually measured or makes perceptible a trend or phenomenon that is not immediately detectable. It gives an indication but not necessarily an exact measurement (Hammond et al., 1995; Niemeijer, 2002). An indicator provides an answer to several questions about the investigated system: are we moving in the correct, predetermined direction? How far are we from our goal? Therefore, a sound indicator is relevant, sensitive, repeatable, feasible, evaluable and understandable (Bockstaller et al., 1997; Schomaker, 1997; Girardin et al., 1999; Meul et al., 2008). Environmental indicators provide insight into the state and dynamics of the environment and typically include physical, biological and chemical indicators (Smeets and Weterings, 1999) and generally comprise indicators of environmental pressures, conditions and (societal) responses (OECD, 1993; Niemeijer, 2002). The relevance of an indicator for PPP use involves a clear relationship between the use of PPPs and its environmental effect. Two main types of indicators are described in literature (Reus et al., 2002; Kruijne et al., 2007; Labite et al., 2011; Wustenberghs et al., 2012).

- (1) use indicators: volume of active substance used or frequency
- (2) risk indicators: pesticide impact assessment systems (PIASs)

Pressure indicators describe developments in emissions (of chemicals, waste, radiation, noise) to air, water and soil, the use of resources and the use of land (Smeets and Weterings, 1999). It is acknowledged that use indicators are not adequate proxies for assessing pressure exerted by PPP use. The toxicity of PPPs and their degradation time vary depending on the active substances, so a useful indicator has to take these characteristics into account (Barnard et al., 1997; Levitan, 2000; Van Bol et al., 2003; Tzilivakis et al., 2004; Stenrod et al., 2008). Single risk indicators consider risk for one environmental compartment or one type of organism. Multi-risk indicators calculate the pressure on several compartments, e.g. POCER (Vercruysse and Steurbaut, 2002; Kruijne et al., 2007). Indicators provide a useful tool to highlight environmental conditions and trends for policy purposes. Environmental indicators have the ability to isolate key aspects from an otherwise overwhelming amount of information and help policy makers to see the larger patterns of what is happening and help them determining appropriate action (Niemeijer, 2002). Policy makers need simple indicators to evaluate trends (Hammond et al., 1995; Saisana and Tarantola, 2002). Flanders adopted an easy-to-use risk indicator in their environmental policy plan, i.e. the Seqindicator ( $\Sigma$ Seq) (De Smet et al., 2005).

## 1.3 Objectives

The objective of this chapter was to modify the currently used Seq-indicator in order to narrow the gap with reality, i.e. the state of the environment. Total PPP use estimates, in this case estimates based on PPP sales, were compared to estimates based on usage registration like FADN. Accurate usage estimates are essential to all PPP risk indicator calculations. In addition, this research refined and updated the Seq-indicator on three different aspects. First, the assessment of the distribution of the quantities sold in agriculture and non-agriculture was improved. Different crops in agriculture were reconsidered. This allows a better assessment of the different croppings regarding pressure on the water compartment. Second, the effect of the application method of the PPPs was included. Finally, the most recent toxicity data based on new European authorisations were processed in the calculation of the indicator.

## **Materials and Methods**

## 2.1 Description and application of the Seq-indicator

 $\Sigma$ Seq is an indicator that estimates the pressure of PPP use in both the different agricultural and horticultural crops as well as the pressure of the non-agricultural use on aquatic life. This indicator is calculated based on the following formula (De Smet et al., 2005; Peeters et al., 2010):

$$\sum Seq = \frac{E \times DT_{50}}{MAC}$$
 Equation 2-1

E = annual sales of PPPs (kg active substance/year)
DT<sub>50</sub> = degradation time of 50% of the active substance in the soil (years)
MAC = Maximum Allowable Concentration (mg/l)

∑Seq is a single risk indicator that estimates the pressure exerted on one environmental compartment, i.e. the aquatic life (algae, daphnia, fish) (Smeets and Weterings, 1999; Niemeijer and de Groot, 2008a, 2008b; Wustenberghs et al., 2012). As such, the Seq-indicator only estimates the risk to aquatic organisms and does not take into account possible bioaccumulative capacity, potential endocrine disrupting characteristics nor synergistic effects. In addition, handling of the Code of Good Agriculture Practice is not taken into account and the indicator is not suitable for local assessment since local conditions and toxicity parameters may vary from area to area. The use of more appropriate formulation methods, certain cultivation techniques and strict compliance guidelines concerning the cleaning and rinsing of PPP tanks play an important role in reducing the burden of surface waters. Yet, these elements are not reflected in this indicator. Other indicators may display specific emission scenarios (SEPTWA: Pussemier and Beernaerts, 1997) or estimate the risk for multiple components (POCER: Vercruysse and Steurbaut, 2002). Still, these indicators do

not include the annual amount of active substances used (De Smet and Steurbaut, 2002; De Smet et al., 2005).

### 2.2 Estimation of total regional/countrywide PPP use

The Federal Government (FPS) provided the Belgian sales figures of PPPs, while Belgian nonagricultural sales figures were provided by the industry (Phytofar). Sales figures were not available at regional level; furthermore, use figures in Flanders had to be combined from FADN (agricultural use estimates) and the Flemish Environment Agency (VMM, nonagricultural use). Finally, agricultural use of PPPs also includes seed treatment. Since the available data provided no information on seed treatment, seed treatment was considered a separate group under agriculture.

Up to 2012, the total use of PPPs has been seen as the sum of active substances, adjuvants and biological products. Additives and wetting agents are not effective themselves, but contribute to the active substance with a better targeting as result (De Smet and Steurbaut, 2002). For this study, the Laboratory of Crop Protection Chemistry (UGent) decided to include only chemical products based on the European list out of Annex III from the regulation of the European Union (Regulation (EU) No 656/2011). Beside the chemical products, some biopesticides registered by FADN that were not found in the European list (Regulation (EU) No 656/2011), were included in the calculation. It concerned biopesticides listed in Annex II of the EU-regulation (Commission Regulation (EC) No 889/2008) and authorised in Belgium (Fytoweb, 2012), i.e. mint oil, granulose virus, 1-dodecanol, 1-tetradecanol, potassium salts of fatty acids and paraffin oil (high & low sulf. Index). Every year the PPP list had to be updated, as some active substances are banned and new ones are added.

### 2.3 Agricultural sales and use of PPPs in different crops

Reconsidering the distribution of the quantities of PPPs sold in Belgium was twofold. Firstly, PPP use in different crop groups was based on FADN. Secondly, although the agricultural and amateur use of products was split following the new legislation (KB 10/01/2010), the non-separated sales figures still had to be used in this study, since the division was not realised for the years studied here yet. De Smet and Steurbaut (2002) classified agriculture in 13 crop groups. A 14<sup>th</sup> group was added, i.e. the pulses. Dry harvested pulses used to taken up in a separate group under agriculture in the context of the indicators of soil balance (Lenders et al., 2012). A 15<sup>th</sup> group of green manure was also added. Horticulture represents fruits, vegetables and ornamental crops in field and in greenhouses. The cultivation of potatoes (*Solanum tuberosum*), beets (*Beta vulgaris*), maize (*Zea mays*), cereals, industrial crops, fodder, meadows and pasture, pulses and green manure is referred to as agriculture.

## 2.3.1 Sales figures of PPPs collected by FPS

In Belgium, the Federal Government (FPS) annually requests the sales figures of PPPs from companies (KB 28/02/1994; KB 16/10/2007). Nowadays, the Belgian sales figures are available from 1979 to 2013. The distribution of the quantities sold in agriculture and non-agriculture were reconsidered according to the following method. This method based on sales figures multiplied the FPS sales figures with the fractions of agricultural use on farms, seed treatment and non-agriculture. These fractions were estimated based on available data from FADN, industry (Phytofar) and VMM. This is illustrated by an example for the active substance of sulphur.

- The quantities of sulphur used in an agricultural context on farms, seed treatment and non-agriculture were divided by the total estimated use. The sum of the fractions equated to 3.9% (2.7% on farms, 0% in seed treatment and 1.2% in non-agriculture).
- The fractions were rescaled to 100%: 69% of sulphur was used on farms and 31% in non-agriculture.
- These obtained fractions were multiplied by the sales figures (FPS) of sulphur. These figures were used to determine the ∑Seq that is further on in this research referred to as ∑Seq-value characterised by 'method according to sales'.

## 2.3.2 Use figures of PPPs collected by FADN

The division for Policy Analysis of the Department of Agriculture and Fisheries provided data on the use of each product per crop for the years 2007 to 2012. Data concerning active substances used in PPPs are registered by FADN. The following numerical data were calculated and delivered for this study: the number of observations, the applied amount of active substance, the area of cultivation group and a weighted average expressed in kg of active substance per hectare of a cultivation group.

The data obtained from the Department of Agriculture and Fisheries were calculated based on several assumptions. The list of active substances registered in the survey included 378 substances of which 109 substances were not listed in the European list (Regulation (EU) No 656/2011). These 109 substances were excluded from the European list but still registered in the farmers' use and therefore included in this study. Not included in this evaluation were organic farms (too few farms to be reliable), biological PPPs with an amount of zero use, products applied on animals, active substance/crop combinations that are prohibited – however, as a total herbicide is often used for the destruction of the ground cover plants before sowing of the crop, the use is assigned to the following crop according to FADN guidelines – and outliers on the total amount of active substance per hectares of maize, potato, beet and cereals. Plant protection products that are banned but still used, were part of the analysis (stock consumption). If holdings per year were insufficient to estimate a representative figure of the agricultural use of a PPP in a crop, the weighted average over the entire period (2007-2012) was included where possible. Outliers were defined as average uses larger than four times the standard deviation (Lenders et al., 2010). Areas of the different crop groups required to express the obtained data of FADN in kg active substance were provided by Statistics Belgium (DGSEI). Data from FADN were delivered on both regional (Flanders) and national (Belgium) level. The conversion factors (ratio of the areas) for various crops were based on the relationship between the growing areas per crop type in Flanders and Belgium (DGSEI, 2013). These factors were used to transform the use estimates of PPPs from Flemish level to Belgian level.

### 2.3.3 Seed treatment

The seed treatment data were calculated by using various assumptions. For various active substances, desk research was conducted to determine whether they could be applied as seed treatment formulation. The provision of data for seed treatment was based on the products authorised in 2013 in Belgium. Seed treatment is used in many crops. These crops can be divided into four major groups:

- beet;
- cereals: (winter and spring) barley (Hordeum vulgare), maize, oats (Avena sativa), rye (Secale cereale), spelt (Triticum spelta), triticale (Triticum x Secale), (winter and spring) wheat (Triticum aestivum);
- industry: chicory (Cichorium intybus var. sativum), flax (Linum usitatissimum);
- vegetables: cabbages (*Brassica oleracea*), carrots (*Daucus carota*), endive (*Cichorium endivia*), leek (*Allium porrum*), lettuce (*Lactuca sativa*), onions (*Allium cepa*), peas (*Pisum sativum*), shallots (*Allium ascallonicum*), vegetables.

The crop areas of Belgium were taken into account to quantify the amount of PPPs used in seed treatment. The amount of seed used per crop and the amount of active substance (dose/kg seed) required of that specific product to treat this seed were determined by using the seed quantity sown per hectare per crop. The amount of seed per crop was taken from Lenders et al. (2012). The amount of seed (per hectare) for wheat, maize and barley was determined by taking the average of respectively winter and spring wheat, grain and fodder maize, and winter and spring barley. In order to determine for certain products the exact amount of active substance, the parameter 'number of seeds per hectare' was needed. This number was obtained from Pannecoucque et al. (2013) or calculated from the seed quantity expressed in kilograms per hectare using the 1000 kernel weight (Alberta Agriculture and Food, 2007; Steve, 2016). Specific values used for onions, leek, lettuce, endive and cabbage were based on data which were found in various sources (Vanparijs et al., 1986; CAD-AGV, 1987; De Moel, 1993; De Kraker, 1994; Kennisakker, 2013; Starke Ayres, 2014a, 2014b). The seed quantities expressed in 'kg seed per hectare' and 'number of seeds per hectare' used for the year 2009 are shown in Table 2-1.

The obtained quantities of active substance for seed treatment were multiplied by a fraction representing the average number of seed treatments across the total crop. These confidential data were obtained by means of experts of industry (Phytofar); phytofar group manufacturers and formulators of PPPs (phytosanitary or phytopharmaceutical products). The way in which data were provided by Phytofar is illustrated by an example based on data from Dutry (2013). According to this article, 50% of all pea seeds are treated with neonicotinoids (seed treatment factor of 0.50). As concerns winter barley, even two-thirds of all seed is treated. The calculation of the amount of PPP on treated seed is illustrated here with peas. According to Fytoweb, peas are treated with cymoxanil, fludioxonil, metalaxyl-M and thiamethoxam. Only thiamethoxam is a neonicotinoid and got the seed treatment factor of 0.50 (Dutry, 2013). This factor was multiplied by the seed quantity of 10 kg/ha, the Belgian peas area of 9200 ha, the product dose of 0.15 I/100 kg seeds and active substance concentration of 350 g/l. Therefore, the amount of thiamethoxam in seed treatment of peas in Belgium was 24 kg. Following this method, the amount of seed treatment was calculated for each active substance and for all the crop groups.

Crop	Seed quantity	Number of seed	Imber of seeds 1000 kerne	
	(kg/ha)ª	(#seeds/ha	) <sup>b</sup> weight (g)	) <sup>c</sup> (ha) <sup>d</sup>
beet	5	90000	16.7-25	63206
barley	160		30-45	44810
spring barley	185			4298
winter barley	135	2500000		40512
maize	32.5	115000	180-380	238844
oats	150		30-45	4876
rye	180		30-35	459
spelt	178		47-52	9562
triticale	165		42-48	6666
wheat	200		30-40	209331
spring wheat	225			3250
winter wheat	175	3500000		206281
chicory	0.272	160000	1.1-1.7	8126
flax	120	2000000	5-6.5	11048
cabbage	0.186	35000	3.3-5.3	6061
carrots	5	1800000	0.8-2.5	3761
endive	10	7000000	1.1	84
leek	10	3700000	2.5-2.9	3383
lettuce	0.136	80000	0.8-1.7	181
onions	10	1625000	2.5-4	1658
peas	10		125-300	9200
shallots	10	1625000	3.3-3.8	6
vegetables	10			40940

Table 2-1 Seed quantities (kg/ha, #seeds/ha), 1000 kernel weight (g) and crop area (ha) of various seed treated crops in Belgium for the year 2009.

<sup>a</sup>(Lenders et al., 2012); <sup>b</sup>(Vanparijs et al., 1986; CAD-AGV, 1987; De Moel, 1993; De Kraker, 1994; Kennisakker, 2013; Pannecoucque et al., 2013; Starke Ayres, 2014a, 2014b); <sup>c</sup>(Alberta Agriculture and Food, 2007; Steve, 2016); <sup>d</sup>(DGSEI, 2013).

The conversion of the national seed treatment data to the Flemish level was done by using the ratio of the cultivation areas of each crop group in Flanders and Belgium (DGSEI, 2013). Subsequently, the conversion factors for the various crops for the period 2009-2012 were calculated under the assumption that the part of treated seeds used in Wallonia equals the part of treated seeds used in Flanders.

### 2.4 Non-agricultural use of PPPs

Non-agricultural use of PPPs in Flanders so far was calculated on the basis of population ratio Flanders/Belgium. The population database could be retrieved from the DGSEI website (De Smet and Steurbaut, 2002). In this study however, data for non-agricultural use were estimated by another way and based on two data sources. First, confidential national sales figures of amateur use were provided by industry (Phytofar). These sales figures apply to Belgium. Second, several data were recorded by the Flemish Environment agency (VMM). In the context of the project 'Zonder is gezonder' (VMM, 2004), an online inventory was put in place: each municipality has to register its PPP use since then. These data only apply to Flanders and are available for the period 2010-2013.

A conversion (CF) was performed by the amount used on the level of non-agricultural use in Flanders to the level of non-agricultural use in Belgium. Both surface area and use were charged (Table 2-2). Use by municipality in Wallonia and Brussels, the other two regions in Belgium, was determined using the following formula:

Use in Flanders + Use in Wallonia + Use in Brussels = Total use in Belgium Equation 2-2

Use in Flanders + (Use in Flanders ×  $CF_{Wallonia}$ ) + (Use in Flanders ×  $CF_{Brussels}$ ) Equation 2-3 = Total use in Belgium

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	Surface area (km²) <sup>ª</sup>	# Municipalities <sup>a</sup>	Surface area (km²/municipality)	Conversion factor (CF)	Use <sub>region</sub> (kg/year)	Use <sub>municipality</sub> (kg/year)
Flanders	13521	308	44	1.00	15143 <sup>°</sup>	49
Wallonia	16844	262	64	1.46	22108 <sup>b</sup>	84
Brussels	161.38	19	8	0.19	2929 <sup>b</sup>	154

Table 2-2 Conversion of data by public services of regional level (Flanders) to Wallonia and Brussels based on surface area (km<sup>2</sup>) and use (kg/year).

<sup>a</sup>(DGSEI, 2013; Portaal Belgium.be, 2013); <sup>b</sup>PPP use in Wallonia and Brussels was extrapolated from use in Flanders based on surface area per municipality; <sup>c</sup>Known data from VMM.

The overall use of PPPs in non-agriculture was determined by taking the sum of data obtained through Phytofar (Belgian level) and data obtained through VMM (Flemish level), and then converted to Belgian use. Since the  $\sum$ Seq was supplied in the first place for Belgium, data of VMM were – in a first step – rescaled to Belgian use based on the number of municipalities and use per municipality. In order to estimate the share of non-agriculture in Flanders, Belgian usage figures (Phytofar sales figures + VMM use figures) were multiplied by the conversion factor of 0.38. This factor was estimated by taking the ratio of non-

agricultural use in Flanders (VMM) to non-agricultural use in Belgium (data obtained by conversion to Belgium based on the number of municipalities and use by each municipality). This conversion factor had the same value for the period 2010-2012.

## 2.5 Introduction of a weighting factor

## 2.5.1 Emission pathways to surface water

PPPs can migrate to the surface water through different pathways which vary according to the properties of the active substance, the formulation, the local topography and the climatic conditions. An important distinction can be made between direct losses and diffuse losses (Carter, 2000; De Wilde et al., 2007; Peeters et al., 2010).

The term direct losses includes leaking storage tanks, spills when filling spray tanks, tank washings or pour out of surpluses. Pollution carried out locally or on a limited scale in the vicinity of the source. Diffuse losses occur after applying PPPs on a field and are spread over a large area. The identification of a diffuse source is more difficult than of a direct source. Diffuse losses include mainly runoff, volatilisation, drift and drainage (Pussemier and Beernaerts, 1997; Bedos et al., 2002; Reichenberger et al., 2007; Peeters et al., 2010).

Only direct losses and drift were taken into account in this study. Data were insufficiently available to take the remaining three pathways runoff, drainage and volatilisation into consideration since these pathways especially depend on the properties of the chemicals.

### 2.5.2 Applications and emission factors

PPPs can be sprayed or applied by means of seed treatment or a number of other application techniques. The application method of a specific PPP can be consulted via the official Belgian website displaying the authorised PPPs (Fytoweb). The website was consulted and every active substance was studied for the formulation it belongs to and whether it was possible to express the formulation (spray or seed treatment) in terms of percentage of each active substance's use.

The result of the above exercise led to the necessity of introducing a specific weighting factor related to the use of the active substances with a particular method of application. A link with the crop type seemed the most reliable method to attribute the percentage distribution of the application method between the active substances. A literature review was done to address the question how to include the application methods in the pressure calculation. Different emission factors related to various application methods were described (Adriaanse et al., 1997; Pussemier and Beernaerts, 1997; Beernaerts et al., 2005; Claeys et al., 2007; FOD, 2009). Regarding direct losses, sources Claeys et al. (2007), Beernaerts et al. (2005) and Pussemier and Beernaerts (1997) indicate that the parameters are within the same order of magnitude. As regards drift, the parameters found in Claeys et al. (2007) and Adriaanse et al. (1997) are higher than the parameters found in Beernaerts et al.

al. (2005) and Pussemier and Beernaerts (1997). The model used in Beernaerts et al. (2005) and Pussemier and Beernaerts (1997) is based on expert judgment and relies on the state of studies conducted for each particular transport pathway applied to the specific situation of Belgium, which was not the case in the other studies mentioned above. This explains the difference in parameters for drift. Pussemier and Beernaerts (1997) describe in detail the most different emission factors for drift and direct losses based on the System for the Evaluation of Pesticide Transport to Waters (SEPTWA) model. SEPTWA is empirically built based on emission factors, which assess and determine the emission of PPPs in specific circumstances. This model allowed us to assign an emission factor to various application methods. Table 2-3 illustrates the fractions of applied dose (fder, fdir, fcondm) of various application methods responsible for drift and direct losses according to the SEPTWA-model. This table also shows the required various surface fractions (f<sub>sup</sub>, f<sub>dirsup</sub>, f<sub>m</sub>) under specific circumstances to determine the emission factors. For direct losses, the value was 0.05 throughout the study. The default value for drift was 0.01, except for orchards where more drift was expected, the fraction was increased to 0.03. The 'worst-case' scenario was assumed for greenhouse, i.e. the application method with the highest fraction of applied dose carried with condensation (Low Volume Application with  $f_{condm} = 0.001$ ).

Application	Emissions	Fraction	IS <sup>a</sup>	Emission factors (%)		
spraying orchards	drift	f <sub>der</sub> = 0.03	$f_{sup} = 0.03$	0.09		
	direct losses	f <sub>dir</sub> = 0.05	$f_{dirsup} = 0.05$	0.25		
spraying fields and pastures	drift	$f_{der} = 0.01$	$f_{sup} = 0.01$	0.01		
	direct losses	f <sub>dir</sub> = 0.05	$f_{dirsup} = 0.05$	0.25		
backpack spraying	drift	$f_{der} = 0.01$	$f_{sup} = 0.01$	0.01		
	direct losses	f <sub>dir</sub> = 0.05	$f_{dirsup} = 0.05$	0.25		
seed treatment, pheromones and	drift	$f_{der} = 0$	$f_{sup} = 0.01$	0		
granules	direct losses	$f_{dir} = 0.01$	$f_{dirsup} = 0.05$	0.05		
non-agriculture	drift	$f_{der} = 0$	$f_{sup} = 0.01$	0		
	direct losses	$f_{dir} = 0$	$f_{dirsup} = 0.05$	0		
greenhouses		$f_{condm} = 0.001$	f <sub>m</sub> = 0.16	0.008		

Table 2-3 Parameters to determine the emission factors (%) of various application methods for drift and direct losses according to the SEPTWA-model (Pussemier and Beernaerts, 1997).

 ${}^{a}f_{der}$  = fraction of applied dose that is responsible for drift;  $f_{dir}$  = fraction of applied dose that is responsible for direct losses;  $f_{dirsup}$  = fraction of the water that can reach the surface water;  $f_{sup}$  = fraction of land area covered by surface water;  $f_{condm}$  = fraction of applied dose carried with condensation in method m;  $f_{m}$  = frequency of use in method m.

### 2.5.3 Determination of weighting factors

The choice of weighting factors (wf) for various application methods was based on the emission factors determined in Table 2-3. In the last step, the sum of emission factors for drift and direct losses of spraying orchards – a particular application that includes fruit (outdoor) – was assigned a weighting factor of 1 (Table 2-4). The spraying of fields and pastures comprises the crops of potato, beet, cereals, vegetables (outdoor), maize, industrial crops, ornamentals (outdoor), fodder, meadows and pasture, pulses and green manure. The

weighting factors for these application methods and backpack spraying got a value of 0.76. Applications in greenhouse cultivation, seed treatment and non-agriculture received the weighting factors 0.02, 0.15 and 0.76 respectively. According to the SEPTWA-model, the emission factor for use in non-agriculture is zero. However, drift and direct losses play a role in the application of PPPs in non-agriculture. Therefore, the application in non-agriculture was assigned the same weighting as backpack spraying. The final weighting factors were used to calculate the pressure of the use of PPPs in the respective crops and for non-agricultural purposes.

	Application	Crop group	Sum of emission factors	V	Veighting
			(% <sub>drift</sub> + % <sub>direct losses</sub> )		factor
agriculture	spraying orchards	fruit (outdoor)	0.34	$\rightarrow$	1.00
	spraying fields	beet, cereals, fodder,	0.26	$\rightarrow$	0.76
	and pastures	green manure, industrial crops			
		maize, meadows and pasture,			
		ornamentals (outdoor), potato	, pulses		
		and vegetables (outdoor)			
	greenhouses	fruit (greenhouse),	0.008	$\rightarrow$	0.02
		ornamentals (greenhouse) and	l		
		vegetables (greenhouse)			
	seed treatment		0.05	$\rightarrow$	0.15
non-agriculture	backpack spraying		0.26	$\rightarrow$	0.76

Table 2-4 Weighting factors (wf) for agriculture and non-agriculture according to application method based on the emission factors for drift and direct losses determined in Table 2-3.

### 2.6 Overview of the Seq-indicator according to new methods

Until now, the Seq-indicator was determined based on the sold quantities of PPPs (kg a.s./year) which were obtained by FPS. The Seq-indicator has in the present study been calculated in two different ways by using the above described methods. A specific weighting factor was introduced in both methods ( $_{incl wf}$ ) to link the substances with a particular method of application. First, in order to determine the  $\Sigma$ Seq, the estimated quantities of used PPPs obtained from FADN were handled. The term used in this study for this  $\Sigma$ Seq-value is 'method according to use' (1). Second, the sales figures of FPS (2.3.1) were used to determine the  $\Sigma$ Seq and supply the  $\Sigma$ Seq-value 'method according to sales' (2). In Equations 2-4 to 2-11, the abbreviations ag, seed and non-ag refer to agriculture, seed treatment and non-agriculture respectively.

### (1) method according to use

a) Seq<sub>use incl wf</sub>:

 $Total \ use_{incl \ wf}(kg) = wf_{ag} \times use_{ag}(kg) + wf_{seed} \times use_{seed}(kg) + wf_{non-ag} \times use_{non-ag}(kg)$ Equation 2-4

**Equation 2-5** 

$$Seq_{use\ incl\ wf} = Total\ use_{incl\ wf}(kg) \times \frac{DT_{50}}{MAC}$$

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PPP use

b) Seq<sub>use</sub>:

$$Total use(kg) = use_{ag}(kg) + use_{seed}(kg) + use_{non-ag}(kg)$$
 Equation 2-6

$$Seq_{use} = Total \, use(kg) \times \frac{DT_{50}}{MAC}$$
 Equation 2-7

#### (2) method according to sales

a)Seq<sub>sales incl wf</sub>:

Total sales <sub>incl wf</sub> (kg)	Equation 2-8
$= wf_{ag} \times sales_{FPS}(kg) \times use_{ag}(\%) + wf_{seed} \times sales_{FPS}(kg) \times use_{seed}(\%)$	
$+ w f_{non-ag} \times sales_{FPS}(kg) \times use_{non-ag}(\%)$	
$Seq_{sales\ incl\ wf} = Total\ sales_{incl\ wf}(kg) \times \frac{DT_{50}}{MAC}$	Equation 2-9

b)Seq<sub>sales</sub>:

$$Total \ sales \ (kg) = sales_{FPS}(kg) \times use_{ag}(\%) + sales_{FPS}(kg) \times use_{seed}(\%) + sales_{FPS}(kg)$$
Equation 2-10  
$$\times use_{non-ag}(\%)$$
Equation 2-11  
$$Seq_{sales} = Total \ sales(kg) \times \frac{DT_{50}}{MAC}$$
Equation 2-11

#### 2.7 Update of toxicity data

The residence time (persistence) of PPPs in the environment varies from several days to years. Soil half-life (DT<sub>50</sub>) is the time it takes for 50% of the compound to break down in the soil, by biological or physicochemical processes. The longer DT<sub>50</sub>, the more likely the PPP is to leach through the soil and contaminates water bodies (US EPA, 2012). The Maximum Allowable Concentration (MAC-value) is determined on the basis of six different toxicity values for the representative aquatic organisms, i.e. the acute or chronic toxicity to three trophic levels: algae, crustaceans and fish (EC<sub>50algae</sub>, NOEC<sub>algae</sub>, LC<sub>50crustacea</sub>, NOEC<sub>crustacea</sub>, LC<sub>50fish</sub> and NOEC<sub>fish</sub>). In this study, toxicity values were based on the acute EC<sub>50</sub> (Effect Concentration), concentration at which 50% of the test species cause an adverse effect (not necessarily mortality) or the acute LC<sub>50</sub> (Lethal Concentration), the concentration at which 50% of the test species cause mortality in a single dose and the chronic NOEC (No Observable Effect Concentration), the concentration at which prolonged exposure has no observable effect on the test species. Toxicity values for the same species are always chosen, as sensitivity within the same class may differ. If no data are available, the lowest available toxicity value is opted for. Available toxicity values are often incomplete and therefore a safety factor has to be taken into account. This stems from the precautionary principle - if toxicity values are missing – to absorb the differences in sensitivity to pollutants between various classes of indicator organisms. The less parameters for determining toxicity at different trophic levels are available, the higher the safety factor used (De Smet and Steurbaut, 2002; Peeters et al., 2010). These safety factors are derived from the recommendations for standards according to the European Water Framework Directive (Directive 2000/60/EC). The MAC-values are calculated through dividing the lowest toxicity value by the safety factor (De Smet and Steurbaut, 2002; De Smet et al., 2005). Table 2-5 gives an overview of the safety factors needed to derive MAC-values of the toxic properties.

Table 2-5 Safety factors for derivation of Maximum Allowable Concentration (MAC) values of the toxic properties (Vryzas et al., 2009, 2011).

Available data	Safety factor	MAC
NOEC-values of at least 3 trophic levels (algae, crustacea and fish)	10	Lowest toxicity/10
NOEC-values of at least 2 trophic levels (algae, crustacea or fish)	50	Lowest toxicity/50
NOEC-values of 1 trophic level (crustacea or fish)	100	Lowest toxicity/100
only NOEC-value of algae or just $L(E)C_{50}$ -values of aquatic species	1000	Lowest toxicity/1000

NOEC = No Observable Effect Concentration; EC = Effect Concentration; LC = Lethal Concentration.

Previous work showed that especially the MAC-values in surface waters differ (De Smet and Steurbaut, 2002; De Smet et al., 2005). Since 2010, the Laboratory of Crop Protection Chemistry (UGent) disposes of a full revised database that includes the properties of all known (authorised and unauthorised) active substances. These data are in line with the new official data from the review program for PPPs in the EU. The following sources for parameter values, ranked as function of importance, were used to create the database: authorisation files of the EU, the Footprint database (Lewis and Green, 2011) and the database of UGent supplemented with new products until 2009 including the information of the Tomlin Pesticide Manual (BCPC, 2006).

Concerning the priority substances 2,4-D, chloridazon, dimethoate, diuron, malathion, MCPA and mecoprop that pose a risk to the aquatic environment, parameter values were derived from background documents used for deriving the Environmental Quality Standards for Priority Substances (Directive 2008/105/EC). The Fraunhofer Institute was appointed by the European Commission in order to set up these documents (Table 2-6). Please note that the minimum requirements were not in line with those of the Water Framework Directive (Directive 2000/60/EC).

Table 2-6 Maximum Allowable Concentration (MAC) values (mg/l) for the priority substances according to Fraunhofer institute different from EU authorisation files and Footprint database (Lewis and Green, 2011).

Active substance	Fraunhofer institute MAC (mg/l)
2,4-D	0.0185
chloridazon	0.0100
dimethoate	0.00002
diuron	0.0002
malathion	0.000008
MCPA	0.0007
mecoprop	0.0130

### **Results and discussion**

#### 3.1 Use and sales figures of PPPs

Figures 2-1 to 2-3 show a comparison of the estimated total Belgian PPP use based on purchases by farmers and amateurs or on total sales recorded by FPS. In order to modify the Seq-indicator, the objective of this research was to relate the national sales figures to a regional level (Flanders). The sum of the usage estimates of agricultural use on farms, seed treatment and non-agriculture should be comparable to the sales data of the Federal Government (FPS). In general, Figure 2-1 shows a slight decrease in the use estimates of PPPs for the period 2009-2012 whereas the sales figures reflect a capricious pattern. For 2009, 2011 and 2012, use estimates of PPPs in all groups (except for agriculture in 2009 and horticulture in 2012) were lower than PPP sales. In 2010, the estimated use of PPPs for agriculture and horticulture was higher than sales figures whereas the estimates of nonagricultural use and seed treatment were again lower than sales figures. The trend between use and sales can be explained by Table 2-7, which summarises the active substances with the largest influence on the total use estimates and sales of PPPs. For example, in 2009 the estimated use of mancozeb was 825539 kg, while the FPS sales figures indicate that 1189363 kg mancozeb was sold in 2009. In 2010, the estimated use of mancozeb was higher than the sales figures while the use estimates in 2011 and 2012 were again lower than the sales. Mancozeb is mainly used in the cultivation of potatoes against mildew. This fungicide may be applied 10 to 30 times on the same crop. The control of mildew consumes over 80% of PPPs applied in the cultivation of potatoes. However, the sales of mancozeb decreased by 43% in 2010 compared to 2009. This decrease can be related to more favourable weather conditions in 2010. In fact, the development of mildew is strongly determined by meteorological factors such as precipitation, temperature, humidity and radiation (Lenders et al., 2013). Furthermore, one of the basic principles of Integrated Pest Management (IPM) is monitoring. This includes the use of warning systems which provide advice on the optimal spray date and the type of PPPs to be used depending on the evolution and intensity of the epidemic, and of the resistant varieties. These warning systems issued by different organisations also influence the amount of PPPs sold, e.g. the number of mildew warnings appeared to be positive correlated with the sold quantities of mancozeb (Lievens et al., 2012). The use estimates of glyphosate were lower than the FPS sales figures throughout the period. Glyphosate can be used in both agricultural and non-agricultural areas. In agriculture, glyphosate is mainly applied to control annual broadleaf weeds and grasses that compete with crops. The use of glyphosate is also affected by meteorological factors (Lievens et al., 2012; Lenders et al., 2013).

According to Pissard et al. (2005), the amounts of active substances actually used are less than the quantities sold in 80% of cases. In practice, PPPs purchased in a given year might not be used during that time of the year due to the non-presence of a particular disease or pest. Hence, it is logical that during a certain year the use is lower than sales indicate. Then

again, a proportion of the seed is also treated abroad and imported into Belgium, e.g. treated seeds of maize are mainly processed in France and Germany (Lievens et al., 2012). Please note that the import figures of PPPs on seeds are not taken into account in FPS' sales figures. These seed treatments may exert pressure on the environment in Belgium and Flanders. The difference between sales and use estimates can also be explained by economic reasons, such as budgets that need to be spent in one year or commercial actions recommending certain products which results in a stock of PPPs.

Table 2-7 PPP usage estimates (kg) obtained by Farm Accountancy Data Network (FADN), industry (Phytofar) and Flemish Environment Agency (VMM) and PPP sales figures (kg) obtained by Federal Public Service (FPS) of the active substances in Belgium with the largest influence on results for the period 2009-2012.

Active substa	ince	Use es	timates (kg	;)	FPS sales figures (kg)				
	2009	2010	2011	2012	2009	2010	2011	2012	
mancozeb	825539	807470	567374	620074	1189363	672230	966081	941778	
glyphosate	340584	334168	360808	384538	434797	371465	552861	699387	
total	5372250	5585301	4885710	5080962	6156252	5080852	6568943	6510267	



Figure 2-1 Total PPP usage estimates  $\blacklozenge$  (million kg a.s.) obtained by Farm Accountancy Data Network (FADN), industry (Phytofar) and Flemish Environment Agency (VMM) and total PPP sales figures  $\blacklozenge$  (million kg a.s.) obtained by Federal Public Service (FPS) per group (agriculture  $\blacksquare$ , horticulture  $\blacksquare$ , non-agriculture  $\blacksquare$  and seed treatment  $\blacksquare$ ) in Belgium for the period 2009-2012.

Figure 2-2 compares the estimated use of PPPs in Belgium in the major agricultural crops to the FPS sales figures. From this figure, it is clear that the total sales and use estimates largely follow the trend in the cultivation of potatoes. The cultivation of potatoes is more susceptible to various pests and diseases and differs from other agricultural crops due to its higher application rate per hectare (Table 2-8). The cultivation of potatoes may present variations in terms of PPP usage from one year to another given the variability of meteorological factors in Belgium. Besides the possible reasons mentioned in the previous section, sales and use of PPPs may also fluctuate annually depending on, for instance, the acreage of the crop (Lievens et al., 2012; Lenders et al., 2013). Figure 2-3 shows the comparison of the use estimates of PPPs in Belgium in the main horticultural crops to the national sales figures. The graph especially follows the profile of the use in the fruit crop group. Striking here is that the use of PPPs for cultivation in greenhouses and ornamentals is hardly remarkable. The estimated usage of fruit (outdoor) is much higher than sales. The question here is whether the deleted statistical relevant crops with less than six response data by FADN caused any rupture of the trend. The total figures per year remain within the same order of magnitude. However, the division put more emphasis on the fruit whereas reality might be different. Special focus on collecting data on greenhouse cultivation and ornamentals in the future can improve representativeness.



Figure 2-2 Total PPP usage estimates  $\blacklozenge$  (million kg a.s.) obtained by Farm Accountancy Data Network (FADN), industry (Phytofar) and Flemish Environment Agency (VMM) and total PPP sales figures  $\blacklozenge$  (million kg a.s.) obtained by Federal Public Service (FPS) per agricultural crop (potato  $\blacksquare$ , beet  $\blacksquare$ , cereals  $\blacksquare$ , maize  $\blacksquare$ , industrial crops  $\blacksquare$ , fodder  $\blacksquare$ , meadows and pasture  $\blacksquare$ , pulses  $\blacksquare$  and green manure  $\blacksquare$ ) in Belgium for the period 2009-2012.



Figure 2-3 Total PPP usage estimates  $\blacklozenge$  (million kg a.s.) obtained by Farm Accountancy Data Network (FADN), industry (Phytofar) and Flemish Environment Agency (VMM) and total PPP sales figures  $\blacklozenge$  (million kg a.s.) obtained by Federal Public Service (FPS) per horticultural crop (in field: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$ ; in greenhouses: fruit  $\blacksquare$ , vegetables  $\blacksquare$  and ornamental crops  $\blacksquare$  and ornamental crop

Table 2-8 shows the average amount of active substances applied per hectare for the agricultural and horticultural crops in Belgium based on usage registration data (FADN) and national sales figures (FPS) during the period 2009-2012. In general, the results from FADN are comparable to those obtained from FPS. The average application rate per hectare in the cultivation of potatoes is characterised by its high intake of active substances in relation to other agricultural crops. FPS sales figures show higher values in terms of average application rate for potatoes compared to the use estimates of FADN (except for 2010). In 2009 and 2010, the average amounts of active substances used per hectare of beets based on FADN use estimates were higher than the FPS sales figures but these were lower than the sales in the period 2011-2012. Regarding the crops cereals and maize, estimates of application rates were approximately the same for both methods. The average amounts of active substances used per hectare for industrial crops, fodder, meadows and pasture and pulses were all higher for FADN use estimates. Application of PPPs in meadows and pasture is usually performed on specific areas and is not done systematically, which means that the average application rates should be treated with caution. Both methods indicated no PPP use in green manure. Regarding horticultural crops, high amounts of active substances were applied per hectare for both methods. Horticultural crops are cultivated on a rather small acreage and thus represent high amounts used per unit area. Only the average application rates per hectare of fruit based on FADN use estimates were higher than those obtained from FPS. Especially cultivation of vegetables, fruits and ornamentals in greenhouses

requires high amounts of PPP consumption. Whereas several harvests per year can be expected for greenhouse crops, the number of harvests should be accounted for in these figures. Therefore, the figures showed in Table 2-8 present the total use per year per hectare and not necessarily the PPP consumption required for a single harvest. As concluded in the previous section, data collection on greenhouse cultivation and ornamentals in the future can improve the representativeness of PPP use on these crops.

Crop group	FADN	use estim	ates (kg a	.s./ha)	FPS sales figures (kg a.s./ha)				
	2009	2010	2011	2012	2009	2010	2011	2012	
agriculture									
potato	19.6	18.9	17.6	20.0	21.2	14.0	24.8	25.7	
beet	6.1	6.4	6.8	6.7	4.2	5.2	8.6	8.0	
cereals	3.0	2.8	2.6	2.8	2.9	2.8	2.8	2.7	
maize	1.4	1.4	1.4	1.4	1.4	1.3	1.5	1.5	
industrial crops	5.3	4.9	4.6	5.5	3.9	3.7	4.5	3.3	
fodder	0.2	0.3	0.3	0.3	0.1	0.1	0.1	0.1	
meadows and pasture	0.3	0.4	0.3	0.3	0.1	0.1	0.1	0.1	
pulses	0.4	0.4	0.4	0.7	0.2	0.2	0.2	1.0	
green manure	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
horticulture									
fruit (outdoor)	48.8	72.4	56.2	59.0	38.0	24.2	48.5	41.2	
vegetables (outdoor)	11.5	13.8	13.4	12.3	16.7	17.2	17.3	16.8	
ornamentals (outdoor)	23.5	22.3	17.8	20.8	171.1	79.3	43.1	38.1	
fruit (greenhouse)	3.4	1.6	0.9	1.6	2.7	1.3	0.6	1.2	
vegetables (greenhouse)	37.5	45.6	34.0	35.4	84.1	102.3	77.7	73.2	
ornamentals (greenhouse)	41.3	35.6	35.2	47.5	43.8	37.8	64.0	66.9	

Table 2-8 Average amounts of active substances applied per hectare (kg a.s./ha) in agricultural and horticultural crops in Belgium based on use estimates from Farm Accountancy Data Network (FADN) and national sales figures from Federal Public Service (FPS) for the period 2009-2012.

### 3.2 ∑Seq-values

Figure 2-4 presents the  $\sum$ Seq-values for Flanders. The  $\sum$ Seq based on sales figures (determined by the percentage of use) is lower than the  $\sum$ Seq based on usage figures. Figure 2-4 describes the sum of  $\sum$ Seq for PPPs used in agriculture, non-agriculture and seed treatment by using the usage figures and the sales figures (with and without weighting). In Figure 2-4, a clear difference between method 1 & 2 (use and weighted use estimates) and method 3 & 4 (sales and weighted sales data) is noted. The lower  $\sum$ Seq-values obtained by using the method based on sales, can be explained by certain active substances still in use even if they were no longer authorised, sold or imported. Table 2-9 summarises the active substances with the largest influence (sorted by year 2009) on the determination of the  $\sum$ Seq. For example in the period 2009-2011, endosulfan, fentin hydroxide and paraquat were still used in Belgium whereas the national sales figures indicate that 0 kg of these active substances was sold. This leads to a  $\sum$ Seq-value (sales) of zero and explains why the  $\sum$ Seq-value

values based on sales show much lower values since not all active substances were taken into account. The results of the  $\sum$ Seq should be treated with caution. Although the sales of some active substances were no longer allowed in the period 2009-2011 (e.g. endosulfan), they were still used in small amounts. In 2012, endosulfan was no longer used in Belgium. Toxicity parameters also affect the  $\sum$ Seq-value, even if only a small amount of a certain active substance is used.



Figure 2-4 Total  $\sum$ Seq of PPPs  $\diamond$  (billion Seq) used in agriculture  $\blacksquare$ , horticulture  $\blacksquare$ , non-agriculture  $\blacksquare$  and seed treatment  $\blacksquare$  in Flanders for the period 2009-2012 by using the method based on usage estimates and based on sales figures (with or without weighting).

Table 2-9 National estimated use (kg) based on data from Farm Accountancy Data Network (FADN), industry (Phytofar) and Flemish Environment Agency (VMM) and sales figures (kg) obtained by Federal Public Service (FPS),  $\sum$ Seq based on use estimates and sales figures of the active substances in Flanders with the largest influence on results for the period 2009-2012.

Active substance	Use estimates (kg) FPS sales figures (kg)			∑Seq <sub>use</sub> (billion Seq)				∑Seq <sub>sales</sub> (billion Seq)								
	2009	2010	2011	2012	2009	2010	2011	2012	2009	2010	2011	2012	2009	2010	2011	2012
endosulfan	167	59	59	0	0	0	0	0	17.67	5.90	5.82	0	0	0	0	0
fentin hydroxide	770	776	0	0	0	0	0	0	1.88	1.96	0	0	0	0	0	0
paraquat	1393	1278	992	1615	0	0	0	0	1.27	1.26	0.92	1.58	0	0	0	0
copper oxychloride	48189	43647	31846	23849	36076	28730	24894	0	1.07	1.03	0.69	0.44	0.83	0.61	0.52	0
copper hydroxide	20530	21555	28027	36381	46210	49020	59505	71495	0.73	0.76	1.00	1.30	1.83	1.94	2.35	2.83
total	5.37E+06	5.59E+06	4.89E+06	5.08E+06	6.16E+06	5.08E+06	6.57E+06	6.51E+06	24.74	13.03	10.56	5.34	5.50	4.96	6.32	6.49

## 3.3 Effect of weighting factors

The effect of weighting factors according to the environmental pressure of an application in method 1 & 2 and method 3 & 4 is low (Figure 2-4) but is more apparent when looking at different crops. The  $\sum$ Seq-value of the crop group agriculture, decreases between method 1 and 2 with approximately 25%. A decrease of approximately 25% is obvious, since only 76% of the estimated use is taken into account in SSeq calculations for agricultural crops when weighting is applied. Horticulture, including fruit, greenhouse cultivation and ornamentals, shows a decrease in pressure on aquatic life of about 40% due to incorporating weighting factors (the weighting factor is 0.02 for greenhouses, 0.76 for ornamentals and vegetables). Using different active substances in the  $\Sigma$ Seq every year strongly influenced the total  $\Sigma$ Seq per crop. Compared to 2009, the 5Seq for ornamentals (outdoor), for example, decreased to almost zero in 2012 (Figure 2-6). As of 2010, endosulfan was no longer used in ornamentals (outdoor), which explains the decrease of the  $\Sigma$ Seq. The effect of the weighting factors between methods 1 & 2 or 3 & 4 in Figure 2-5 follows the same trend, as the same weighting factors are used for the different crops. The effect of the weighting factors is clearly visible in Figure 2-6. This figure shows ∑Seq-values of PPPs used in horticulture. The ∑Seq-value for ornamentals (greenhouse) decreases tremendously due to incorporating a weighting factor of 0.02 assigned to the environmental pollution.



Figure 2-5 Total  $\Sigma$ Seq of PPPs  $\blacklozenge$  (billion Seq) used in agricultural crops (potato  $\blacksquare$ , beet  $\blacksquare$ , cereals  $\blacksquare$ , maize  $\blacksquare$ , industrial crops  $\blacksquare$ , fodder  $\blacksquare$ , meadows and pasture  $\blacksquare$ , pulses  $\blacksquare$  and green manure  $\blacksquare$ ) in Flanders for the period 2009-2012 by using the method based on usage estimates and based on sales figures (with or without weighting).



Figure 2-6 Total ∑Seq of PPPs ◆ (billion Seq) used in horticultural crops (in field: fruit ■, vegetables ■ and ornamental crops ■; in greenhouses: fruit ■, vegetables ■ and ornamental crops ■) in Flanders for the period 2009-2012 by using the method based on usage estimates and based on sales figures (with or without weighting).

#### 3.4 Discussion

Sales figures of PPPs recorded by FPS, are relatively simple to collect and fairly inexpensive. However, sales figures give rise to confidentiality issues and restrictions on the release and use of data for commercial reasons. The FPS sales figures contain no information about the crop, timing, regional variation in use, dose applied, number of applications to the crop or percentage of crop treated (Chapter 1). Usage estimates cover all kinds of data on the actual use of PPPs by farmers and growers, but are not always quick and easy to produce (2.3.2). On the other hand, reliable data on usage of PPPs are critical for the development of indicators of the effects of PPPs on the environment. As shown in Figures 2-1 to 2-3, the differences between sales figures and usage estimates of PPPs are not as impressive. However, the identified differences exert a large influence on the determination of the pressure on aquatic life (3.1, 3.2). It is acknowledged that use indicators (kg a.s.) are not adequate proxies for assessing pressure exerted by PPP use (De Smet and Steurbaut, 2002; De Smet et al., 2005). Furthermore, the distribution of PPP use among crops established by using usage estimates instead of sales figures of PPPs is based on an approach that is per definition more reliable. The comparative analysis of the two methods (FADN usage registration and FPS sales figures) helped to highlight the similarities and differences in terms of active substances applied per hectare for the period 2009-2012. In general, both methods showed similar results. However, differences were indicated for some specific crops and varied from one year to another (e.g. potato and beet). Figures 2-4 to 2-6 show

differences between the  $\sum$ Seq-values based on sales and based on usage estimates. The  $\sum$ Seq-values based on sales showed much lower values since not all active substances were taken into account. In addition, the toxicity parameters of the active substances also exert an influence on the results of the indicator. An active substance can be highly toxic to the environment even if only a small amount of the active substance is used (e.g. endosulfan). So accurate usage estimates and toxicity parameters of PPPs are essential to all PPP risk indicator calculations to better reflect the state of the environment. In addition, sales figures may be used to adjust and improve surveys on use of PPPs and to produce national estimates of PPP usage. The application method was included into the risk indicator calculations based on weighting factors (3.3). Taking into account the weighting factors into the calculations of the  $\sum$ Seq, provides a strong reduction of the indicator.

The adjusted method applied to estimate the total use of PPPs was used for the period 2009-2012. Still, this period is too short to see a long-term evolution. In 2009-2012, agriculture and horticulture were responsible for approximately 95% of the total use estimates. The use of PPPs in arable farming was circa 50% larger than in horticulture. Throughout the time frame, the average for non-agriculture and seed treatment was respectively 3.5% and 2%. The most commonly used PPPs were fungicides and herbicides. In fact, a limited number of PPPs determine the total  $\sum$ Seq (Table 2-9). Horticulture had over the entire period the largest influence on the  $\sum$ Seq followed by agriculture, non-agricultural use and seed treatment. In this short period, the  $\sum$ Seq-value based on use estimates of PPPs declined between 2009 and 2012. This decline is particularly caused by a reduced use of endosulfan (insecticide, prohibited in 2007). The  $\sum$ Seq is a simple indicator that requires limited data input and provides an easy tool for environmental policy planning. However, it is necessary to complement the databases with new information and research results to ensure the transparency of the applied data and to avoid misinterpretations of the policy makers.

## Conclusion

The objective of this study was to modify the currently used Seq-indicator to narrow the gap with reality. Total PPP use estimates, in this case estimates based on PPP sales, were compared to estimates based on usage registration. In general, this chapter showed the difference between use estimates and sales figures of PPPs. Use estimates were lower than national sales figures and particularly followed the trend of cultivation of potatoes and fruit. The estimated use of PPPs is more useful compared to sales figures in order to perform risk calculations. The  $\Sigma$ Seq was calculated in two different ways: based on usage estimates and based on FPS sales figures. However, the  $\Sigma$ Seq-values determined by the method based on sales figures were much lower. A certain number of PPPs can be sold in a certain year, but are not necessarily applied in that year due to the non-presence of a particular disease or pest. This is clearly shown in this chapter. A PPP like endosulfan, fentin hydroxide or paraquat, not registered in sales figures (prohibited in 2007, 2007 and 2002 respectively)

were still in use and had a remarkable pressure on surface water. The  $\sum$ Seq-values based on sales showed lower values, since not all active substances were taken into account. Another remark concerns toxicity parameters, which affect the  $\sum$ Seq-value, even if only a small amount of a certain active substance is used. Accurate usage estimates and toxicity parameters of PPPs are essential to the  $\sum$ Seq-indicator calculations to better reflect the state of the environment.

This research also refined and updated the Seq-indicator on at least three other aspects. First, the distribution of the quantities sold in agriculture and non-agriculture and the different crops in agriculture have been reviewed. The calculations were carried out based on usage figures of PPPs. Non-agricultural use and seed treatment data were also incorporated. Second, weighting factors were calculated to include the application method. Taking into account the weighting factors into the calculations of the  $\Sigma$ Seq, provides a strong reduction of the  $\Sigma$ Seq-value. Finally, the most recent toxicity data based on new European authorisations were processed in the calculation of the indicator.

Finally in the present study, non-agricultural use was obtained through data from industry related to sales figures of PPPs to amateur users. In the future, these data source from industry will be replaced by more complete data from the Federal Government. In addition, the determination of the amount of PPPs used in seed treatment was difficult during this research as well.

According to Directive 2009/128/EC, the awareness and behaviour of PPP users at farm level has to be improved. However, it is difficult to directly assess on-farm response measures from any data currently available. The first part of **Chapter 3** describes the collection of PPP application data by means of face-to-face interviews to investigate how farmers in Flanders handle and apply PPPs (D-R) (**RQ3**). The second part of **Chapter 3** deals with two case-studies for which collected information was used to refine calculations of the POCER indicator in order to verify if risk assessments (P) can be improved by having extra insights in the practice (D-R) (**RQ4**).

## This chapter has been compiled from:

Garthwaite, D., Sinclair, C.J., Glass, R., Pote, A., Trevisan, M., Sacchettini, G., Spanoghe, P., Ngoc, K.D., Fevery, D., Machera, K., Charistou, A., Nikolopoulou, D., Aarapaki, N., Tskirakis, A., Gerritsen-Ebben, R., Spaan, S., González, F.E., Stobiecki, S., Śliwiński, W., Stobiecki, T., Hakaite, P., 2015. Collection of pesticide application data in view of performing Environmental Risk Assessments for pesticides. EFSA supporting publication 2015:EN-846. 246pp.

Wustenberghs, H., Fevery, D., De Schaetzen, C., Delcour, I., D'Haene, K., Lauwers, L., Marchand, F., Taragola, N., Steurbaut, W., Spanoghe, P., 2014. Playing the trump of duality in discuss: upgrading POCER with questionnaire results. Communications in Agricultural and Applied Biological Sciences. 79(3), 525-534.

# Abstract

In response to Regulation (EC) No 1107/2009, the European Food Safety Authority (EFSA) funded a project to address cumulative exposure to PPPs and the potential combined non-target effects of multiple applications of PPPs by means of carrying out surveys in eight EU Member States, using a specifically designed survey form. Existing PPP usage surveys throughout the EU provide little information on how the products are applied by operators nor do they give details of the mitigation measures used to reduce exposure, hours worked, specific times of application or other working activities performed by the operator that may contribute to the exposure. PPP surveys in this project collected information on a wide range of factors for operators such as the number of hours worked each day for the specific principle operator, other worker activities, personal protective equipment used and details of sprayers. The risk of exposure from combined toxicity resulting from the cumulative non-dietary exposure of operators to multiple active substances used for crop protection can be determined from such data. The first part of this chapter describes the collection of PPP

application data in Belgium and more precisely in Flanders performed in the context of the EFSA project. Together with the principal operator information, the crops maize, potato and sugar beet were selected and specific information was collected on twenty fields per crop. These fields were designated 'environmental fields' and information was gathered on the multiple PPP applications to that field in 2013 together with information concerning in- and off-field margin characteristicts. These data were compared with the environmental fields of other countries cultivating similar crops. The second part of this chapter describes how collected information (by means of questionnaires) was used to refine calculations of the Pesticide OCcupational and Environmental Risk indicator (POCER). First, a case study of fruit production was performed in the context of DISCUSS. DISCUSS, the Dual Indicator Set for Sustainable Crop protection Sustainability Surveys, was designed to help farmers achieve more sustainable crop protection. The indicator set pairs risk indicators (i.e. POCER) with response indicators (i.e. a management questionnaire). Both components of DISCUSS are indicators themselves but the dual risk-response setup has the additional trump that the questionnaire reveals farm level information. Simulations with personal protection and drift mitigation measures illustrated how DISCUSS can be used to support the farmers' sustainable crop protection decision-making process. Second, information collected by means of face-to-face interviews conducted in the context of the EFSA project (first part of this chapter) was used to perform a case study of potatoes in order to upgrade POCER as well. Both case studies illustrated how responsive modifications in crop protection practices can reduce human and environmental risk.

## Introduction

The use of PPPs is an integral part of modern agriculture, and contributes to the productivity and the quality of the cultivated crop (Oerke, 2006; Verger and Boobis, 2013). Despite their ability to protect crops and to assure farmer's profit, they have an impact on the environment and human health (Millet et al., 1997; Grung et al., 2015; John and Shaike, 2015; Lerro et al., 2015). Farmers are now being called to use PPPs more consciously and to implement a more sustainable crop protection in general. Directive 2009/128/EC on the sustainable use of PPPs stipulates, among other things, a minimisation of the hazards and risks to human health and the environment from PPP use. In contrast to former legislation, which focused on PPP authorisation and residues, this directive focuses on the use of PPPs at farm level. It aims to improve the awareness and behaviour of PPP users, to promote good practices (e.g. careful handling and storage of empty PPP packaging or surpluses, regular inspection of application equipment), to improve low-input farming and to protect the aquatic environment (Wustenberghs et al., 2012). As a result, legislation – that was already quite strict – is now also targeting farm-level PPP management. On-farm response measures include more and more precautions to prevent resistance, residues or pollution and the use of non-chemical crop protection techniques. However, on-farm response measures or good management practices cannot be evaluated by means of risk indicators. Furthermore, it is difficult to directly assess on-farm response measures from any data currently available.

Existing PPP usage surveys focus on single PPP applications whereas the potential combined non-target effects of multiple PPP applications are not sufficiently known. In order to better inform farmers how to consciously deal with PPPs, the current behaviour of farmers in Flanders related to PPP handling and application has to be investigated. These insights in the current crop protection practices of Flemish farmers can then be used to improve risk assessments at farm level. These risk assessments can serve as decision support tools by helping farmers to decide on the most sustainable alternatives.

### 1.1 Collection of PPP application data in view of performing ERAs for PPPs

The EFSA Panel on Plant protection products and their Residues (PPR) was tasked in December 2008 to revise the Guidance Documents on Aquatic Ecotoxicology (EC, 2002a) and Terrestrial Ecotoxicology (EC, 2002b), resulting in the respective Mandates 2009-0001 and 2009-0002. Due to the complexity of the task, the revision will result in a series of updated Guidance Documents (GDs) covering different organism groups and spatial scales for the Environmental Risk Assessment (ERA) of PPPs. Nowadays, risk assessment for PPP authorisation is mainly based on single substance assessment. However, different PPPs can be applied sequentially or as mixtures in the environment. In the current risk assessment, it is assumed that if effects on non-target organisms occur and do not exceed a certain level, recovery from these effects will occur. This might, however, be impaired by multiple applications of PPPs, as they might have a combined action causing a lower or higher toxic effect than would be expected from knowledge of the single compound (US EPA, 2002; EFSA, 2008a, 2009; Glass et al., 2012; EC, 2014). The lack of knowledge of multiple applications of PPPs on the same crop and on crop sequence on one field in different areas of the EU made it necessary to collect more information on it. Understanding the time frames (peak effects, recovery time) associated with the multiple applications of PPPs will help develop a revised methodology assessing a realistic PPP use scenario. Therefore, a PPP use data collection is needed to investigate to which extent ecological recovery can be expected and considered in ERA. This information will provide an essential support to the revision of the GDs on Ecotoxicology. In view of the ongoing revision of the GDs on Aquatic and Terrestrial Ecotoxicology (Mandates 2009-0001 and 2009-0002), collecting data on realistic PPP use patterns in different crops or crop combinations in different areas of Europe is needed. The PPP use scenarios will then be taken into account when developing the risk assessment schemes for the aquatic and terrestrial compartment in order to give appropriate recommendations regarding the potential for ecological recovery in the revised GDs.

In response to Regulation (EC) No 1107/2009, EFSA funded a project to address cumulative exposure to PPPs and the potential combined non-target effects of multiple applications of PPPs by means of carrying out surveys in eight EU Member States (MS), using a specifically designed survey form. The eight MS represent the Northern (Lithuania), Central (Belgium, Netherlands, Poland and United Kingdom) and Southern (Greece, Italy and Spain) regulatory zones. This project is built upon knowledge and experience gained during the previous EFSA

pilot survey performed in six EU MS to collate information on cumulative non-dietary exposure (Glass et al., 2012). Whilst PPP usage surveys are performed in some countries in the EU (e.g. Lithuania, Poland and United Kingdom), these provide little information on how the products are applied by operators nor do they give details of the mitigation measures used to reduce exposure (e.g. personal protective equipment, design of the sprayer cab, qualifications), hours worked, specific times of application or other working activities performed by the operator that may contribute to the exposure (Hamey et al., 2008; Glass et al., 2012).

## **1.2 Upgrading POCER with questionnaire results**

DISCUSS, the Dual Indicator Set for Sustainable Crop protection Sustainability Surveys, was designed to help farmers achieve more sustainable crop protection (Wustenberghs et al., 2012). Indicators can serve this purpose, as they can perform multiple functions. They enable monitoring the farm's progress and become decision support tools when used to compare farmers' practices or in simulations. Indicators synthesise information and help decision makers (farmers) understand complex systems (crop protection) (Girardin et al., 1999; Trevisan et al., 2009). Moreover, indicators are assumed to have the capacity to support social learning processes, i.e. learning through group interaction with peers. Interaction with other farmers fosters learning about farming practices' sustainability and about more sustainable alternatives (Leeuwis, 2004; Röling, 2009). In such a peer encouragement setting not only knowledge can be acquired, but also changes in attitudes, norms, perceptions and behaviours can take place (Marchand et al., 2010; De Mey et al., 2011). Triste et al. (2014) emphasise that all three potential indicator functions require different implementation settings. Given all potential functionalities and their preconditions, the DISCUSS indicator set was designed to synthesise the multitude of issues raised concerning crop protection ('use less harmful PPPs', 'make the cloud disappear', 'wear gloves', 'protect the bees', etc.). The indicator set is intended for implementation in small farmer groups, coached by an advisor, where its results serve as starting point for discussion (Wustenberghs et al., 2013).

DISCUSS pairs risk indicators with response indicators (Figure 3-1). Risk for human health and the environment exerted by chemical crop protection is quantified by POCER, the Pesticide OCcupational and Environmental Risk indicator (Vercruysse and Steurbaut, 2002). Farmers' response to this risk is revealed by a questionnaire, enquiring the farmers about both their background and their management actions concerning Integrated Pest Management (IPM), human safety and environmental pollution mitigation. Both parts of DISCUSS are indicators themselves but the dual risk-response setup has the additional trump that the questionnaire reveals farm level information, by which POCER calculations can be refined. Although POCER was originally designed as a farm level indicator, over the past decade it has mainly been used for sector and regional level assessments. As such, POCER is usually calculated using the worst-case scenario and/or prevailing legislation. Pairing POCER with a questionnaire,

using it as a farm level indicator, offers an opportunity to calculate risk indicators (RIs) that concur as closely as possible with the individual farmer's actual crop protection practices. Moreover, the RIs can serve as decision support tools if the adoption of risk mitigation measures is simulated.



Figure 3-1 Dual structure and hierarchy of indicators in DISCUSS (Wustenberghs et al., 2014).

### 1.3 Objectives

First, the objective of this study was to collect detailed data on real PPP applications from farms producing crops for direct consumption (such as potatoes) and crops for processing (such as sugar beet). To meet this objective, detailed data on real PPP applications over a period of one year were collected in order to gather information on overall PPP input and application patterns on a field for different crop types in Flanders. Furthermore, information relevant for non-dietary exposure of operators over the period of one year was collected as well. This included application parameters relating to the equipment and application technique as well as the personal protective equipment (PPE) and operator behaviour and training. The aim of the current survey was to collect data in order to improve models of operator and worker cumulative exposure; it was not intended to produce national estimates of PPP usage. Second, this chapter discusses how the POCER calculation system was refined with questionnaire results. It showed how the trump of having additional response information improved POCER as a risk indicator. To start off, a case study of fruit production was performed in the context of DISCUSS. Subsequently, based on information collected in the first part of this chapter (EFSA project), a case study of potatoes was performed as well.

# **Materials and Methods**

The first part of this section presents how PPP application data were collected in the context of the EFSA project (Garthwaite et al., 2015). The second part of this section describes the multi-risk indicator POCER and the adjustments that were made to the POCER calculation system for both case studies.

## 2.1 Collection of PPP application data in view of performing ERAs for PPPs

## 2.1.1 Survey

## 2.1.1.1 Design of the farm survey

A detailed questionnaire (Appendix A: Survey questionnaire) used in the survey of the principal spray operator on each farm was developed by expanding the survey forms used for the pilot study in 2011 (Glass et al., 2012). The latter had originally taken into account the expertise from previous surveys carried out by the Food and Environment Research Agency (Fera) in the United Kingdom (UK) to survey PPP usage and working practices for operators and workers (Garthwaite, 2002, 2004a, 2004b). However, the survey form for this EFSA project differed from existing Fera survey forms and the approach was significantly different from the one used in the pilot study (Glass et al., 2012). In particular, additional data on the individual farmer or grower's approach to IPM were collected as well as field margin data and historical PPP usage on selected fields on each farm. However, the main thrust of the survey was to ascertain the extent of an individual operator's exposure during a 12-month period. This included not only spray applications on the sampled farm but on all farms sprayed by the operator. PPP usage also included non-crop areas, such as roadways, the farm yard, gravel drives, cereal stores and barren strips.

The questionnaire comprised a number of forms (Appendix A: Survey questionnaire). Form 1 (Cropping details) was used to provide background information on the farm. Farm business details including size, location, number of spray operators, use of agronomists, buffer strips and IPM were collected by means of Form 2 (Farm business details). Form 3 (PPP application) was the most critical form of the survey and included PPP application details for the principial spray operator on the farm including date, crop stage, product, method of application, application rate, area treated, start time and duration of application. On each farm one or more fields were selected as environmental fields (ERA fields). From the ERA field additional information was collected on the field margins and, where available, historical PPP usage data for the last five years were collected as well. Personal details on the principal spray operators, their behaviour relating to spraying activities and sprayer details were collected by means of Form 4 (Spray operator details) and Form 5 (Spraying equipment details) respectively. Form 6 (Principal spray operator – work activity details) was used to collect information on the work conducted by the principal spray operator on the farm.

### 2.1.1.2 Crop selection

The study had to include a representative sample of the current agronomic and PPP application practices which would be suitable for informing risk assessments for the cumulative non-dietary exposure and environment. To achieve this, major crop types were selected by means of expertise of the consortium and data from Eurostat (Directorate-General of the European Commission located in Luxembourg) to ensure that agronomic practices for the EU area were included, together with some of the possible worst-case scenarios for operator exposure. The Eurostat database was also considered as a source of land use data, together with the report of Agriculture in the EU (EC, 2010), containing information on land use for agricultural and horticultural crops. Based on the existing and published data for cropping in each of the eight MS carrying out the surveys, and to give a good range of crops in the project as a whole, nine crop types were selected. Each crop type was included in a country where that particular crop is important nationally, and a range of PPP application techniques and worker activities are covered. Similarly, for other countries the surveys were focused on particular regions rather than attempting to obtain a nationally representative survey sample. In Belgium and more precisely in the region of Flanders, the crop types potato, sugar beet (Beta vulgaris subsp. vulgaris var. altissima) and maize were selected for the survey.

### 2.1.1.3 Farm selection

Farm selection was based on the guidelines for the collection of statistics on the usage of plant protection products within agriculture and horticulture (Thomas, 2000). In Flanders, the crops potato, sugar beet and maize were sampled. These crops could have been present on a single sampled farm. With 20 samples of each crop being required and multiple crops being present on a single farm, 37 farms were sampled in total. Of those 37 farms, 31 cultivated potatoes and maize while 25 farms cultivated sugar beets. For each of these crops, sub-groups were distinguished based on the crop areas in Flanders provided by Statistics Belgium (DGSEI, 2013). Table 3-1 shows the distribution and size of the sampled farms. The UK experience showed that the behaviour of operators and workers varies significantly between smaller and larger farms (Garthwaite, 2002, 2004a, 2004b). It was therefore important to include a wide range of farm sizes in order to ensure that the survey was representative of these behaviours. The basic principle of the sampling frame was to have five different sizes of groups, each with approximately 20% of the area grown (commodity group or selected crops) within that group – not 20% of the number of farms in each size group. This normally results in much smaller numbers of farms in the upper size groups but large numbers of farms in the smaller size groups. For Flanders, only four size groups could be distinguished for the crops potato and sugar beet and three groups for maize. Creating five size groups was not possible since the stated 20% of the area grown would not have been reached.

Farm areas	Farm size classes								
	Α	В	С	D	Total				
potato									
area per farm (ha)	< 5	5-10	10-20	> 20					
area in Flanders (ha)	9884	12117	10678	13970	46659				
number of farms in Flanders	4734	1764	803	328	7629				
sugar beet									
area per farm (ha)	< 5	5-10	10-20	> 20					
area in Flanders (ha) <sup>a</sup>	6282	8404	7551	6120	28357				
number of farms in Flanders <sup>a</sup>	2071	1211	553	169	4004				
maize									
area per farm (ha)	< 10	10-20	> 20	-					
area in Flanders (ha) <sup>b</sup>	39613	42612	67035	-	149261				
number of farms in Flanders <sup>b</sup>	9381	3039	1960	-	14380				

Table 3-1 Farm size classes for potato, sugar beet and maize farms in Flanders (Belgium) based on Flemish crop areas of 2010 provided by Statistics Belgium (DGSEI, 2013).

<sup>a</sup>Sugar beet data were based on the commodity group 'Industrial crops'; <sup>b</sup>Maize data were based on the commodity group 'Cereal grains' (DGSEI, 2013).

## 2.1.1.4 Farmer contact and recruitment

In Flanders, an initial letter was sent to all members of farmers' associations who grew the necessary crops. This letter explained the purpose of the survey, the confidential nature thereof and the fact that it was on a voluntary basis and not an inspection. Key issue was finding farmers willing to cooperate. Main reasons for not willing to cooperate were (1) no time, (2) the extent of data required for the survey, and (3) crop protection is a sensitive topic (farmers may use products that are not allowed in Belgium but allowed elsewhere e.g. the Netherlands; farmers do not want any more products to be taken out of the market). All the farms were visited in March-April because this period suited the farmers' busy schedule best. The visits took on average 2.5 hours depending on the size of the farm and recording system used by the farm.

### 2.1.1.5 Data capture

Crop management record sheets are the most commonly used recording system in Belgium. These sheets are filled in on a field-by-field basis and provide the following information:

- date of application, applied product, product dose (per ha), treated area and operator (farmer or spray contractor); and
- date of sowing/planting, fertilisation activities, date of harvest.

Although crop management record sheets are most common, other recording systems are often used by the farmer:

• Application schemes provided by a crop advisor. These are usually very simple and contain information on the application date, applied product and product dose.
• Excel sheets, calendars and agendas. These are very common but the level of detail varies among farmers, depending on the purpose of records (e.g. calculation of spray application costs, farmers' interest). Some farmers will only record the date of application, applied product and applied dose, whereas others may also record spray volume, weather conditions, etc. Information on the main crop activities (sowing, planting, fertilisation and harvesting) may also be recorded.

**Form 3** information, i.e. application date, applied product and product dose, were usually available. Other information (e.g. spray volume, duration of application) was usually not recorded. **Form 6** information, such as dates of sowing/planting, fertilisation activities and harvesting, were usually available on the crop management record sheets. Other activities, such as ploughing or mechanical and thermal weeding, were not recorded. In general, records of 'other activity' data were non-existent or limited to the dates of sowing/planting, fertilisation and harvesting. All farmers were asked to keep detailed records of worker activities. Most farmers were able to do this, but the level of detail of the records seemed to be quite variable. The farmers who filled out the forms quite well were contacted and were asked whether it was possible to obtain some data of the past five years as well. The response was negative, caused by the same reasons, resulting in the lack of certain data (2.1.1.4).

## 2.1.2 Database and data entry

The database developed (Capex2) was made available to consortium members on the internet allowing the entry of data. The database used a MySQL relational database (an open-source relational database running on Debian Linux) and the web front-end using Adobe ColdFusion (a web application and server software running on commodity hardware and Linux). The data input by each consortium member, via a web portal, was regulated through a detailed user interface that ensured the use of the finite list of controlled terminology. The data checking protocols to identify any incorrect data entries were developed to identify obvious erroneous data. Although the database construction included features to minimise entry of erroneous data, significant resources were required to modify the database during the data input period in response to feedback from the consortium.

To assess collected data of the current survey with regard to Environmental Risk Assessment, data were extracted from the Capex2 database using SQL queries. Subsequently, data analysis was performed using Microsoft Access and Excel statistical programs. The approach adopted divided the analysis into two complementary parts:

### (1) Analysis of the farm general practices in the use of PPPs

The general practices on the farms in their use of PPPs were analysed considering their application of products and landscape management practices. When PPP application was investigated, whether the farms surveyed were using an agronomist to advise them on PPP

use, if they were practising IPM on the farm and the range of IPM practices they follow were taken into account. Analysis of whether the ERA fields had permanent or temporary watercourses adjacent to them and whether the farm used buffer strips/windbreaks to prevent drift was also performed (**Form 2**).

### (2) Detailed farm practices in the use of PPPs

Analysis of the specific practices in the use of PPPs using detailed data concerning the ERA fields was performed in an attempt to identify whether usage patterns could be identified based on the cultivated crop and the country. These data especially took into account the landscape management undertaken on farms. This information was compared with the ERA fields of other countries cultivating similar crops.

- Regarding the application, the average number of PPPs applied in 2013 per hectare (broken down by PPP group) and the period when the application was made (broken down by month) were taken into account to demonstrate complete scenarios of exposure pattern/PPPs that were applied over a full year (2013). Data were obtained by analysing the results from **Form 3** of the questionnaire.
- Regarding landscape management, the type of the different in/off-field margins identified were taken into consideration. For the analysis, data from Form 2 and Form 3 of the questionnaire were used.

### 2.2 Upgrading POCER with questionnaire results

In POCER, RIs for human health were calculated for the operator, re-entry worker, bystander, resident and consumer. For the environment, RIs were calculated for persistence, groundwater, aquatic organisms, birds, earthworms, bees and beneficial arthropods (Figure 3-1). The RIs were calculated as the ratio of the Predicted Environmental Concentration (PEC) to a toxicological reference value, such as the Acceptable Operator Exposure Level (AOEL) or the No Observable Effect Concentration (NOEC) for aquatic organisms (Vercruysse and Steurbaut, 2002; Garreyn et al., 2007). The toxicological reference values were taken from the authorisation files according to the European Council Regulations with regard to placing plant protection products on the market (Directive 91/414/EEC; Regulation (EC) No 1107/2009), from the Footprint database (Lewis and Green, 2011) or from other literature. The absolute RI values for each PPP application were transformed and benchmarked between a lower and an upper limit, resulting in a dimensionless value between 0 and 1 for each compartment, 1 indicating unacceptable risk and 0 implying negligible risk. To calculate farm risk for each human or environmental compartment, these values were summed over all applications in a year, resulting in 12 risk indicators. To calculate the total farm risk, the compartment-specific values were again summed into an aggregate indicator.

## 2.2.1 Case study fruit production

In the context of DISCUSS, POCER was paired with a questionnaire, enquiring about the farmer's management actions (Figure 3-1). This not only allowed assessing response to PPP risk as such, but also fed back farm level information to the risk indicator calculation. Information, such as PPE use, PPP application technology or the use of coated seeds was retrieved from the questionnaire and incorporated into the RIs concerned.

The questionnaire established for the case study of fruit production consisted of a human safety and an environmental section. The human safety section of the questionnaire comprised three questions on PPE use: (1) by the operator while mixing and loading PPPs, (2) by the operator during application, (3) by re-entry workers. For each case farmers had to indicate on a 1-5 scale never-always whether gloves, coverall or mask were worn. Each additional piece of PPE worn reduces RI<sub>operator</sub> or RI<sub>re-entry</sub>. In the environmental safety section of the questionnaire, the drift-reducing technology available on the farm (type of sprayer and nozzles) and the way it was used, was surveyed. Appropriate use of drift-reducing technology reduces the RIs for bystanders, residents and aquatic organisms. In arable or vegetable crops the use of coated seeds increases risk for birds.

Compared to earlier sector and regional level implementations, POCER risk calculation was adapted as follows. In Rl<sub>operator</sub> and Rl<sub>re-entry</sub>, instead of using the worst-case scenario (no PPE worn), reduction factors for dermal and inhalation exposure were introduced to match PPE use reported in the questionnaire. Reduction factors were taken from EFSA (2010a) and the EUROPOEM model (1996, 2002). New re-entry scenarios were added, e.g. re-entry after herbicide application in orchards. Instead of assuming PPPs were applied according to prevailing legislation, i.e. by using drift-reducing technology and buffer strips along surface water stipulated on the product labels, Rl<sub>bystander</sub>, Rl<sub>resident</sub> and Rl<sub>aquatic organisms</sub> now took questionnaire answers into account. Drift, crop interception and runoff factors were adjusted for non-spray applications. New types of PPPs were introduced, e.g. pheromones in dispensers, and bird feeding scenarios were updated. Instead of assuming that PPPs were always applied at maximum rates, application rates were retrieved from the farm's PPP application log whenever available.

## 2.2.2 Case study potatoes

Information collected by means of face-to-face interviews conducted in the context of the EFSA project was paired with POCER. Details on the location and size of the farm were extracted from **Form 1**. **Form 2** supplied information on the number of spray operators, spray decisions and the range of worker activities carried out on the farm. Detailed records of the crop, areas grown, dates, rates and methods of application of all PPPs applied to an individual field, information on the operator, the sprayer and nozzles were extracted from **Form 3**. Personal details on the principal spray operators, their behaviour relating to spraying activities and sprayer details were obtained from **Form 4** and **Form 5** respectively.

In addition to the adjustments that were made for the case study of fruit production, POCER risk calculations for the case study of potatoes also included a number of assumptions. Workers re-entering sprayed fields can be exposed to PPPs and thus incur risk while performing various hand-labour tasks, e.g. fruit thinning, harvesting, crop inspection, etc. Exposure routes during post-application activities are similar to operator exposure, although sources are different, i.e. foliage, surfaces, soil or dust may contribute. Transfer factors for dermal exposure while performing tasks in potato fields were taken from EUROPOEM (2002). Re-entry risk is calculated based on a default value of 8 h/d for the number of working hours. However, potatoes do not need any manual labour during the growing season and they are usually harvested mechanically. Professional potato growers or their staff might come into contact with sprayed potato plants while scouting for pests or diseases, or while checking a treatment's effectiveness. For such activities, a default value of 2 h/d was assumed (van Hemmen et al., 2002). A buffer strip is a strip of the parcel in the vicinity of a water surface, which must not be treated. It is measured from the last sprayed line up onto the banks of the water surface. The buffer strip is product dependent and is listed on the label. The width of the buffer strip can range from 2, 5, 10, 20 to 30 m whereas some products do not have a buffer strip. The width of the buffer strip can be reduced in size by the use of drift-reducing technologies. However, a minimum buffer strip of 1 m in the cultivation of potatoes should be respected (Vandewalle et al., 2014). Nozzle data were collected by its manufacturer name and not by its type. Nozzles were verified using the web tool for more accurate choice of spray technology and less diffuse pollution when using PPPs in order to determine which spray technology was used (Nuyttens et al., 2015). Cumulated human and environmental risk for the entire spray records of Flemish farms growing potatoes were calculated. In the context of the EFSA project, farms were classified in four size groups (2.1.1.3). POCER results were compared between farms within each farm size class.

## **Results and discussion**

The first part of this section presents the results obtained by means of face-to-face interviews conducted in the context of the EFSA project. The second part of this section presents the POCER calculations for the case studies of fruit production and potatoes.

## 3.1 Collection of PPP application data in view of performing ERAs for PPPs

## 3.1.1 Details of the farms surveyed

In Flanders, 37 farms were sampled at which a total of 30 farms were completely surveyed, including 209 fields with a maximum of 15 fields per farm. The total area of the surveyed farms was on average 62.5 (± 58.5) ha. The largest farm had a total area of 330 ha while 3.6 ha belonged to the smallest holding. Table 3-2 illustrates the number of fields per crop. During the farm selection, focus was on three crops, i.e. potato, maize and sugar beet. This explains the high number of fields for these crops surveyed in 2013 in Flanders.

Furthermore, 24 ERA fields were surveyed for potatoes, 25 and 20 for maize and sugar beets respectively.

Сгор	Crop group	Number of fields
barley	arable crops	5
beans (with pods)	vegetables	9
carrots	vegetables	5
cauliflower	vegetables	3
celeriac	vegetables	1
grassland grass	grassland and fodder	· 4
head cabbage	vegetables	2
leek	vegetables	4
maize	maize	52
oats	arable crops	1
onions	vegetables	2
other cereals	arable crops	2
other kind of root and tuber vegetables except sugar beet	vegetables	2
peas (with pods)	vegetables	5
potato	potato	46
strawberries	soft fruit	2
sugar beet	sugar beet	35
wheat	wheat	29

Table 3-2 Overview of number of fields per crop surveyed in 2013 (Flanders; based on Form 3).

#### 3.1.1.1 Details of the active substances and products applied on the farms surveyed

The treated area per holding was on average 660.3 ( $\pm$  670.5) ha. The largest area treated per holding was 3476 ha and the minimum was 128 ha. Further, the treated area per field per holding was on average 90.7 ( $\pm$  174.7) ha. The maximum and minimum area treated per field per holding were 1981 and < 0.1 ha respectively. These data include repeated applications to the crops but take into account tank mixing of a number of products within a spray round (entry into a field). Table 3-3 illustrates the area treated per field per crop group. Of all crop groups, potatoes indicated the highest treated area, i.e. 175 ha. The amount of active substance applied per holding per active substance ranged between 0 and 90 kg with an average amount of 3.8 ( $\pm$  6.9) kg. The total number of active substances applied per holding varied between 10 and 55 substances resulting in an average number of 37 ( $\pm$  13.2). The treated area per spray round per field per holding ranged between 0.03 and 50 ha with an average area of 7.4 ( $\pm$  7.9) ha. Table 3-4 gives a summary of the number of active substances were applied on wheat and potato, followed by sugar beet, barley, strawberries (*Fragaria*) and other cereals. Only five active substances were used on grass and none on oats.

Crop group	Number of fields	Area treated (ha)
arable crops	8	22.3
grassland and fodder	4	4.2
maize	52	36.6
potato	46	175
soft fruit	2	4.8
sugar beet	35	97.1
vegetables	33	68
wheat	29	93.9

Table 3-3 Overview of number of fields and area treated per field (ha) per crop group surveyed in 2013 (Flanders; based on **Form 3**).

Table 3-4 Summary of the number of active substances and products used on each crop surveyed in 2013 (Flanders; based on **Form 3**).

Сгор	Number of active substances	Number of products
barley	25	20
beans (with pods)	12	18
carrots	15	21
cauliflower	16	15
celeriac	10	11
grass	5	5
head cabbage	18	17
leek	20	23
maize	18	29
oats	-	1
onions	20	20
other cereals	21	12
other kind of root and tuber vegetables except	sugar beet 10	10
peas (with pods)	11	13
potato	44	86
strawberries	22	22
sugar beet	29	54
wheat	55	69
total	351	446

The average number of active substances applied to the ERA fields of maize, potato and sugar beet in 2013 were 6.3, 29.4 and 19.4 respectively (Table 3-5). Spray round is the term used to describe the entry into a crop or field by a sprayer. It refers to the constituents of a spray tank and can include single products or a group of products. Where multiple products are used in a single sprayer tank the spray round number is the same for all. It therefore acts as a linking number for the contents of a tank mix. When the average number of sprays is 1.0 in Table 3-5, it indicates a potential infrequent use, and in fact it could have been applied to only a single field. Only herbicides were used on the maize fields. On the potato fields, especially fungicides were applied followed by herbicides, and to a lesser extent insecticides, growth regulators and defoliants. Herbicides were the dominant group of PPPs used in sugar

beet fields. On these fields, a limited amount of fungicides, insecticides and molluscicides were applied as well (Table 3-5). A total of 17, 42 and 27 active substances were applied on the ERA fields of maize, potato and sugar beet respectively. Table 3-6 illustrates the top five active substances used on the ERA fields of maize, potato and sugar beet surveyed in 2013 in Flanders. The average application rates of active substances/formulated mixtures per crop in 2013 on the ERA fields in Flanders are described in Table A-3 and Table A-4 of Appendix A.

Crop	PPP group	Average number of	Average number of	Average number
		sprays	active substances	of products
maize	herbicide	1.4	6.3	4.1
	total	1.4	6.3	4.1
potato	defoliant	1.3	1.3	1.3
	fungicide	11.7	23.2	16.1
	growth regulator	1.0	1.3	1.3
	herbicide	2.1	5.2	4.6
	insecticide	1.4	1.4	1.4
	total	13.5	29.4	21.8
sugar beet	fungicide	1.0	1.9	1.0
	herbicide	4.8	17.9	13.5
	insecticide	1.0	1.3	1.0
	molluscicide	1.0	1.0	1.0
	total	5.5	19.4	14.6

Table 3-5 T	he average	number	of sprays (s	spray ro	unds),	active s	ubstances	and pr	oducts a	applied	to
each of the	ERA field cr	ops surve	yed in 201	3 (Flande	ers; bas	sed on <b>F</b>	orm 3).				

Table 3-6 The top five active substances (by area sprayed) used on the ERA fields surveyed in 2013 (Flanders) presented per crop (based on **Form 3**).

Сгор	PPP group	Active substance
maize	herbicide	terbuthylazine
	herbicide	nicosulfuron
	herbicide	mesotrione <sup>a</sup>
	herbicide	dimethenamid-P
	herbicide	flufenacet
potato	fungicide	cymoxanil
	fungicide	mancozeb
	fungicide	cyazofamid
	fungicide	mandipropamid
	fungicide	propamocarb
sugar beet	herbicide	phenmedipham
	herbicide	ethofumesate
	herbicide	metamitron
	herbicide	desmedipham
	herbicide	chloridazon

<sup>a</sup>Sum of mesotrione and MNBA (4-methylsulfonyl-2-nitrobenzoic acid), expressed as mesotrione.

### 3.1.1.2 Details of the sprayers used on farms

In Flanders, a total of 37 sprayers were surveyed (36 farm owned, 1 contractor owned). All of these sprayers were hydraulic boom sprayers (downward). On average, a sprayer had a main tank capacity of 2149 ( $\pm$  1258.8) I, an auxiliary tank capacity of 268 ( $\pm$  228.9) I, a boom width of 25.5 ( $\pm$  6.0) m and was 11.8 ( $\pm$  8.2) years old. The largest main tank had a volume of 4100 I whereas the smallest had a volume of only 750 I. The tank capacity of the auxiliary tanks varied between 0 and 1200 I and the boom width between 18 and 36 m. The oldest sprayer in this survey was 30 years old. Especially closed cab sprayers were used, i.e. 75.7%. The other sprayers were open cab (8.1%) or contained a carbon filter (16.2%). 46% of the sprayers contained two spray nozzle sets, only 22% contained more than two nozzle sets. Especially Flat Fan nozzles were used (Table A-8 of Appendix A). These nozzles were used for 72 months maximum and were on average replaced after 35.6 ( $\pm$  17.8) months. Detailed sprayer characteristics like average speed of use, boom height, number of farms on which the sprayer was used, number for each set of nozzles on the sprayer, etc. are described in Appendix A: Detailed sprayer characteristics.

### 3.1.2 Assessment of the collected data with regard to operator exposure

The following paragraph outlines the analysis of farm principal operator information from survey year 2013. Data were only collected on the principal spray operator on 80% of the farms surveyed; the remaining farms had information collected on two operators. Most of the additional operators (i.e. not the principal operator) were recorded because of their input on the ERA field. The average age of Flemish spray operators was 48 (± 10.2) years and the average amount of spraying experience was 24.9 (± 13.9) years. The oldest spray operator was 74 whereas the youngest operator was 23. The spraying experience varied between 1 and 60 years. All spray operators were male and were owner or tenant of the farm (except one was a contractor). 83% of all spray operators worked full time on the farm while the rest of the operators were operating on the farm as a part-time job. Only 42% of all operators had a nationally recognised spray certificate. Daily spraying hours varied from 0.2 to 17.3 hours with an average of 4 ( $\pm$  3.0) spraying hours per day. Seed treatments (ST) were excluded from most tables as the times relate more to seed drilling rates rather than spray applications. Seed drum (SDr) applications were also excluded as this is more an application in-situ rather than a field applied process. Molluscicides incorporated (MI) and Molluscicides broadcast (MB) were both excluded as well since the former is normally applied at the time of seed drilling and the latter can often be used at the same time as the sprayer and may therefore double up the number of hours spent spraying. Vertebrate control/repellent (VC) was excluded as well. Default or unknown values were recorded as 99 and were excluded. The number of applications per holding is depending on the type and number of crops and varied between 7 and 73. Furthermore, the average application time per farm ranged between 1.2 and 8.1 hours. The minimal application time per farm ranged between 0.2 and 3.3 hours, while the maximal application time per farm varied between 2.5 and 17.3 hours. Table 3-7 shows the main application methods used in the survey. This table illustrates that hydraulic boom sprayers (HDB) were the most regularly used sprayer. The range of uses of hydraulic boom sprayers is extremely variable, including applications to field grown arable and vegetable crops to herbicide strip applications made to row crops such as grapes (*Vitis vinifera*), blackcurrants (*Ribes nigrum*) and apples (*Malus domestica*). The average application duration was highest for granules and a pressurised knapsack. The hydraulic boom had an average application duration of 3.7 hours.

Table 3-7 Overview of average application duration (h) per application method (excluding application
methods ST, SDr, MB, MI, VC) surveyed in 2013 (Flanders; based on Form 3).

Application method	Code	n	Average application duration (h)
drench (soil drench)	DR	1	0.5
granules broadcast (vehicle mounted)	GB	1	8.0
hydraulic boom (downward)	HDB	764	3.7
hydraulic boom with air assistance (downward)	HA	28	3.9
pressurised knapsack	KN	3	6.3
motorised knapsack	MK	1	0.5

n = number of applications; ST = seed treatment; SDr = Seed drum; MB = molluscicides broadcast; MI = molluscicides incorporated; VC = vertebrate control/repellent.

An examination of the exposure of the principal operator to individual active substances throughout the 2013 cropping year was performed. The principal operators' daily duration of exposure per farm per active substance used varied between 0.02 and 17 hours. On average, principal operators were exposed to active substances for  $3.8 (\pm 2.8)$  hours per day. However, the number of individual active substances the principal operator could be exposed to was on average 37 in Flanders. Where two active substances were present in a formulated product, they were separated and would be cumulative to the total number of active substances present. Principal operators in Flanders spent on average 2.4 ( $\pm$  2.3) hours per spray round per field per holding. The maximum duration indicated during the survey was 17 hours per spray round per field per holding. However, it must be noted that this high value can include amalgamated fields which can represent a significant crop area and are likely to represent a full day's work. As mentioned before, spray round is the term used to describe the entry into a crop or field by a sprayer. Spray round can be used to calculate the number of sprayer entries into a field.

When considering non-dietary exposure of the operators, cleaning the sprayer, and mixing and loading the sprayer are two operations/activities that bring the operator closer to PPPs than would be experienced when in a spray cab, particularly one that is air conditioned and filtered. The average time spent cleaning the sprayer was approximately 0.7 ( $\pm$  0.4) hours. The maximum time used to clean the sprayer was 2 hours. The number of times the sprayer was cleaned per year varied between 1 and 180 times. On average, the sprayer was cleaned 17.6 ( $\pm$  29.7) times per year. For mixing and loading the sprayer, an average of 0.2 ( $\pm$  0.1) hours was spent performing this activity with a range of between 0.02 and 0.33 hours.

Mixing and loading events ranged between 1 and 10 times per day. On average, mixing and loading took place 3.3 ( $\pm$  1.6) times a day.

During the survey, detailed information on the usage of PPE when performing PPP application, mixing and loading, sprayer cleaning and other work activities was collected. A summary of these data are provided in Table 3-8. Breathable workwear in combination with leather/fabric or rubber boots were especially used as PPE while using a hydraulic boom. Other PPE were hardly used by the principal spray operators. The use of gloves (vinyl, neoprene, nitrile), coveralls and face shields were similar when operators were mixing and filling solids and liquids, but the use of respiratory protective equipment (RPE) was higher when mixing and filling solids. During the cleaning of the sprayer, breathable workwear in combination with leather/fabric or rubber boots and to a lesser extent gloves (vinyl, nitrile, neoprene) were used as PPE. While performing other work activities on the field, operators especially used breathable workwear in combination with leather boots. Depending on the weather, they often do not wear PPE but only shorts and t-shirt. Detailed operator characteristics like percentage of all spraying conducted by the operator on the sampled farm, type of certification, year of most recent training, type of PPE worn during operator activities, average daily/weekly hours worked during each month are described in Appendix A: Detailed operator characteristics.

Method of application	Type of PPE	Number of principal operators wearing PPE		
hydraulic boom (downward)	coat-padded	1		
	full length trousers	1		
	gloves-fabric/leather	1		
	gloves-latex	2		
	gloves-neoprene	1		
	gloves-nitrile	1		
	gloves-vinyl	3		
	leather/fabric boots	21		
	long sleeved shirt	1		
	respirator-disposable f	iltering half mask 2		
	respirator-full face mas	sk 1		
	respirator-half mask, re	eusable with filters 3		
	respirator-valved filter	ing half mask 1		
	rubber boots	15		
	work wear: breathable	(cotton/polyester) 31		
	work wear: rainwear 1	piece (vinyl, etc.) 2		
	work wear: rainwear 2	piece (vinyl, etc.) 1		

Table 3-8 The number of principal spray operators that wore specific personal protective equipment (PPE) while using a hydraulic boom sprayer surveyed in 2013 (Flanders; based on **Form 4**).

### 3.1.3 Assessment of the collected data with regard to Environmental Risk Assessment

#### 3.1.3.1 Analysis of the farm general practices in the use of PPPs

The use of an agronomist (or professional advisor) to advise on PPP use and IPM practices were commonly implemented by almost every farm in Flanders where the surveys were performed. In Table 3-9, information is presented about the range of IPM practices farmers tend to follow. Crop rotation is widely used in Flanders. Also very common is the use of predictive models/early warning systems and the selection of resistant varieties. Furthermore, it is interesting to see that the use of biological control agents is not common. However, only few biological control agents are available for use in arable farming. Water courses were commonly present among the farms surveyed, i.e. 78% in case of permanent water courses and 81% in case of temporary water courses. Measures to prevent drift were not common: buffer strips (41%), windbreaks (19%) and in-crop buffer strips (0%).

Table	3-9	Percentage	(%)	of	farms	implementing	Integrated	Pest	Management	(IPM)	practices
survey	/ed ii	n 2013 (Fland	ders;	bas	sed on	Form 2).					

IPM practices	IPM practices applied (%)
crop rotation	95
maintaining and increasing populations of beneficial parasites and predate	ors 43
selection of PPPs to minimise risk to beneficial parasites and predators	49
selection of resistant varieties	59
use of biological control agents	11
use of monitoring traps	19
use of predictive models/early warning systems	84
others	16

### **3.1.3.2** Detailed farm practices in the use of PPPs

Analysis of the specific practices in the use of PPPs using detailed data concerning the ERA fields of maize, potato and sugar beet was performed in an attempt to identify whether usage patterns could be identified based on the crop cultivated and the country. These data especially took into account the landscape management undertaken on farms. ERA fields of maize, potato and sugar beet were also surveyed in 2013 in Italy, Poland, Lithuania, the Netherlands and United Kingdom. These data were used to compare the use of PPPs on ERA fields in Flanders with other countries cultivating similar crops.

Data considering the number of applications could be presented based on products, formulated mixtures and/or active substances. The following data were analysed and presented on a product basis. The use of an active substance would artificially increase the number of applications when products containing more than one active substance were considered. Formulated mixtures would artificially reduce the number of applications when products containing the same active substance(s) were considered. Presentation of the data on a product basis was considered a compromise for the data analysis. Off-field

characteristics are those surrounding a field: either (semi-)natural habitats with high ecological value (such as hedgerow or woodland) or simple structures (fence or bare strip of land). Normally, no short-term changes in cultivation are made and in most cases they are not influenced by the farmer. Other off-field categories comprise man-made structures, e.g. an adjacent field, roads and ditches. In-field structures are characteristically a piece of cropped land, typically managed by one farmer. In-crop margins are normally areas that remain unsprayed, either by all active substances or by selected active substances, such as chlorpyrifos applied to apples in the United Kingdom. Here the outer edge of the apple orchard remains untreated with chlorpyrifos in order to prevent drift into neighbouring habitats, including watercourses. Buffer strips are defined as an in-field, cropped or uncropped zone of a defined width at the edge of a field which is influenced by the farmer's action (e.g. spray drift). The buffer strip can be enforced by authorities, for example if it is part of an agri-environmental scheme and has prescribed management actions in order to meet the off-field specific protection goal (for example spray drift into watercourses). In addition, buffer strips may provide a recovery potential for the cropped areas. The use of incrop margins was rarely exercised for all PPP applications to a field and in most cases applied to specific active substances or spray rounds. Details of the off- and in-field margins per crop on the ERA fields surveyed in 2013 are described in Table A-5 of Appendix A.

### <u>(1) Maize</u>

Table 3-10 shows the average number of products applied in 2013 broken down (by percentages) into the different PPP groups. On average, four products per hectare were applied in 2013 on ERA maize fields. Poland appeared to have a lower number of products applied than Belgium and Italy. Herbicides were frequently used, i.e. on 85% of the surveyed farms. On the other hand, percentages among PPP groups seemed very similar to the exception of insecticides which were more frequently used in Italy (24%) than in Belgium (0%) and Poland (3%).

Maize		n	Perc	entage of	Total average number			
			Fu	Gr	He	In	Other	per hectare
total		66	0	0	85	11	4	4
country	BE	25	0	0	99	0	1	4
	IT	21	0	0	67	24	9	5
	PL	20	0	0	97	3	0	1

Table 3-10 Average number of products applied in 2013 per hectare (broken down by PPP group) for ERA fields of maize (based on **Form 3**).

n = number of ERA fields; Fu = fungicide; Gr = growth regulator; He = herbicide; In = insecticide; BE = Belgium and more precisely Flanders; IT = Italy; PL = Poland.

Table 3-11 shows when products were applied during the year. While in Italy a higher application of products was registered in April, May and June (24%, 26% and 36%, respectively), in Belgium and Poland the number of applications was higher in May and June (13% and 87% for Belgium, 49% and 24% for Poland, respectively).

Maize		n	F	Percentage of products applied in each month (%)											%)	Total average number
			J	F	Μ	Α	Μ	J	J	Α	S	0	Ν	D	UN	per hectare
total		66	0	0	0	11	23	57	5	2	0	0	0	0	2	4
country	BE	25	0	0	0	0	13	87	0	0	0	0	0	0	0	4
	IT	21	0	0	0	24	26	36	11	3	0	0	0	0	0	5
	PL	20	0	0	0	7	49	24	0	3	0	0	0	0	17	1

Table 3-11 Average number of products applied in 2013 per hectare (broken down by month) for ERA fields of maize (based on **Form 3**).

n = number of ERA fields; UN = unknown; BE = Belgium and more precisely Flanders; IT = Italy; PL = Poland.

In relation to the landscape management, Table 3-12 shows a similar situation depending on the country in relation to the off-field margins. In every country the most part of the off-field margins was on average comprised of other fields (42%) and roads and other artificial structures (21%). On the other hand, the situation about in-field margins was very different (Table 3-13): while in Belgium and Poland in-field margins seldom had margins (98% in Belgium and 78% in Poland), in Italy having herbaceous margins (23%) and mixed margin types combined (19%) was possible.

Table 3-12 Average percentage (%) of off-field margins for ERA fields of maize surveyed in 2013 (based on **Form 3**).

Maize		n	Types of off-field margins (%)									
		-	FI	HD	OF	OR	PA	RO	TR	WB	wo	Other
total		66	0	1	42	0	5	21	1	0	8	22
country	BE	25	0	0	40	1	12	24	0	0	9	14
	IT	21	0	4	32	0	2	25	0	1	1	35
	ΡL	20	1	0	54	0	0	14	4	0	13	14

n = number of ERA fields; FI = arable field; HD = hedgerow; OF = other field; OR = orchard; PA = pasture; RO = roads and other artificial structures; TR = track, drove, etc.; WB = wind break; WO = woodland, spinneys, copses, forests, etc.; BE = Belgium and more precisely Flanders; IT = Italy; PL = Poland.

Maize	-	n	n Types of in-field margins (%)									
		-	HM	IC	MM	NM	NR	SM	Other			
total		66	8	2	6	76	8	0	0			
country	BE	25	2	0	0	98	0	0	0			
	IT	21	23	6	19	46	6	0	0			

Table 3-13 Average percentage (%) of in-field margins for ERA fields of maize surveyed in 2013 (based on **Form 3**).

n = number of ERA fields; HM = herbaceous margin; IC = in-crop margin; MM = several (mixed) margin types combined; NM = no margin; NR = natural regenerating margin; SM = sown or planted margin; BE = Belgium and more precisely Flanders; IT = Italy; PL = Poland.

0

78

22

0

0

PL

20

0

0

## <u>(2) Potato</u>

Table 3-14 shows the average number of products applied in 2013 broken down (by percentages) into the different PPP groups. In Lithuania, a lower number of products were applied in 2013 (4 against 22 for Belgium and 26 for the Netherlands). Furthermore, percentages among PPP groups appeared different: while in Belgium and the Netherlands most of the products applied were fungicides (74% and 64%, respectively), product usage in Lithuania was more distributed among fungicides, herbicides and insecticides (40%, 23% and 34%, respectively).

Table 3-15 shows when products were applied during the year. Belgium and the Netherlands had a similar distribution throughout the year, whereas Lithuania showed a peak in application during the month of June (44% against the 16% of Belgium and the 21% of the Netherlands).

Table 3-14 Average number of products applied in 2013 per hectare (broken down by PPP group) for ERA fields of potato (based on **Form 3**).

Potato		n	Perc	entage of	)	Total average number		
			Fu	Gr	He	In	Other	per hectare
total		58	66	1	22	9	2	15
country	BE	24	74	1	21	3	1	22
	LT	24	40	0	23	34	3	4
	NL	10	64	1	22	11	2	26

n = number of ERA fields; Fu = fungicide; Gr = growth regulator; He = herbicide; In = insecticide; BE = Belgium and more precisely Flanders; LT = Lithuania; NL = Netherlands.

Table 3-15 Average number of products applied in 2013 per hectare (broken down by month) for I	ERA
fields of potato (based on <b>Form 3</b> ).	

Potato		n		Percentage of products applied in each month (%)											%)	Total average number
			J	F	Μ	Α	Μ	J	J	Α	S	0	Ν	D	UN	per hectare
total		58	0	0	0	3	13	21	28	25	10	0	0	0	0	15
country	BE	24	0	0	0	3	15	16	29	26	11	0	0	0	0	22
	LT	24	0	0	0	1	12	44	25	12	6	0	0	0	0	4
	NL	10	0	0	0	4	11	21	26	26	11	0	1	0	0	26

n = number of ERA fields; UN = unknown; BE = Belgium and more precisely Flanders; LT = Lithuania; NL = Netherlands.

In relation to the landscape management, Table 3-16 and Table 3-17 show the situation per country. On the one hand, in every country, the largest part of the off-field margins was on average comprised of other fields (37%), roads and other artificial structures (11%), and pastures (11%). On the other hand, while in Belgium and the Netherlands in-field margins seldom had margins (100% in Belgium and 70% in the Netherlands), in Lithuania having other in-field margin types was possible (Table 3-17).

(100000		,.										
Potato		n				Types o	of off-fie	eld marg	ins (%)			
			FI	HD	OF	OR	ΡΑ	RO	TR	WB	WO	Other
total		58	2	0	37	0	11	11	4	0	5	30
country	BE	24	0	0	34	0	12	20	0	0	6	28
	LT	24	5	0	45	0	15	1	9	0	5	20
	NL	10	0	0	24	0	3	12	0	0	0	61

Table 3-16 Average percentage (%) of off-field margins for ERA fields of potato surveyed in 2013 (based on **Form 3**).

n = number of ERA fields; FI = arable field; HD = hedgerow; OF = other field; OR = orchard; PA = pasture; RO = roads and other artificial structures; TR = track, drove, etc.; WB = wind break; WO = woodland, spinneys, copses, forests, etc.; BE = Belgium and more precisely Flanders; LT = Lithuania; NL = Netherlands.

Table 3-17 Average percentage (%) of in-field margins for ERA fields of potato surveyed in 2013 (based on **Form 3**).

Potato		n	n Types of in-field margins (%)									
		-	НМ	IC	MM	NM	NR	SM	Other			
total		58	3	16	2	75	3	1	0			
country	BE	24	0	0	0	100	0	0	0			
	LT	24	8	25	4	55	6	1	1			
	NL	10	0	30	0	70	0	0	0			

n = number of ERA fields; HM = herbaceous margin; IC = in-crop margin; MM = several (mixed) margin types combined; NM = no margin; NR = natural regenerating margin; SM = sown or planted margin; BE = Belgium and more precisely Flanders; LT = Lithuania; NL = Netherlands.

### (3) Sugar beet

Table 3-18 shows the average number of products applied in 2013 broken down by the different PPP groups. This table illustrates that a similar number of chemical product was applied in 2013 (15 in Belgium and 12 in United Kingdom). Furthermore, the percentages among the PPP groups were similar although in United Kingdom use of fungicides and insecticides (11% and 9%, respectively) was higher than in Belgium (4% and 1%, respectively).

Table 3-18 Average number of products applied in 2013 per hectare (broken down by PPP group) for ERA fields of sugar beet (based on **Form 3**).

Sugar bee	t	n	Perc	entage of	Total average			
			Fu	Gr	Не	In	Other	number per hectare
total		40	7	0	84	4	5	14
country	BE	20	4	0	89	1	6	15
	UK	20	11	0	77	9	3	12

n = number of ERA fields; Fu = fungicide; Gr = growth regulator; He = herbicide; In = insecticide; BE = Belgium and more precisely Flanders; UK = United Kingdom.

Table 3-19 shows when products were applied during the year. The distribution of applications throughout the year for both countries appeared similar although in Belgium a higher percentage of PPPs applied was registered in May (55% versus 41% of the UK).

Table 3-19 Average number of products applied in 2013 per hectare (broken down by month) for ERA fields of sugar beet (based on **Form 3**).

Sugar be	et	n	Ρ	Percentage of products applied in each month (%)											(%)	Total average number
			J	F	Μ	Α	Μ	J	J	Α	S	0	Ν	D	UN	per hectare
total		40	0	0	1	17	49	24	2	4	1	1	0	0	1	14
country	BE	20	0	0	0	15	55	24	2	4	0	0	0	0	0	15
	UK	20	0	0	2	20	41	25	2	5	1	1	1	0	2	12

n = number of ERA fields; UN = unknown; BE = Belgium and more precisely Flanders; UK = United Kingdom.

In relation to the landscape management, Table 3-20 and Table 3-21 show the situation per country. In Belgium, the largest part of the off-field margins comprised of other fields (41%) and roads and other artificial structures (22%) and the in-field margins were comprised of no margins (100%). The largest part of the off-field margins in the United Kingdom was comprised of hedgerows (17%) and roads and other artificial structures (13%). The in-field margins were comprised of natural regenerating margins (54%), no margins (20%) and sown or planted margins (19%).

Table 3-20 Average percentage (%) of off-field margins for ERA fields of sugar beet surveyed in 2013 (based on **Form 3**).

Sugar beet		n	Types of off-field margins (%)										
			FI	HD	OF	OR	ΡΑ	RO	TR	WB	WO	Other	
total		40	3	9	22	0	3	18	2	0	3	40	
	BE	20	2	0	41	0	7	22	0	0	3	25	
	UK	20	5	17	4	0	0	13	4	0	2	55	

n = number of ERA fields; FI = arable field; HD = hedgerow; OF = other field; OR = orchard; PA = pasture; RO = roads and other artificial structures; TR = track, drove, etc.; WB = wind break; WO = woodland, spinneys, copses, forests, etc.; BE = Belgium and more precisely Flanders; UK = United Kingdom.

Table 3-21 Average	percentage (%)	of in-field	margins for	ERA fields	of sugar	beet surveyed	l in 2013
(based on Form 3).							

Sugar beet		n	Types of in-field margins (%)						
			НМ	IC	MM	NM	NR	SM	Other
total		40	3	0	0	60	27	9	1
country	BE	20	0	0	0	100	0	0	0
	UK	20	5	0	0	20	54	19	2

n = number of ERA fields; HM = herbaceous margin; IC = in-crop margin; MM = several (mixed) margin types combined; NM = no margin; NR = natural regenerating margin; SM = sown or planted margin; BE = Belgium and more precisely Flanders; UK = United Kingdom.

#### 3.1.4 Discussion

Detailed data on real PPP applications over a period of one year were collected in 2013 in order to gather information on overall PPP input and application patterns on a field for different crop types in Flanders. The surveys focused on particular regions rather than attempting to obtain a nationally representative survey sample. During the selection of the farms, emphasis was put on three crops, i.e. maize, potato and sugar beet. These crops were important in Flanders, and they covered a range of PPP application techniques and worker activities. Furthermore, information relevant for non-dietary exposure of operators over a period of one year was collected as well. This included application parameters relating to the equipment and application technology as well as the PPE and operator behaviour and training. All this information was collected by means of conducting face-to-face interviews. Face-to-face interviews are appropriate to come into contact with the operator and are also quite accurate. The surveyor can give an explanation to the questions in order to achieve a more honest and clear answer. On the other hand, the words or actions of the surveyor can intentionally influence respondents to answer in a particular way. Face-to-face interviews, however, are time-consuming and also cover a smaller geographic area than other data collection methods (Eurostat, 2008). A particularly important issue of this study which contrasted strongly with the pilot study (Glass et al., 2012) was the additional workload put onto the field surveyors who had to collect data, not only from the sampled farm, but also any farm that was sprayed by the principal operator. This increased the duration of the farm visits significantly and in a number of cases additional appointments had to be made to collect all necessary input. Because of the volume of PPP data collected, new priorities had to be decided on which meant that some of the operator/worker data were not gathered. Especially the work activity details of the principal spray operator were likely to be incomplete for many farms. These data could however contain some useful information on weekly work and the range of work activities conducted by the principal operator, but these could not be further discussed due to the lack of accurate and complete data.

A total of 30 farms were surveyed in Flanders mainly cultivating maize, potato and sugar beet. Of all crop groups surveyed in 2013, potatoes indicated the largest treated area. The treated area of sugar beet and maize were even two and five times smaller compared to potatoes (Table 3-3). Most active substances were applied on wheat and potato, followed by sugar beet, barley, strawberries and other cereals (Table 3-4). Potatoes are susceptible to various diseases and pests. During the season, most farmers spray every 7 to 10 days, depending on the weather, with a PPP against late blight (*Phytophthora infestans*). The need for chemical control can only be limited by choosing disease-resistant varieties. A total of 37 sprayers were surveyed and all of these sprayers were hydraulic boom sprayers. When the sprayer cab is fully closed and fitted with an operational active carbon filter, it offers the same level of protection as the highest level of PPE (Garreyn et al., 2007). Results of this survey indicated that especially closed cab sprayers were used, but only 16.2% contained a carbon filter (3.1.1.2). However, keeping all windows closed may be a problem during

summer and most farmers probably do not replace the carbon filter often enough either. Especially Flat Fan nozzles were used (Table A-8 of Appendix A), but nozzle data for future surveys could be improved. On the one hand, the use of for example Tee Jet as a category has already been mentioned as unnecessary, as it is a manufacturer's name and not a nozzle type. On the other hand, it was only possible to add low-drift status information where the actual name of each nozzle was recorded. A number of nozzle types could be low-drift, but could not be recorded as such because of insufficient information. Adding a question on the Local Environmental Risk Assessment for Pesticides (LERAP) or drift status of other nozzle types would be useful. Technology, such as low-drift nozzles, can reduce PPP drift up to 90% (Reichenberger et al., 2007; Roettele et al., 2012; Doruchowski et al., 2013). Collecting detailed nozzle data is very useful in order to perform ERAs for PPPs, e.g. to estimate the risk indicator for aquatic organisms (POCER, 1.2). The average age of Flemish spray operators was 48 years, just like the age observed in studies conducted in the UK (Garthwaite, 2002, 2004a, 2004b). In 2013, not everyone already disposed of a spray certificate as it was not mandatory yet, i.e. only 42% of all operators had a nationally recognised spray certificate. This certificate is called 'Phytolicence' in Belgium and is delivered by the Federal Public Service Health, Food Chain Safety and Environment (FPS) that ensures that all professional use of PPPs is based on sufficient knowledge and proficiency, and that is regularly updated. The phytolicence is mandatory as of 25 November 2015 for all professional users, distributors and advisors. Operators who mix, load and apply PPPs are usually considered to receive the highest exposure due to the nature of their work, and are therefore at highest risk for acute intoxications. Exposure during specific handling events can be modified by several factors, such as type of equipment used, PPP formulation, packaging, environmental conditions, PPE, hygienic behaviour, duration of the activity, etc. (Fenske and Day, 2005; Harrington et al., 2005). An examination of the principal operator to individual active substances throughout the 2013 cropping year was also performed. The principal operator's daily duration of exposure per farm per active substance used was on average 3.8 hours. Further, the number of individual active substances the principal operator could be exposed to was on average 37. Wearing personal protective equipment – and especially gloves – can reduce the risk of dermal exposure during mixing and loading (Harrington et al., 2005) and is therefore highly recommended. Almost all operators surveyed in 2013 protected themselves with gloves, coveralls and boots during mixing and loading. During PPP application, gloves were hardly used (Table 3-8; Table A-14 of Appendix A). Application of PPPs was especially performed by using a hydraulic boom sprayer, so PPP exposure to hands was limited. Cleaning the sprayer, for example, is an activity that brings the operator closer to PPPs than would be experienced when in a spray cab. Therefore, also during the cleaning of the sprayer and performing other work activities in the field, the use of gloves, coveralls and boots is recommended to reduce the risk of dermal exposure of PPPs. Some general information on the adoption of IPM practices was collected during the survey in 2013 (3.1.3.1). However, detailed information on IPM practices could be useful in order to perform ERAs for PPPs, e.g. the buffer strip width which was not registered in the survey of 2013. In Belgium, authorisations for most PPPs containing active substances that are known to pose riks to aquatic organisms stipulate a minimum buffer strip width. Further, buffer strips may provide a recovery potential for the cropped areas.

Together with the principal operator information, the crops maize, potato and sugar beet were selected and information was collected on twenty ERA fields for each crop. As described in 3.1.3.2, ERA fields of maize, potato and sugar beet were also surveyed in 2013 in Italy, Poland, Lithuania, the Netherlands and United Kingdom. These data were used to compare the use of PPPs on ERA fields in Flanders with other countries cultivating similar crops. The average number of products applied to the ERA fields of maize in Italy (Southern zone) was little higher than in Flanders. The average number of spray rounds was even twice as high compared to Flanders. On the ERA fields of maize in Poland, belonging to the Central zone like Belgium, on average only one product was applied per hectare whereas the average number of spray rounds was approximately the same as in Flanders (Table 3-5; Table 3-10). Both herbicides and insecticides (especially in Italy, i.e. 24%) were applied and a total of 21 and 15 active substances were used on the ERA fields of maize in Italy and Poland respectively (17 a.s. used in Flanders). The top five applied active substances differed per country. In Italy, terbuthylazine, tefluthrin, dicamba, nicosulfuron and lambda-cyhalothrin were used, whereas especially the herbicides nicosulfuron, mesotrione, metolachlor, terbuthylazine and acetochlor were applied in Poland. In Belgium, the insecticide tefluthrin is not authorised for use in the crop of maize. Further, the herbicide acetochlor is not authorised as herbicide in the EU since 2008. As originally formulated, metolachlor (herbicide) was applied as a racemate, a 1:1 mixture of the (S)- and (R)-stereoisomers. This mixture is not authorised in Belgium, only S-metalochlor can be used. In Italy, PPP application already started in April while in Belgium and Poland spraying of PPPs began as of May (Table 3-11). In the Northern zone (Lithuania), the number of products applied on the ERA fields of potato were on average five times lower than in Flanders and also the average of 3.4 spray rounds was four times lower. Especially fungicides were applied followed by insecticides and herbicides, and to a lesser extent defoliants and desiccants (Table 3-5; Table 3-14). The total number of active substances applied on these fields was lower than in Flanders. Dimethomorph, ametoctradin, alpha-cypermethrin, mancozeb and propamocarb hydrochloride were especially used. In the Netherlands (Central zone), both the average number of products as the average number of spray rounds were higher than in Flanders (Table 3-5; Table 3-14). Especially fungicides were applied followed by insecticides and herbicides, and to a lesser extent growth regulators, sprout suppressants and nematicides. A total of 39 active substances were used. The top five applied active substances were mancozeb, cyazofamid, fluopicolide, propamocarb and cymoxanil. Belgium and the Netherlands had a similar distribution throughout the year, while Lithuania appeared to be more focused on applications in June (Table 3-15). The UK ERA fields of sugar beet were treated on average with approximately the same number of products as in Flanders. The average number of spray rounds was the same as well (Table 3-5; Table 3-18). Herbicides were mainly applied on the UK ERA fields of sugar beet, just like in Belgium. On these fields, a limited amount of fungicides, insecticides and molluscicides were applied as well and a total of 28 active substances were used. In the UK, the top five applied active substances were phenmedipham, ethofumesate, desmedipham, lenacil and metamitron. The distribution of PPP applications throughout the year for both countries appeared to be quite similar (Table 3-19).

In general, the pattern of PPP use differs between countries. Countries belonging to the same regulatory zone show similarities in PPP use. Lithuania's climate (Northern zone) ranges between maritime and continental and is relatively mild. Some winters can be very cold. The United Kingdom has a temperate climate, with plentiful rainfall all year round. The temperature varies with the seasons seldom dropping below -11 °C or rising above 35 °C. In Belgium, the climate is maritime temperate with significant precipitation in all seasons. The average temperature is lowest in January at 3 °C and highest in July at 18 °C. The predominant wind direction in the Netherlands is southwest, which causes a moderate maritime climate, with cool summers and mild winters, and typically high humidity. In Poland, the climate is mostly temperate throughout the country. The climate is oceanic in the north and west and becomes gradually warmer and continental towards the south and east. Summers are generally warm, winters are rather cold. All these countries belong to the Central zone. Italy belongs to the Southern regulatory zone and has a Mediterranean climate, which is characterised by warm to hot, dry summers and mild to cool, wet winters. Differences in the weather, influencing the range of pest, disease and weed problems requiring control, or affecting the ability of the farmer to apply the PPP under suitable conditions. Further, pest pressure may be higher during warm weather resulting in higher inputs to some crops, e.g. during the summer than in the winter (Eurostat, 2008; Regulation (EC) No 1107/2009; Wais, 2012). The crop type also influences PPP usage, some crops e.g. potatoes are more susceptible to various pests and diseases.

## 3.2 Upgrading POCER with questionnaire results

## 3.2.1 Case study fruit production

In this section, some simulations of the effects of modifications in crop protection practices on POCER RIs are discussed. All simulations and application schemes were for mechanical (vertical) spray applications in apple plantations.

## 3.2.1.1 Risk for operator and re-entry worker

Figure 3-2A shows how the operator's risk can be reduced when he wears PPE instead of jeans and a T-shirt. RI<sub>operator</sub> decreases by 33% when wearing gloves while mixing and loading (ML) maneb WP into the spray tank. Wearing a mask reduces risk with an additional 1%. By wearing gloves and coveralls while spraying (or in a fully closed cab), RI<sub>operator</sub> again decreases strongly. When the operator is wearing full PPE, his risk is reduced by 85%. Analogously, the re-entry worker's risk (RE), when manipulating sprayed trees, is reduced by

79% by wearing gloves and coveralls (Figure 3-2B). Risk is mainly reduced by wearing body protection, as in fruit trees the whole body comes into contact with treated leaves, e.g. when thinning fruit. In this open air case, the additional protection provided by a mask is marginal, unlike in greenhouses. Another possibility to reduce the operator's risk is to use a granulated formulation (WG), instead of a powder (WP), as the chances are smaller that undiluted granulates are blown onto the skin or that they are inhaled. Even without PPE, RI<sub>operator</sub> is 37% smaller when handling maneb WG instead of maneb WP (Figure 3-2C). Once the WP or WG formulations are diluted into spray mixtures with the same concentrations, risk exerted by either one is similar and RI<sub>operator</sub> during application (AP) or RI<sub>re-entry</sub> are not reduced (Figures 3-2C and 3-2D).



Figure 3-2 The effect of additional personal protective equipment (PPE) on the operator's risk while handling maneb WP (A) and maneb WG (C) and the re-entry worker's risk while working in fruit trees treated with maneb WP (B) and maneb WG (D). Application rate = 2.4 kg active substance/ha; maneb WP (80% maneb, Trimangol 80, wettable powder (WP), UPL Europe LTD) and maneb WG (75% maneb, Trimangol WG, water dispersible granules (WG), UPL Europe LTD); no PPE = wearing no PPE ; ML = mixing and loading :  $ML_{hand}$  = hand protection during ML (gloves);  $ML_{body}$  = body protection during ML (gloves + coveralls);  $ML_{mask}$  = inhalation protection during ML (gloves + coveralls + mask); AP = application :  $AP_{hand}$  = hand protection during AP (full PPE-ML + gloves);  $AP_{body}$  = body protection during AP (full PPE-ML + gloves + coveralls + mask); RE = re-entry :  $RE_{hand}$  = hand protection during RE (gloves);  $RE_{mask}$  = inhalation protection during RE (gloves); Most RIs are proportional to the PPP application rate. Figure 3-3A and 3-3B show how the operator's and the re-entry worker's risk are reduced when the application rate of the fungicide mancozeb WP is reduced from 3 to 1.8 kg/ha in apples. RI<sub>operator</sub> is reduced by 45% as he needs to handle smaller amounts of PPPs to protect the same area. RI<sub>re-entry</sub> is also reduced by 45% as he comes into contact with smaller residue concentrations. Applying PPPs at smaller rates, when possible, not only reduces cost, but also human risk. Risk is further reduced by wearing PPE (Figure 3-3C and 3-3D).



Figure 3-3 The effect of application rate and the effect of additional personal protective equipment (PPE) at 1.8 kg product/ha on the operator's risk while handling mancozeb WP (A, C) and on the reentry worker's risk while working in fruit trees treated with mancozeb WP (B, D). Mancozeb WP (75% mancozeb, Dequiman MZ WP, wettable powder (WP), UPL Europe LTD); no PPE = wearing no PPE  $\blacksquare$ ; ML = mixing and loading  $\blacksquare$ : ML<sub>hand</sub> = hand protection during ML (gloves); ML<sub>body</sub> = body protection during ML (gloves + coveralls); ML<sub>mask</sub> = inhalation protection during ML (gloves + coveralls + mask); AP = application  $\blacksquare$ : AP<sub>hand</sub> = hand protection during AP (full PPE-ML + gloves); AP<sub>body</sub> = body protection during AP (full PPE-ML + gloves + coveralls); RE = re-entry  $\blacksquare$ : RE<sub>hand</sub> = hand protection during RE (gloves); RE<sub>mask</sub> = inhalation protection during RE (gloves) + coveralls + mask).

The examples above show how DISCUSS and POCER can be used as decision-making support tools, e.g. when apple growers need to decide on how to handle a particular PPP. The following example shows how it can help decide between PPPs. When a PPP consists of two or more active substances in POCER the RIs are added up. Figure 3-4A shows RI<sub>operator</sub> when

fluquinconazole and pyrimethanil are combined (e.g. in the formulation of Vision). Without PPE RI<sub>operator</sub> = 1 for fluquinconazole and a small additional risk is posed by pyrimethanil. RI<sub>operator</sub> can be reduced by 84% by wearing full PPE. However, it would be even better to substitute fluquinconazole by another fungicide from the same group, difenoconazole, which poses only a very small risk to humans. Even without PPE, RI<sub>operator</sub> is thus reduced by 97% to 0.03 (Figure 3-4B). Since May 2014, the product Vision is no longer authorised in Belgium. However, this example illustrates the effect of substituting between PPPs to reduce the risk.



Figure 3-4 The effect of plant protection product substitution on the operator's risk while handling combinations of pyrimethanil  $\blacksquare$  with fluquinconazole (A) or difenoconazole (B). Vision (50 g/l fluquinconazole, 200 g/l pyrimethanil, suspension concentrate (SC), Bayer CropScience, 1.5 l product/ha), Geyser (250 g/l difenoconazole, emulsion concentrate (EC), Syngenta Crop Protection, 0.15 l product/ha) and Scala (400 g/l pyrimethanil, SC, BASF Belgium, 1.125 l product/ha); no PPE = wearing no personal protective equipment  $\blacksquare$ ; ML = mixing and loading  $\blacksquare$ : ML<sub>hand</sub> = hand protection during ML (gloves); ML<sub>mask</sub> = inhalation protection during ML (gloves + coveralls + mask); AP = application  $\blacksquare$ : AP<sub>hand</sub> = hand protection during AP (full PPE-ML + gloves); AP<sub>body</sub> = body protection during AP (full PPE-ML + gloves + coveralls).

### 3.2.1.2 Risk for aquatic organisms

Rl<sub>aquatic organisms</sub> can be mitigated by leaving buffer strips unsprayed along surface water or by using drift-reducing technology. Technology, such as specific sprayer types (e.g. shielded tunnel sprayers, sensor-equipped sprayers that avoid spraying in the gaps between trees) or low-drift nozzles, can reduce PPP drift up to 90% (Reichenberger et al., 2007; Roettele et al., 2012; Doruchowski et al., 2013). Figure 3-5 shows some examples of the effect of buffer strip width and drift-reducing technology on the risk posed to aquatic organisms.

In Belgium, authorisations for most products containing active substances that are known to pose risks to aquatic organisms stipulate a minimum buffer and/or drift reduction.

Application of the fungicide metiram in apple plantations, for example, is only allowed with a 30 m buffer and 90% drift reduction. Under these conditions  $RI_{aquatic organisms}$  is only 0.04. In standard POCER calculations compliance with the application restrictions is assumed. Figure 3-5A shows how  $RI_{aquatic organisms}$  would increase if buffer width or drift reduction were to be reduced. For example, with only a 5 m buffer and 50% drift reduction  $RI_{aquatic organisms}$  becomes > 1, i.e. unacceptable, but with the best drift-reducing technology it would remain limited to 0.23. Without any buffer strip, not even the best technology could achieve  $RI_{aquatic organisms} < 1$  though.



Figure 3-5 The effect of buffer strip width (m) and drift-reducing spray technology (0% ■, 50% ■, 75% ■ and 90% ■ drift reduction) on the risk posed to aquatic organisms (aq. org.) by (A) metiram (80% metiram, Polyram WG, water dispersible granules (WG), BASF Belgium, 2.625 kg product/ha) and (B) spinosad (120 g/l spinosad, Boomerang, suspension concentrate (SC), Dow AgroSciences B.V., 0.9 l product/ha).

Figure 3-5B shows an analogous simulation for the insecticide spinosad. Here Belgian authorisation stipulates at least a 20 m buffer and 50% drift reduction, resulting in Rl<sub>aquatic</sub> organisms = 0.79. A similar risk could be achieved with only a 10 m buffer, but 75% drift reduction. Such precautionary measures are indispensable, as with < 50% drift reduction and < 20 m buffer Rl<sub>aquatic</sub> organisms becomes > 1. Spinosad is a biopesticide but not per definition safe for aquatic organisms. Larger buffer strips and more drift-reducing technology could further reduce Rl<sub>aquatic</sub> organisms. In apples, the maximum application rate for spinosad is 0.1 kg/ha in Belgium while this is 0.2 kg/ha in pears (*Pyrus*). As described above for human risk (3.2.1.1), also the risk exerted on aquatic organisms increases with the application rate. This means that stricter precautions are needed when applying spinosad at the maximum authorised rate in pears than when applying the maximum authorised rate in apples.

### 3.2.1.3 Spray record analysis

Even very detailed farm analyses and recommendations can be made based on their spray records. Figure 3-6 shows an example of such an analysis: per spraying event total human and environmental risk are set out over an entire growing season. In this example the risk summed over 5 human or 7 environmental compartments is quite acceptable for most spraying events. A peak in environmental risk was incurred with the very first treatment of the season on March 22<sup>nd</sup>. A preventative copper oxychloride spray was carried out against fire blight and scab, resulting in RIgroundwater = 1, RIpersistence = 1 and RIaquatic organisms = 0.55. Since fire blight is a quarantine pest (EPPO, 2014), treatment can hardly be omitted. It would be better to search for an alternative though, since the risks for groundwater and persistence cannot be mitigated. Other active substances authorised in Belgium are fosetyl, laminarin and Aureobasidium pullulans. All pose very small environmental risk (as far as data are available), but all three also require several sprayings during the flowering season, making them costly treatments. The second risk peak on April 6<sup>th</sup> was caused by a combined spray application of imidacloprid against rosy apple aphid, causing RI<sub>bees</sub> = 1, RI<sub>persistence</sub> = 0.74 and RIgroundwater = 0.48, and dodine against scab, with RI<sub>aquatic organisms</sub> = 0.55. Imidacloprid entails unacceptably large risk for bees. Fortunately, this application was done pre-bloom. If flowering weeds should be present, an alternative (although more expensive) could be thiacloprid. The peaks in human risk on July 16<sup>th</sup> and 23<sup>rd</sup> were caused by thiram. They could be mitigated by the use of protective clothing as illustrated before.

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Figure 3-6 Total human and environmental risk per spraying event and cumulated risk for the entire spray records of a randomly selected Belgian farm growing Jonagold apples in 2012 (RI<sub>human</sub> ■, RI<sub>environment</sub> ■, Cumulated RI<sub>human</sub> ■ and Cumulated RI<sub>environment</sub> ■).

## 3.2.2 Case study potatoes

Spray records of 25 farms were simulated. For each farm size class, farms with the highest and lowest RIs were compared and discussed. Only 10 farmers used buffer strips to prevent drift on any fields on their farm but the exact buffer strip width was not registered. A minimum buffer strip of 2 m was assumed for these farmers whereas the worst-case situation (buffer strip of 1 m) was used for the remaining farmers (prevailing legislation, Vandewalle et al., 2014). Only 20% and 12% of the farmers used a 50% and 75% driftreducing spray technology respectively.

## 3.2.2.1 Farm size class A (< 5 ha)

Figure 3-7 illustrates the cumulated human and environmental risk for the entire spray records of three farms belonging to farm size class A. The results of farm 3 indicated the highest RI in terms of human risk whereas the lowest RI<sub>human</sub> was obtained for farm 2. RI<sub>operator</sub> of farm 3 was even > 1 which is unacceptable. Farmer 3 wore only gloves during mixing and loading but no further PPE during application and the performance of other work activities. Wearing full PPE during mixing and loading, such as farmer 2, reduced RI<sub>operator</sub>. During PPP application, farmer 2 used body and hand protection so RI<sub>operator</sub> was again lower compared to farm 3. During re-entry into the field, only farmer 2 wore body protection

indicating the lowest RI<sub>re-entry worker</sub>. Further, the use of prosulfocarb on all farms resulted in higher RIs for both operators and re-entry workers compared to the use of other active substances. Prosulfocarb is classified under Class III substances that permit no strong initial presumptions of safety, or that may even suggest significant toxicity. They thus deserve the highest priority for investigation (Cramer et al., 1978; Lewis and Green, 2011). Farmer 3 also used diquat, which poses a large risk to humans (Class III, Lewis and Green, 2011).



Figure 3-7 Cumulated human and environmental risk for the entire spray records of three Flemish farms belonging to farm size class A (< 5 ha) surveyed in 2013 (Farm 1 , Farm 2 , Farm 3 ).

Both farm 2 and 3 indicated high RIs in terms of environmental risk. Especially the risk for aquatic organisms and beneficial arthropods were > 1 for all farms. Farm 3 also had a high RI<sub>persistence</sub> because of the use of the highly persistent diquat ( $DT_{50 \text{ soil}} = 5475$  days). All farmers used a classical spray technology (0% drift-reducing) and only farm 3 used a minimum buffer strip of 2 m. Despite the buffer strip of 2 m, the highest RI<sub>environment</sub> was obtained for farm 3, but RI<sub>aquatic organisms</sub> was lower than farm 2 having a buffer strip of 1 m. The use of various active substances resulted in RI<sub>aquatic organisms</sub> > 1, i.e. aclonifen, prosulfocarb, mancozeb and famoxadone. All farmers used aclonifen (in the formulation of Challenge) which should be applied with a 20 m buffer and 50% drift reduction. Prosulfocarb which poses a large risk to aquatic organisms, was used by farmer 1 and 2 in the formulation of Defi (1 m and classical) and Roxy EC (10 m and classical) respectively. Farmer 2 and 3 applied mancozeb in different formulations whereby the legal buffer strip width and spray

technology were not respected, e.g. Ebrimax WG (10 m and classical) and Festival (20 m and 75% drift-reducing). Further, farmer 2 also used famoxadone in the formulation of Tanos which should be applied with a buffer strip of 20 m using a classical spray technology. Risk for beneficial arthropods was high for all farms. This high risk was caused by the use of active substances which pose potentially high risk to beneficial arthropods, i.e. prosulfocarb, linuron, cymoxanil, mancozeb, pyraclostrobin and ametoctradin. Farmer 3 applied most of these substances resulting in the highest RI<sub>beneficial arthropods</sub>.

## 3.2.2.2 Farm size class B (5-10 ha)

The cumulated human and environmental risk for the entire spray records of two farms belonging to farm size class B are displayed in Figure 3-8. Both farm 1 and 2 had a Rl<sub>operator</sub> and Rl<sub>re-entry worker</sub> < 1. However, a difference between the farms was noticed. Both farmers used gloves and body protection during mixing and loading, and wore coveralls during application. During re-entry into the field, farmer 1 wore no PPE while farmer 2 used body protection. Farmer 1 applied diquat (Class III, Lewis and Green, 2011) resulting in a higher Rl<sub>operator</sub> and RI<sub>re-entry worker</sub> compared to farm 2. Further, farmer 2 also applied prosulfocarb which affected the RIs for both operator and re-entry worker (Class III, Lewis and Green, 2011).



Figure 3-8 Cumulated human and environmental risk for the entire spray records of two Flemish farms belonging to farm size class B (5-10 ha) surveyed in 2013 (Farm 1 , Farm 2 ).

Rl<sub>environment</sub> was lower for farm 2 than for farm 1, but the risk for aquatic organisms, groundwater and beneficial arthropods were > 1 for all farms. Farm 1 also had a high Rl<sub>persistence</sub> because of the use of the highly persistent diquat. All farmers used a buffer strip of 2 m. Farmer 1 used a classical spray technology, while farmer 2 applied PPPs using a 75% drift-reducing technology resulting in a lower Rl<sub>aquatic organisms</sub> compared to farm 1. The higher risk for aquatic organisms of farm 1 was also determined by the application of two active substances, i.e. the use of aclonifen (in the formulation of Challenge) and the use of maneb (in the formulation of Trimangol 80) which should be applied with a 20 m buffer, 50% drift reduction and a 5 m buffer, classical spray technology respectively. Both the use of metribuzin and fluopicolide (potential leaching behaviour, mobile PPPs), resulted in high risks for groundwater on both farms (Lewis and Green, 2011). Risk for beneficial arthropods was caused by the use of various active substances, i.e. pencycuron, linuron, cymoxanil, flonicamid, pyraclostrobin, prosulfocarb and mancozeb. All of these substances are harmful to beneficial arthropods.

#### 3.2.2.3 Farm size class C (10-20 ha)

Figure 3-9 presents the cumulated human and environmental risk for the entire spray records of two farms belonging to farm size class C. Only the results of farm 1 indicated a high RI in terms of human risk. RI<sub>operator</sub> of farm 1 was even > 1. Both farmers used body protection during mixing and loading, application and re-entry into the field. Only farmer 2 used gloves during mixing and loading. The high risk for operator and re-entry worker of farm 1 was especially caused by the use of both diquat and prosulfocarb (Class III). Furthermore, farmer 2 also applied glufosinate which is classified under Class III substances (Lewis and Green, 2011).

Rl<sub>environment</sub> was lower for farm 1 than for farm 2, but the risk for both aquatic organisms and beneficial arthropods was > 1 for all farms. The high risk for soil persistence of farm 1 was caused by the use of diquat (highly persistent). Further, Rl<sub>groundwater</sub> of farm 2 was high due to the use of two mobile PPPs, i.e. metribuzin and fluopicolide (Lewis and Green, 2011). All farmers used a classical spray technology (0% drift-reducing) and only farm 1 used a minimum buffer strip of 2 m. The higher risk for aquatic organisms of farm 2 was especially determined by the application of two active substances, i.e. the use of flufenacet (in the formulation of Artist) and the use of mancozeb (in the formulation of Cymozeb WG) which should be applied with a 20 m and 10 m buffer, classical spray technology respectively. Both farmers also applied aclonifen in the formulation of Challenge (20 m and 50% drift-reducing). Risk for beneficial arthropods was high for both farms. This high risk was caused by the use of active substances which pose potentially high risk to beneficial arthropods, i.e. prosulfocarb, linuron, cymoxanil, mancozeb, flonicamid, flufenacet and ametoctradin. Farmer 2 applied more of these substances than farmer 1, resulting in the highest Rl<sub>beneficial</sub> arthropods.

Chapter 3



Figure 3-9 Cumulated human and environmental risk for the entire spray records of two Flemish farms belonging to farm size class C (10-20 ha) surveyed in 2013 (Farm 1 , Farm 2 ).

## 3.2.2.4 Farm size class D (> 20 ha)

The cumulated human and environmental risk for the entire spray records of three farms belonging to farm size class D are illustrated in Figure 3-10. The results of farm 2 indicated the highest RI in terms of human risk whereas the lowest RI<sub>human</sub> was obtained for farm 1. RI<sub>operator</sub> of farm 2 was even > 1, and also RI<sub>re-entry worker</sub> of farm 2 and 3 exceeded the upper limit. All farmers used hand and body protection during mixing and loading, and wore coveralls during application and re-entry into the field. All farmers applied diquat (Class III, Lewis and Green, 2011). Farmer 1 and 3 applied diquat only once in the formulation of Quad Glob 200 SL and Mission 200 SL respectively. Farmer 2 used the product Diqua two times. All these products contain 200 g/l diquat and a maximum application rate of 4 l/ha for potatoes. Farmer 1 used an applied prosulfocarb which poses a risk to humans indicating a higher RI<sub>operator</sub> and RI<sub>re-entry worker</sub> compared to farm 3.

Both farm 1 and 2 indicated high RIs in terms of environmental risk. Especially the risk for beneficial arthropods and soil persistence were > 1 for all farms.  $RI_{persistence}$  was high due to the use of the highly persistent diquat. High RIs for groundwater of farm 2 and 3 were caused due to the use of metribuzin. The mobile fluopicolide was also applied on farm 2.

Farmer 1 and 2 used a classical spray technology, farmer 3 used a 75% drift reduction technology resulting in the lowest RI in terms of environmental risk. Only a minimum buffer strip of 1 m was used on farm 1. Risk for aquatic organisms exceeded the upper limit for farm 1 and 2 due to the use of various active substances. Both farmers used flufenacet in the formulation of Artist (20 m and classical). Mancozeb was applied as Cymozeb WG on farm 1 and as Acrobat Extra WG on farm 2. These products should be applied with a 10 m and 20 m buffer using classical and 75% drift-reducing technology respectively. On farm 2, indicating the highest RI<sub>aquatic organisms</sub>, also the active substances maneb (Trimangol WG: 5 m, classical) and aclonifen (Challenge: 20 m, 50% drift-reducing) were used. Risk for beneficial arthropods was high for all farms. This high risk was caused by the use of active substances which pose potentially high risk to beneficial arthropods, i.e. prosulfocarb, linuron, cymoxanil, mancozeb, pyraclostrobin, ametoctradin, flufenacet, pendimethalin, acetamiprid and chlorpropham. Farmer 2 applied more of these substances compared to farmer 1 and 3 resulting in the highest RI<sub>beneficial arthropods</sub>.



Figure 3-10 Cumulated human and environmental risk for the entire spray records of three Flemish farms belonging to farm size class D (> 20 ha) surveyed in 2013 (Farm 1 , Farm 2 , Farm 3 ).

### 3.2.2.5 Discussion

The UK experience showed that the behaviour of operators and workers varies significantly between smaller and larger farms (Garthwaite, 2002, 2004a, 2004b). Therefore a wide range

of farm sizes were included in order to ensure that the survey was representative of these behaviours. However, an obvious relation between risk for human health and the environment and farm size classes, and more precisely area of potatoes grown, could not be observed for the farms investigated here. Within each class, variation is also quite large. Despite, some general conclusions concerning human and environmental risk could be listed as follows.

The difference between farms could be explained by the use of PPE. Potential exposure to PPPs is highest during mixing and loading, and this mainly at the height of hands and chest (Fenske and Day, 2005; Harrington et al., 2005). Wearing personal protective equipment and especially gloves and coveralls – can reduce RI<sub>operator</sub> during mixing and loading (3.2.2.1; 3.2.2.3). As described in 3.1.1.2, farmers used especially closed cab sprayers and farmers tend to rely heavily on the protection it offers. However, not all sprayers contained a carbon filter. Only when the sprayer cab is fully closed and fitted with an operational active carbon filter, it offers the same protection level as the highest level of PPE (Garreyn et al., 2007). Keeping all windows closed may also be a problem during summer and most farmers probably do not replace the carbon filter often enough. Consequently, by only wearing gloves and coveralls during application of PPPs, RI<sub>operator</sub> can be strongly reduced (3.2.2.1). During re-entry into the field, exposure to PPPs can also occur. Like the operator, also the reentry worker can protect himself by wearing PPE. In potatoes, wearing coveralls already had a considerable effect and wearing gloves will further reduce the risk (3.2.2.1; 3.2.2.2). The utmost care must be taken to avoid exposure during handling operations and application in the field. In case of herbicides such as diquat, prosulfocarb and glufosinate which pose large risk to humans, wearing full PPE is highly recommended both for operators and re-entry workers. In general, all farmers applied the amount as indicated on the PPP labels. Using a lower application rate resulted in a lower RI<sub>human</sub>, as human risk decreases proportionally to a decreasing application rate (3.2.2.4). However, it is possible that the desired effect is not reached when using a lower dose than recommended.

On most farms, RIs for aquatic organisms were high, but both the use of buffer strips and drift-reducing technology can strongly reduce RI<sub>aquatic organisms</sub>. Authorisations for most Belgian PPPs containing active substances that are known to pose risk to aquatic organisms stipulate a minimum buffer and/or drift reduction. Only 40% of the farmers used buffer strips but the exact buffer strip width was not registered. Still 64% of the farmers applied PPPs with a classical spray technology. To reduce RI<sub>aquatic organisms</sub> PPPs should be applied according to prevailing legislation, i.e. by using drift-reducing technology and buffer strips along surface water stipulated on the product labels. Buffer strips may also provide a recovery potential for the cropped areas. Risk for groundwater exceeded the upper limit for some farms (3.2.2.2; 3.2.2.3; 3.2.2.4). Calculating RI<sub>groundwater</sub> was based on the GUS indicator (Ground Ubiquity Score), which describes the infiltration of the PPP in the soil. The higher the GUS value, the higher the potential for PPPs to move toward groundwater (Gustafson, 1989). The use of mobile PPPs, such as metribuzin and fluopicolide, should be avoided to lower risk for

groundwater. All farms had high RIs for beneficial arthropods (3.2.2.1; 3.2.2.2; 3.2.2.3; 3.2.2.4). This was caused by the use of various substances which are harmful to beneficial arthropods. Beneficial arthropods, as aphid predators and parasites, are the key of an effective aphid control in potato. However, these insects are exposed to PPPs applied during the season, especially fungicides used to control late blight and insecticides used to control aphids and the Colorado beetle. The conservation of natural enemies' population by the use of products that are selective for them is required in the context of IPM. In 2005, a selectivity list of PPPs on beneficial arthropods in potato was established. The results obtained in the study showed that it is possible to control pests and diseases with products that are selective towards main aphid natural enemies, during all periods in which these beneficial insects are active in the field. These selectivity lists can help the farmers choosing the appropriate PPP. The lists are distributed to the farmers yearly by the organisms in charge of the advisory systems and are also used for several guidance documents (IPM, specific labels, etc.). They are regularly updated to include all changes in the list of available products, as some products are banned and new ones are added (Hautier et al., 2004, 2006; Jansen, 2014). The POCER risk index for beneficial arthropods does not take into account the application period and beneficial insects phenology. As all non-target biota within or outside the treated fields should be safeguarded for their intrinsic value, it would be useful for farmers to consult the selectivity lists to choose the product to spray in order to reduce risk for beneficial arthropods. These lists can also complete the information given by potato advisory systems for aphids control. Furthermore, toxicity varies strongly between PPPs. If risk is considered too high, operators may also opt to substitute between PPPs. For example, potato growers might consider substituting the highly persistent diquat by another PPP authorised in Belgium for haulm destruction in potatoes, e.g. carfentrazone-ethyl and pyraflufen-ethyl. These herbicides pose smaller risks for all compartments. When doing so, growers will need to apply the substitutes somewhat earlier before harvest, as the safety period for carfentrazone-ethyl and pyraflufen-ethyl is 14 days, compared to only 4 days for diquat. Finally, when a PPP consists of two or more active substances in POCER, the RIs are added up. This might also result in higher risks compared to the use of single active substances. The number of PPP applications per holding varied between 10 and 24, but exerted no direct influence on the RIs whereas the use of certain PPPs (e.g. diquat) can strongly determine the risks in terms of human health and the environment.

An additional important note is that the results of the risk calculations indicated different risks for bees than for beneficial arthropods. The determination of these risks is based on various assumptions and includes other formulas. In the context of sustainable development and the preservation of ecosystems, it is important to minimise the disruption of essential links. This disruption is difficult to control, but an evaluation of the possible risks for non-target organisms caused by PPP use can be made, e.g. by determining the toxic effects of PPPs on highly sensitive organisms such as bees. Risk for bees is assumed to exist when they visit treated plants for pollination or honey production during the flowering season (Vercruysse and Steurbaut, 2002). Therefore, a risk indicator for bees was established and

included in POCER. Both the application rate and toxicity for bees (expressed as LD<sub>50</sub>) of the applied PPPs are taken into account when calculating the risk for bees. The side-effects of PPPs to beneficial arthropods – other than bees – in general and arthropod natural enemies more specific (e.g. Typhlodromus pyri, Encarsia formosa) are expressed as the reduction in control capacity (i.e. the reduction in natural enemy potential or effectiveness, which if reduced could lead to higher pest numbers) (EPPO, 1994). This refers to effects such as mortality and non-hatching of eggs and pupae and to sub-lethal effects such as reduced fertility, problems with regard to moulting, repellency, etc. The side-effects of beneficials used in the POCER indicator are derived from the online side-effect database of Koppert (Koppert, 2016) and the database of Biobest (Biobest, 1999). The side-effects of PPPs in these databases are classified into four categories (harmless, slightly harmful, moderately harmful and very harmful) according to IOBC guidelines (International Organisation for Biological and Integrated Control) which determine the reduction of control capacity (EPPO, 1994). The risk for beneficial arthropods is set up in such a manner that a PPP which is harmless (according to the side-effect databases) for beneficial arthropods leads to a RI<sub>beneficial arthropods</sub> = 0 whereas a PPP which is very harmful for beneficial arthroprods results in a RI<sub>beneficial arthropods</sub> = 1. This risk assessment focuses on the protection of arthropods which are used as natural enemies to pests, diseases or weeds. This is often the case in integrated farming. Therefore, PPPs are classified according to their ability to use them in integrated farming. The application period, beneficial insects phenology and the specific toxicity parameters (like  $LD_{50}$ ) are not included in the POCER risk index for beneficial arthropods. Due to these limitations, POCER risk calculations for beneficial arthropods should be treated with caution. Therefore, the use of selectivity lists is recommended in order to reduce risks for beneficial arthropods.

# Conclusion

Overall, a large, unique and high quality dataset relating to PPP application and usage was collected in Flanders (Belgium). Analysis of the data was provided in this study but subsequently the dataset can be used to undertake more in-depth investigations of cumulative PPP exposure to spray operators. These in-dept investigations have to focus on mixing and loading, spraying and cleaning activities whether or not combined with other worker activities that will be added to the operator exposure. Moreover, the data collected on the ERA fields can be investigated and used to subsequently build environmental scenarios for the purposes of environmental risk assessment. The data were collected in order to improve models of operator cumulative exposure and to support the revision of ecotoxicological guidance documents; it was not intended to produce national estimates of PPP usage.

By pairing POCER with a farm level questionnaire, the Pesticide Occupational and Environmental Risk indicators can be calculated more accurately than before. The RIs thus concur more closely with the individual farmer's actual crop protection practices. Both case

studies illustrated how responsive modifications in crop protection practices can reduce human and environmental risk. Compared to earlier sector and regional level implementations, for which the RIs used to be calculated using worst-case scenarios and prevailing legislation, the refined RIs make a better starting point for discussion between farmers. It has been shown that these RIs can serve as decision support tools, because simulations of management changes or substitution between PPPs can help farmers to decide on the most sustainable alternatives.
**Chapter 4** provides background information about seed treatment with PPPs to demonstrate what this environment-friendly form of chemical crop protection exactly implies (**RQ5**). Due to its disadvantages related to the coating of the seed and its link to the overall decline of pollinators, the question is raised if the use of seed treatment products, other than neonicotionoids, also exert pressure (P) on pollinators (**RQ6**). This chapter verifies this research question by means of several experiments with treated seed (S). Uptake, translocation and persistence of PPPs in the plant after seed treatment are presented with special focus on the investigation of methiocarb residues in maize. Beeswax and bee bread from different hives located in Flanders are investigated as well.

# Abstract

The use of seed coated with fungicide or insecticide active substances is an effective way to control various pests and diseases in the cultivation of many crops and makes postemergence foliar treatments less necessary. Due to the targeted action, the technology should be seen as part of Integrated Pest Management. Seed treatment is considered to be an environmentally friendly form of chemical crop protection. The applied dose per hectare is limited to the seeds themselves, and is very low compared to leaf or soil spraying. Main disadvantages of this technique include the potential presence of systemic active substance residues in the guttation fluid, plant pollen and nectar of seed treated plants, and the potential emission of abraded seed particles to the environment during sowing. In the last few years, this emission has resulted in bee losses in several countries and contamination of surface water, among other things. The uptake, translocation and persistence in the plant vary greatly among various products. In this study, these three factors were examined for various crops, with special focus on the investigation of methiocarb residues in maize. First, degradation of methiocarb and its metabolites (methiocarb sulfone and methiocarb sulfoxide) in maize of which the seed was treated with Mesurol FS 500 was examined. The results of this greenhouse trial indicated that methiocarb is rapidly metabolised in the plant to its metabolites. Initially, the residue levels of methiocarb sulfone were 50 times smaller than those of methiocarb sulfoxide, but after a few weeks this difference was reduced to a factor of two. The translocation of the active substances from seed to the guttation drops was investigated as well. Shortly after emergence of the seedlings, guttation drops were collected daily and examined on the presence of methiocarb, methiocarb sulfone and methiocarb sulfoxide. Pollinators, such as honey bees, are assumed to consume these drops. Residue values of methiocarb were compared with LD<sub>50</sub>-values of the bee and a lethal dose was never registered in the guttation fluid during this study. Furthermore, a monitoring was performed at which the persistence and translocation of the seed coated active substances in various crops were investigated. The samples were mainly taken from mature crops. The spray records and the seed treatment were provided by the visited farmers. The following active substances used in the seed treatment of these crops were examined: fludioxonil, imazalil, prothioconazole, methiocarb, thiram and metalaxyl-M. Residues of fludioxonil, imazalil and methiocarb were found in mature plants, showing that these substances are persistent. Finally, residue analysis of beeswax and bee bread from different hives was performed. The bee bread contained little to no PPPs. Beeswax on the other hand, appeared to be more contaminated with several PPPs, i.e. especially fungicides but also some insecticides. The residue levels of the fungicide pyraclostrobin and the insecticide carbofuran found in beeswax can potentially harm honey bees. The results of this study illustrate that several active substances, used as seed treatment products, are translocated through the plant. However, not a single one of all detected substances in bee bread and beeswax is authorised for seed treatment in Belgium. Based on the obtained results, the risk of exposure to pollinators by means of seed treatment is supposed to be very low for the crops and active substances investigated in this study.

# Introduction

## **1.1 General introduction**

### 1.1.1 Purpose of seed treatment

Plant protection products are applied in agriculture although it is in fact a broader term, as PPPs are also used for non-agricultural purposes. They provide a higher yield by controlling diseases, pests and weeds (Oerke, 2006). However, PPPs can have a negative impact on human health and the environment (Xu et al., 2008). The use of PPPs can be limited by treating the seed. Seed treatment is mainly of interest for the young development stages of the plant as they are quite vulnerable to pathogens. The current seed treatments are subject to high safety and efficiency standards and guarantee a good control of pests and plant diseases. In addition, the environment is less exposed to active substances of PPPs by using a seed treatment compared to the use of foliar and soil products (Hairston, 2013). Figure 4-1 illustrates the differences in treated areas between spray applications, granules and seed treatment.

Furthermore, the use of seed treatment is also consistent with the concept of Integrated Pest Management (IPM), which makes it more attractive to farmers (FIS, 1999). According to IPM, pests are treated by means of an ecological approach using the knowledge of the biology of the pest. IPM is a well-reasoned use of products to control pests that ensures favourable economic and environmental effects. Plant protection products are still used in IPM but always weighted against other alternatives (Sandler, 2010).



Figure 4-1 Differences in PPP treated areas between spray applications (left), granules (centre) and seed treatment (right) (Bayer CropScience, 2012).

### 1.1.2 History and evolution of seed treatment

The history of seed treatments dates back to the times of the Egyptians and the Romans. At that time, the juice of onions was used to drench seeds which led to an antibacterial, -viral and -fungal activity. Further seed treatment development originated during the Middle Ages, like the use of liquid manure and chlorine salts. Salt water treatments were used from approximately 1650 because of its antibacterial effect (Abbasi and Lazarovits, 2006). A technique still used today is the hot water treatment. This method exists since 1765 at which the seed is placed in water of 45°C during two hours. This leads to a control of certain fungal pathogens that are located on the outer surface of the seed. Some other notable elements in the history of the seed treatment are the use and ban of arsenic (1740 to 1808) and mercury (1915 to 1982). Arsenic at low concentrations has a favourable effect on the germination and the growth of foliage and roots. The ban on the use of arsenic was a result of the high toxicity (Chun-xi et al., 2007). The use of mercury was put forward when it turned out it worked effectively against common bunt. However, the use of mercury as seed treatment did not increase because of both its high price and its toxicity (FIS, 1999; Mathre et al., 2001). The launch of the first systemic compound was in 1960. Until then, seed treatments had only been seed sterilants and did not move into the plant. During the 1970s, the first systemic seed treatment fungicides for airborne pathogens were introduced. Since the 1990s, crop protection and seed industries have developed and adopted new classes of fungicides, insecticides and nematicides, expanding pest control while reducing user and environmental impact. In 2005, the first seed treatment nematicide was introduced (FIS, 1999; Goggi, 2011).

Nowadays, various definitions of seed treatment are described in literature (TeKrony, 1976; The Ohio State University, 1988; Taylor and Harman, 1990; Paulsrud et al., 2001; Taylor et al., 2001; Nault et al., 2004; Hairston, 2013; ASTA, 2013a). Some definitions only focus on the chemical treatment while other definitions also include the physical and biological treatments. Some examples of complete definitions are:

"Seed treatments are physical, chemical or biological treatments applied to seeds or vegetative propagation materials to control disease organisms, insects or other pests" (Goggi, 2011).

"Seed treatment is the process of applying fungicidal and/or insecticidal seed dressing products onto various types of seed as a protective coating to create a 'protective zone' of active ingredient in the soil against soil-borne pathogens and insects" (Nuyttens et al., 2013).

### 1.1.3 Seed treatment types

Seed treatment is divided into coating and the physical treatment. The coating of seeds is further subdivided into seed pelleting, seed coating and seed dressing. During pelleting of the seed, inert materials are added to the seed to modify the seed size, shape and weight in order to sow accurately (Kaufman, 1991). A seed pellet consists of two main parts, i.e. the coating material and the binder. The coating material consists of only one component or of a mixture of several minerals and organic substances. The coating material adjusts the size, the shape and the weight of the seed. The binder ensures that the coating material is held together. During seed coating, the seed is placed in a jacket of various substances. This type of coating includes no minerals or organic substances in order to change the shape of the seed, but binders are used in order to adhere additives better to the seed (Hill, 1999). Seed dressing is a widely used technique in which the seed is treated with either solids or liquids.

Different substances are used in the coating process, i.e. PPPs, micro-organisms and hydroabsorbers. During seed pelleting, these substances are applied in the coating material by which direct contact with the seed is small. During seed coating and seed dressing, the substances come into direct contact with the seed and depending on the substances, the germination will be adversely affected. The used PPPs are mainly insecticides and fungicides. The use of fungicides has multiple goals (TeKrony, 1976), i.e.

- seed disinfection: the elimination of a pathogen which has penetrated into the seed and infected it, e.g. loose smut in wheat and barley;
- disinfestation of the seed: the control of spores and other pathogenic organisms which are located on the surface of the seed;
- seed protection: the chemical treatment to protect the seed and the young seedling from pathogenic organisms in the soil.



Figure 4-2 Cross section of a film-coated sugar beet fruit (left) and a pelleted plus film-coated sugar beet fruit (right) (Leubner, 2006).

Each fungicide may be used to achieve one or more of the above described goals. Plant death due to fungal attack can be significantly reduced as a result of seed treatment (Wheeler et al., 1997; Rothrock et al., 2012). Insecticides are commonly used to control or to reduce damage caused by infestation in the soil or during storage. Treatment with micro-organisms provides an improved transfer of nitrogen to the plant and fixation of N<sub>2</sub> (Taylor and Harman, 1990). Treatments with hydro-absorbers facilitate the water supply of the seed. A hydro-absorber saves a small amount of water and makes it available to the growing seedling (Berdahl and Barker, 1980).

Seeds can also undergo a physical treatment, for example the use of heat. Most heat treatments occur by means of water, water vapour and radiation which inactivate the seed bound pathogens. The temperature and duration of the treatment vary according to crop (Miller and Ivey, 2005). Finally, the seed can get a physiological treatment that stimulates the nutrient uptake and mobilisation of nutrients from the seed. Afifi et al. (2014) stated that the use of the neonicotinoid insecticide thiamethoxam has a positive effect on the physiological performance of the young maize plant, i.e. the insecticide improves the germination under stress conditions. The physiological substances from the treatments are added to the seed coating.

## 1.2 Seed coating technology

### 1.2.1 Products used for seed coating

Many products are available to coat seeds, each with its specific function. By using PPPs, each active substance used has its strengths and weaknesses, which often leads to using a mixture of active substances in the treatment process (Russell, 2005). These active substances are divided into two major groups, i.e. the contact and systemic active substances. The effect of contact active substances is local and they do not penetrate into plant. Systemic active substances penetrate into the roots and the germinating seed after which translocation to the stem and leaves takes place. The translocation rate varies depending on the crop and the systemic activity of the active substances (Paulsrud et al., 2001). The active substances authorised as seed treatment products in Belgium are described in Table 4-1. Beside PPPs, additives are also often added to the seeds. Various additives are available on the market and are used in the seed treatment process (Paulsrud et al., 2001), i.e.

- dyes or colorants are added in order to mark the treated seed and to prevent mixing with food grain;
- carriers, stickers and binders are added in order to increase the adherence of the PPP to the seed and to prevent dusting off. These substances tend to have a neutral pH, are non-toxic to humans and cause no apparent damage to the germination of the seed;
- anti-foaming agents suppress the formation of foam;
- lubricants reduce the friction of seed flow through the planter, e.g. graphite or talc;
- micronutrients are added in order to introduce trace elements required for nodulation (soybean (*Glycine max*) seed treatment), e.g. molybdenum.

Table 4-1 Active substances authorised as seed treatment product in Belgium with information according to PPP group (insecticide = In, fungicide = Fu), systemicity (systemic = S, non-systemic = NS, local systemic = LS), pest or disease they control and crops on which the use is authorised (BCPC, 2006; Lewis and Green, 2011; Fytoweb, 2015).

Active substance	PPP group	Systemicity	Activity against	Сгор
cymoxanil	Fu	LS	Phytophtora infestans	beans, lupine, peas
difenoconazole	Fu	S	loose smut and at high label rates against foliar	winter and spring barley, winter and spring oats,
			diseases (e.g. rust)	winter and spring rye, spelt, (winter and spring)
				triticale, (winter and spring) wheat
fludioxonil	Fu	NS	Aspergillus, Fusarium, Penicillium, Rhizoctonia and	(winter and spring) barley, beans, black radish,
			seed-borne wheat scab	cabbage, carrots, Chinese and knob celery, fennel,
				lupine, maize, (winter and spring) oats, onions,
				parsley, peas, radish, root parsley, (winter and
				spring) rye, shallots, spelt, sunflower, (winter and
				spring) triticale, (winter and spring) wheat
fluoxastrobin	Fu	S	fire blight	rye, spelt, triticale, wheat
hymexazol	Fu	S	Pleospora betae and soil-borne fungi	beet
ipconazole	Fu	S	common bunt, Fusarium, loose smut and Pyrenophora	winter and spring barley, oats, rye, spelt, triticale,
			teres	wheat
iprodione	Fu	S	Alternaria, grey mould, Phoma betae	beet, cabbage
metalaxyl-M	Fu	S	Phytophthora and Pythium	beans, lupine, maize, peas
prochloraz	Fu	NS	Pyrenophora teres and Septoria nodorum	barley, flax, oats, rye, spelt, triticale, wheat
prothioconazole	Fu	S	aphids, common bunt, Fusarium and loose smut	winter and spring barley, (winter) oats, (winter) rye,
				spelt, (winter) triticale, (winter) wheat
Pseudomonas chlororaphis	Fu	-	common bunt, Fusarium and Septoria nodorum	rye, triticale, wheat

Active substance	PPP group	Systemicity	Activity against	Сгор
silthiofam	Fu	-	take-all	barley, spelt, triticale, wheat
thiram	Fu	NS	broad spectrum of fungi	beans, beet, chicory, maize, peas
triticonazole	Fu	S	common bunt and loose smut	barley, oats, rye, spelt, triticale, wheat
ziram	Fu	NS	fire blight and scab	maize
beta-cyfluthrin	In	NS	insects of the Coleoptera, Hemiptera and Lepidoptera	beet (export), chicory
clothianidin	In	S	aphids, beet flea beetles, beet leafminers, pygmy	winter barley, beet (export), chicory, winter oats,
			beetles and wireworms	winter rye, spelt, winter triticale, winter wheat
cypermethrin	In	NS	wheat bulb flies and wireworms	winter and spring barley, oats, rye, spelt, triticale,
				wheat
fipronil	In	S	onion fly and thrips	leek, onions, shallots
imidacloprid	In	S	aphids, beet flea beetles, beet leafminers, milli- and	winter barley, beet (export), endive, lettuce, winter
			centipedes, pygmy beetles, thrips and wireworms	oats, winter rye, spelt, triticale, winter wheat
magnesium phosphide	In	-	storage mites and other storage pests	seeds
methiocarb	In	NS	Oscinella frit, pygmy beetles and damage by birds	beet for export, maize
tefluthrin	In	NS	garden centipedes, milli- and centipedes, pygmy	barley, beet, oats, rye, spelt, triticale, wheat
			beetles, wheat bulb flies and wireworms	
thiamethoxam	In	S	aphids, beet flea beetles, beet leafminers, milli- and	beet, cabbage, carrots, endive, lettuce
			centipedes, pygmy beetles, spring tails, thrips and	
			wireworms	

### **1.2.2** Application equipment

PPPs used in seed treatment can be applied to the seed in various forms, i.e. as dry powder, slurry or liquid. Depending upon the PPP formulation, a particular machine is used. Commercial seed treatment machines are designed to add a certain amount of PPP to a given weight of seed. In order to achieve this, two major elements are distinguished in each machine. The first element is a measuring system that tracks the added amount of seed and PPP. The second element is the mixing chamber where seed and PPP are mixed (Paulsrud et al., 2001).

## 1.2.2.1 Dust treater

This machine is used as the PPP is present in the form of a dry powder. The powder is mechanically mixed with the seed without any addition of water. Disadvantages of this treatment are the difficulty of achieving a uniform distribution on the seed and the easy loss of powder on the seed. Nowadays, this machine is no longer used in commercial seed treatment companies, mainly due to problems with dust formation and seed treatment quality (TeKrony, 1976; Paulsrud et al., 2001).

## 1.2.2.2 Metered-slurry treater

This machine uses slurries. Slurries are a suspension of water and water-insoluble substances. For example, if the PPP is an insoluble solid form, it is mixed with water to obtain a slurry. This application system ensures a good and thorough treatment of the seed. The dust problem (1.2.2.1) does not arise here. In addition, a more uniform distribution of the PPP to the seed is obtained by using this kind of treater (TeKrony, 1976; Paulsrud et al., 2001).

# 1.2.2.3 Direct treater

A liquid formulation of the PPP is treated with a direct treater. The PPP may be diluted in water before applying it to the seed. The use of this type of treaters ensures an even better and more uniform coating of the PPP to the seed compared to the previously described treaters. Some examples are the 'Panogen Seed Treater' and 'Mist-O-Matic Seed Treater'. The PPP is added as a liquid to the seed in the 'Panogen Seed Treater'. In the 'Mist-O-Matic Seed Treater', the liquid is first atomized before it comes into contact with the seed (TeKrony, 1976; Paulsrud et al., 2001).

# 1.2.3 Seed treatment quality

Various seed treatment methods may differ in quality. The quality of the treatment depends on the composition of the treatment mixture, the rate at which the mixture is applied to the seed, the conditions in which the treatment is carried out (seed temperature, product temperature, etc.) and the equipment used. These factors should be taken into account in order to obtain high quality treated seed in a safe and effective manner. The quality of the seed treatment plays an important role (ASTA, 2013b). In Europe, a quality assurance system is developed for treated seeds, i.e. the European Seed Treatment Assurance (ESTA). ESTA combines a number of elements to ensure professional and high quality treated seed. In order to indicate that a company produces high quality seed, the company can use the logo of ESTA. Only companies certified by the ESTA are allowed to use this logo. This logo facilitates the free movement of treated seed through the European Union and indicates the monitoring of seed quality standards (ESTA, 2015).

### 1.3 Role of seed treatment at global and regional scale

Seed treatments are used worldwide, however the interest on the market varies from region to region. Table 4-2 shows the world seed treatment by region based on estimates of 1997. Geographically, the use of seed treatment was concentrated in Western Europe and the North American Free Trade Agreement (NAFTA) countries, which together accounted for 63% of the world market. In Asia, Africa and Middle East seed treatments have been substantially underutilised (Agrow Reports, 1999; Stevens, 2002). In 2007, the European Seed Association (ESA) reported that the global seed treatment sector had a value of 1.1 billion euros in 2005. Western Europe represented about 36% of the world total, with North America accounting for 22%, Latin America for 16%, and Asia for 3%. An annual growth of 5% for the global seed treatment sector was predicted by ESA. Europe is, however, expected to show only limited further growth, well below the worldwide rate. European legislation on seed treatments makes it difficult to maintain existing authorisations and obtain new ones. As a consequence, companies are reducing their investment and innovation is slowing down. Finally, ESA also reported that approximately 95% of all seed sown in Europe is treated with one or more seed treatment products (ESA, 2007). More recent data show that the United States currently are the leading seed treatment market, accounting for more than 50% of the studied global markets. Further, Brazil represents about 15% of the world total due to the rapidly growing market for treated soybeans and maize. Compared to 1997 (Table 4-2), the seed treatment sector is increasing in Latin America. Also Argentina, another Latin American country, has a growing seed treatment market especially due to a sharp increase in the use of treated maize (Little Falls, 2010).

Table 4-2 World seed treatment market (%) by region based on 1997 estimates (Agrow Reports, 1999).

Region	Percentage of world market (%)
Western Europe	38
North American Free Trade Agreement Countries (NAFTA)	25
Eastern Europe	19
Latin America	12
Asia	3
Africa, Middle East	3

Fungicide and insecticide seed treatment market by crop is illustrated in Table 4-3. Cereals accounted for 63% of fungicidal treatments and 22% of insecticidal treatments. More than one third of insecticidal seed treatments worldwide were applied to maize only (Agrow Reports, 1999; Stevens, 2002). Nowadays, the amount of insecticide treatments in maize decreases because of the actual problems of systemic insecticides, especially neonicotinoids. Neonicotinoids are a class of neuro-active insecticides chemically similar to nicotine. Used as PPPs or biocides, neonicotinoids are translocated throughout the plant. Due to their systemic activity, these products are used to control insects. The frequent use of neonicotinoids has revealed some problems. Neonicotinoids also end up in pollen, nectar and guttation fluids through which pollinators, e.g. bees, can be exposed to these substances (CTGB, 2015). Since 2013, the use of neonicotinoids (clothianidin, imidacloprid and thiamethoxam) is limited in Belgium. Treated seeds of crops that are not grown in greenhouses may no longer be sown (Fytoweb, 2013).

Table 4-3 Fungicide and insecticide seed treatment market by crop (% of total), data from 1996 (Agrow Reports, 1999).

Сгор	Fungicides (%)	Insecticides (%)
cereals	63	22
canola	-	15
potatoes	8	-
sugar beet	-	14
rice	8	-
cotton	6	12
maize	5	34
vegetables	5	-
other	5	3

The world seed treatment market estimates described in the previous section were based on monetary value. In order to estimate the role of seed treatment in relation to the total use of PPPs in Belgium, the exact amount of PPPs used in seed treatment should be determined. Up to 2012, seed treatment was not included in the determination of Belgian PPP use. In **Chapter 2**, the amount of PPPs used in seed treatment is calculated by using various assumptions since no figures are publicly available in Belgium (2.3.3). The use of PPPs in seed treatment decreased from 0.07 million kg a.s. in 2009 to 0.05 million kg a.s. in 2012 representing 1.3% and 1.1% of the total PPP usage estimates in Belgium respectively. Fungicidal treatments accounted for an estimated 55% of total PPP use in seed treatment in 2009 and increased to 60% in the period 2010-2011. In 2012, insecticidal treatments were higher than fungicidal treatments (55% and 45% respectively).

## 1.4 Seed treatment issues

### 1.4.1 Introduction

Seed treatment is a widespread and effective way to control various pests and diseases using smaller doses with potentially less harmful side-effects. This technology also makes it possible to combine various applications into only one sowing procedure, helping to reduce the use of fuel and risks of soil compression and erosion and assisting low-intensity farming practices and an IPM policy (Ahmed et al., 2001; Koch et al., 2005; Nuyttens et al., 2013). The use of treated seeds also has some disadvantages which are all related to the coating of the seed. Workers who produce or apply the seed treatments can be accidentally exposed to the used substances. Besides, contamination of the food supply can occur by accidental mixing of treated seed with food and also contamination of the environment by emission of abraded seed particles during sowing can occur (Girolami et al., 2009; Nikolakis et al., 2009; Pistorius et al., 2009; Tapparo et al., 2011; Nuyttens et al., 2013). Birds may also eat treated seed, which poses a risk to their health (Vercruysse and Steurbaut, 2002). In the field, pollinators can be exposed to PPPs by various routes, i.e. by contact with residues in the guttation fluid, nectar and pollen. Finally, drift of dust from treated seeds is also one of the potential exposure routes for bees. Several incidents of bee poisoning have recently occurred that were caused by dust from abraded particles of the seed treatment containing bee-toxic products (Girolami et al., 2009; Nikolakis et al., 2009; Pistorius et al., 2009; Reetz et al., 2011; Tapparo et al., 2011; Nuyttens et al., 2013).

Seed treatment is linked to the overall decline of pollinators and the well-known phenomenon of colony collapse disorder (CCD). Pollinators, such as honey bees, come into contact through the above mentioned exposure routes with different PPPs that have been applied to the seed. Neonicotinoids are often used in seed treatment. The effects of this group are consistent with the symptoms of CCD (Suchail et al., 2000; Maus et al., 2003; Cox-Foster et al., 2007):

- rapid loss of its adult bee population and a considerable capped brood and food reserves;
- little or no dead adult bees are found inside the hive or in close proximity to the colony;
- delayed invasion of pests in the hive (e.g. small hive beetles, wax moths, etc.).

The International Commission for Plant-Pollinator Relations (ICPPR) promotes and coordinates research on the relationships between plants and pollinators of all types. This research includes studies of insect pollinated plants, pollinator foraging behaviour, effects of pollinator visits on plants, management and protection of insect pollinators, bee collected materials (e.g. nectar and pollen), and products derived from plants and modified by bees. Especially the Bee Protection Group is the ICPPR's most active working group and has provided leadership for the European Plant Protection Organization's (EPPO) concerns for

pollinators and pollination, and for the ICPPR as a world-wide body. This group brings together experts from the crop protection industry, national regulatory authorities, university research departments, and more recently from beekeeping organisations, to discuss the development of testing methodology as well as the overall assessment of risk to bees from the use of PPPs. As part of their ongoing review of PPP risk assessment for honey bees, a number of issues that require further consideration were identified. Working groups were set up to address the recently emerged problems of systemic effects through seed and soil treatments, of field and semi-field testing, and honey bee brood testing. Concerning honey bees, four working groups focus on following topics: the development of testing methods in semi-field and field (including modelling aspects), risk assessment related to dust and guttation drops (Alix, 2015).

#### 1.4.2 Residues in guttation fluids

Guttation is the phenomenon of water being released from the leaves through pores (hydathodes) or through the ends of the veins on the edge of the leaf (Figure 4-3). An increased root pressure and a decreased transpiration cause this phenomenon. Guttation occurs especially during the night and in the morning several hours after sunrise (Klepper and Kaufmann, 1966). Shawki et al. (2005) stated that honey bees need a lot more water during early spring. During this period, foraging bees visit plants not to collect nectar or pollen but to collect dew and guttation fluids. Because of this, bees can come into contact with systemic active substances. Girolami et al. (2009) investigated the presence of systemic insecticides and more specifically of neonicotinoids in guttation fluids. Research was especially conducted on maize and revealed that the excretion of guttation fluids is limited to the first three weeks after germination (Girolami et al., 2009; Thompson, 2010). During this period, residue levels in the guttation fluids can be quite high. However, active substances residues vary among different environmental conditions. The residue levels of the active substances in the guttation drops will be higher in conditions of lower soil moisture and dryer air as a consequence of the progressive water evaporation (Girolami et al., 2009).



Figure 4-3 Guttation drops on maize leaves in the field (Girolami et al., 2009).

In the study of Girolami et al. (2009), maize seeds were coated with the product Gaucho 350 FS (350 g/l imidacloprid, suspension concentrate for seed treatment (FS), Bayer CropScience, 0.5 mg imidacloprid/seed). Guttation drops collected on plants from neonicotinoid coated seeds contained an imidacloprid concentration of 47 (± 9.9) mg/l. Seeds coated with clothianidin (600 g/l clothianidin, Poncho 600 FS, Bayer CropScience AG Leverkusen Germany, 1.25 mg clothianidin/seed) and thiamethoxam (350 g/l thiamethoxam, Cruiser 350 FS, Syngenta International AG Basel Switzerland, 1 mg thiamethoxam/seed) were also examined. The guttation drops contained 23.3 (± 4.2) mg/l clothianidin and 11.9 (± 3.32) mg/l thiamethoxam. Despite the lower dose of imidacloprid applied on the seeds, the concentration found in the guttation drops was significantly higher compared to the other two neonicotinoids. This indicates a higher translocation from seed to guttation for imidacloprid. Tapparo et al. (2011) observed the same phenomenon. When these guttation drops were consumed by honey bees, irreversible wing paralysis appeared in a time ranging between two and nine minutes from consumption followed by death. The paralysis occured slightly faster at guttation drops contaminated with thiamethoxam as this neonicotinoid is more toxic than imidacloprid for honey bees although it was less concentrated in guttation drops (Girolami et al., 2009).

### 1.4.3 Residues in pollen and nectar

Bees live from nectar and pollen. Pollen are a source of proteins which are necessary for the growth and development of the honey bees. Nectar is a source of carbohydrates which

serves as an energy source for the bee (Rortais et al., 2005). Neonicotinoids are systemic active substances which are transported through the plant. Because of the systemic nature of these active substances, translocation to different plant parts and also to the pollen and nectar can occur. In Schmuck et al. (2001), the translocation of imidacloprid, used as seed coating, in sunflowers (Helianthus annuus) was investigated. Under laboratory conditions at which seeds were treated with 0.7 mg imidacloprid/seed, residues of 3.9 ( $\pm$  1.0)  $\mu$ g/kg and 1.9 ( $\pm$  1.0)  $\mu$ g/kg were found in the pollen and nectar respectively. Furthermore, no residues of imidacloprid or its metabolites were indicated in the nectar and pollen when performing a field study in which 1 mg imidacloprid/seed was used. The discrepancy between results of laboratory and field experiments show that ambient conditions should always be taken into account (Schmuck et al., 2001). Bonmatin et al. (2003) analysed samples of pollen obtained from maize and sunflowers both under field conditions. However, 58% of the sunflower pollens contained imidacloprid at levels from 1 to 11  $\mu$ g/kg, with a mean value of 3  $\mu$ g/kg. 80% of the maize samples contained an average imidacloprid concentration of 2  $\mu$ g/kg. A follow-up of this study also indicated residues in the pollen of maize and this with an average concentration of 3  $\mu$ g/kg (Charvet et al., 2004).

#### 1.4.4 Dust drift from coated seed

During sowing of crops, abraded seed particles can be emitted to the environment. Bees can come into direct contact with these particles when flying through the drift cloud originating from the seed drill (Biocca et al., 2011; Nuyttens et al, 2013). Another way of exposure is by indirect contact when bees walk on contaminated leaves of the vegetation next to drilled fields (Schnier et al., 2003). Since the possibility of dust formation is largely determined by the seed treatment, the Heubach method was developed which allows to verify the abrasion potential of seed coatings. This method is used as a reproducible measuring technique for seed treatment quality and includes the so-called Heubach dust meter. This device consists of a rotating drum where the to be examined product is placed. Dust particles are released due to rotation, further transported by a calibrated air flow and collected on a filter. Finally, the collected dust particles on the filter are quantified. The more particles are retained on the filter, the higher the abrasion potential of the product (Delft Solids Solutions BV, 2009; ESA, 2011; Nuyttens et al., 2013). In the study of Girolami et al. (2012), bees were captured in the vicinity of working seed drilling machines. The used seed was treated with different seed treatment products, including Poncho 600 FS (600 g/l clothianidin, FS, Bayer CropScience AG Leverkusen Germany, 1.25 mg clothianidin/seed) and Gaucho 350 FS (350 g/l imidacloprid, FS, Bayer CropScience, 0.5 mg imidacloprid/seed). The collected bees contained 500 ng active substance/bee after sowing. This amount is potentially lethal as the LD<sub>50</sub> for clothianidin is 21.8 ng/bee and for imidacloprid 17.9 ng/bee. Given the water solubility of neonicotinoids, the effect of humidity on mortality after exposure to dust drift was also investigated. High humidity itself does not cause mortality, but it seems to have a synergistic influence on the toxicity of insecticides that come into contact with honey bees resulting in a faster and higher mortality rate (Marzaro et al., 2011).

# 1.4.5 Toxicity of seed treatment for the honey bee (Apis mellifera)

Residues of active substances the bee can come into contact with after seed treatment were shown in previous paragraphs. To make further interpretations, toxicity values were looked up. Table 4-4 shows LD<sub>50</sub>-values which were obtained from different studies. If the LD<sub>50</sub>-values from Table 4-4 are compared to the exposure via guttation fluids (1.4.2), a lethal exposure is noticed. Girolami et al. (2009) assumed that a bee absorbs 5  $\mu$ l of fluid during drinking. After recalculating the exposure of the bee ( $\mu$ g/bee), the bee is exposed to 0.060  $\mu$ g thiamethoxam, 0.115  $\mu$ g clothianidin and 0.235  $\mu$ g imidacloprid. These calculated values were compared to the acute oral exposure LD<sub>50</sub>-values shown in Table 4-4, 0.005, 0.003 and 0.05  $\mu$ g/bee respectively. This comparison indicates that in the study of Girolami et al. (2009) all neonicotinoids were present in lethal concentrations in the guttation fluids. Exposure to dust drift (1.4.4) was multiple times higher than the LD<sub>50</sub>-values for acute contact exposure and therefore also lethal to bees.

Neonicotinoid	Exposure	LD <sub>50</sub> (µg/bee)	References
clothianidin	Contact (acute)	24h: 0.022	Iwasa et al. (2004)
	Contact (acute)	48h: 0.044	Decourtye et al. (2010)
	Oral (acute)	48h: 0.003	Decourtye et al. (2010)
imidacloprid	Contact (acute)	24h: 0.0179	Iwasa et al. (2004)
	Oral (acute)	48h: 0.070	Suchail et al. (2001)
	Oral (acute)	96h: 0.050	Suchail et al. (2001)
thiamethoxam	Contact (acute)	24h: 0.030	Iwasa et al. (2004)
	Contact (acute)	48h: 0.024	Decourtye et al. (2010)
	Oral (acute)	48h: 0.005	Decourtye et al. (2010)

Table 4-4 Toxicity va	alues of neonicotinoids	(µg/bee) for the ho	ney bee (Apis mellifera).
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 $LD_{50}$  = Lethal Dose at which 50% of the test species cause mortality in a single dose.

### 1.5 Objectives

During this study, residue analysis was carried out to investigate the uptake, translocation and persistence of PPPs in the plant after seed treatment (Figure 4-4). Systemic PPP residues can be present in the guttation fluid, plant pollen and nectar of seed treated plants (1.4). However, the available residue data in nectar and pollen are limited. In all cases where data were available, residues in pollen and nectar were less than 0.1 mg a.s./kg suggesting that translocation of PPPs to fruiting structures is measurably less effective than to other plant parts. Therefore, residue analysis of pollen and nectar was not included in this thesis. First, degradation of methiocarb in maize of which the seed was treated with Mesurol FS 500 was examined in the greenhouse trial. The translocation of the active substance from seed to the guttation drops was investigated as well. Furthermore, a monitoring was performed at which the persistence and translocation of the active substances in the coated seed of various crops were investigated. Finally, residue analysis of beeswax and bee bread from different hives was performed.



Figure 4-4 Objectives of this study.

# **Materials and Methods**

# 2.1 Experimental set-up

## 2.1.1 Greenhouse trial

Various studies already illustrated the issue of neonicotinoids and translocation of these substances through the plant. Furthermore, the use of neonicotinoids is limited in Belgium since 2013. Non-professional use of neonicotinoids is no longer authorised in Belgium. Professional users may no longer use these insecticides on crops that are in bloom and on certain cereals. Furthermore, treated seeds with neonicotinoids may no longer be sown except if these crops are grown in greenhouses and if cereals are sown between July and December (Fytoweb, 2013). In this study, the focus was on a substance that does not belong to the group of neonicotinoids, i.e. methiocarb better known as Mesurol (commercial name of the product). Methiocarb belongs to the group of the carbamates and is used as an insecticide, molluscicide, acaricide and bird repellent. Carbamates provide a reversible inhibition of acetylcholinesterase which explains the neurotoxic contact and stomach action of methiocarb. It can even cause acute toxicity in humans if exposed for long periods of time or to a sufficient dose. Methiocarb is also a known poison to water organisms (BCPC, 2006; Lewis and Green, 2011).

# 2.1.1.1 PPP residue trial during growth of maize

In this greenhouse experiment, maize was grown and samples were taken at various development stages of the plant in order to determine the residues of seed treatment products (Figure 4-5). The seeds of maize were treated with Mesurol FS 500 (500 g/l

methiocarb, FS, Bayer CropScience, 1 l product/100 kg seeds) and TMTD 98% (98% thiram, WP, Bayer CropScience, 36 g product/50000 seeds). Thiram is a non-systemic active substance which belongs to the group of dithiocarbamates. Dithiocarbamates are not stable and cannot be easily extracted or analysed directly. Contact with acidic plant juices rapidly degrades dithiocarbamates, decomposing them into CS<sub>2</sub> and the respective amine (Dasgupta et al., 2012). Because of this, the focus was on methiocarb. Although methiocarb is also a non-systemic substance, it has a few degradation products (methiocarb sulfone and methiocarb sulfoxide) which are well transported in the plant (EFSA, 2006). Two different maize varieties were sown in the greenhouse, i.e. Telexx (Telexx FAO 200, Philip-Seeds) and Ronaldinio (KWS Benelux B.V.). Taking into account the 1000 kernel weight of Telexx maize (286 g), the applied ratio of methiocarb was 1.43 mg/seed. The 1000 kernel weight of Ronaldinio maize was 311 g resulting in an applied ratio of 1.56 mg methiocarb/seed.

Maize is normally sown under field conditions at 12-16 cm distance from each other in the row (depending on the desired plant densities) and at 75 cm from each other between rows (KWS Benelux B.V., 2014; Wageningen UR Livestock Research, 2015). In this study, the distance between plants was 15 cm in the row and 40 cm between rows due to the restricted test area of the greenhouse. Seeds were sown at a depth of 4 to 5 cm in a sandy loam soil. Temperature and daylight hours in the greenhouse were kept at 28-30°C and 12-13 hours respectively. During sampling, entire plants were taken at random from the test area in the greenhouse and placed in resealable bags. A minimum weight (± 50 g) was required to perform the analysis which meant that the number of sampled plants varied between the development stage. Sampling was carried out once a week and the analysis took place on the same day.

The first sampling was performed when the plants were located between the  $3^{rd}$  and  $4^{th}$  leaf stage, the second sampling between the  $4^{th}$  and  $5^{th}$  leaf stage, the third sampling at the  $6^{th}$  leaf stage, and the fourth and last sampling at the  $7^{th}$  leaf stage.



Figure 4-5 Germinating maize in the greenhouse.

Seed treatment

#### 2.1.1.2 Guttation trial

A guttation trial was performed in the greenhouse in order to check the amount of active substance in the guttation drops derived from maize seed (Figure 4-6). The maize variety Telexx was used in this trial and contained the same active substances as the seed described in the previous trial (2.1.1.1), i.e. methiocarb and thiram. Coated seeds of Telexx maize were sown in pots with a diameter of 15 cm (36 pots in total). Temperature and daylight hours in the greenhouse were kept at 28-30°C and 12-13 hours respectively. Guttation drops were collected at random from various plants as soon as the phenomenon occurred, which was usually a few days after germination. The collection of the droplets occurred daily from 8 to 9 a.m. using disposable glass Pasteur pipettes. The droplets were transferred in a vial and then placed in the freezer at  $-21^{\circ}$ C in order to allow analysis of all samples on the same day. The guttation drops were not extracted, but injected directly into the LCMS-MS, whether or not after diluting ten times. The guttation drops were examined on the presence of methiocarb and its metabolites.



Figure 4-6 Experimental set-up of the guttation trial (left) and maize plant with some guttation drops (right).

### 2.1.2 Monitoring

All samples of brown bean (*Phaseolus vulgaris*), chicory, maize, onion seed, rye, spelt, spring and winter barley, sugar beet, triticale and winter wheat were taken at random over the field. At least three entire plants were sampled per field and placed in resealable freezer bags. All samples were cooled after collection and stored at -21°C before residue analysis. Spray records were provided by the farmers. Based on information from the spray records, an indication was given for the persistence of the seed treatment product. Spray records were also useful in order to highlight a potential overlap between the active substances of the spray application and of the seed treatment. Spray records of brown bean, chicory, spring and winter barley, sugar beet, triticale and winter wheat are listed in Appendix B: Spray records of various crops. Seed treatment products without corresponding application date were obtained for the crops maize, onion seed, rye and spelt and are listed in Appendix B as well. The plots of winter wheat and winter barley at which a second sampling was taken, were untreated. Spring barley was treated with Fungazil mlf 50 which is not authorised as seed treatment product in Belgium. The used seeds were derived from the Netherlands and were certified by the Dutch General Inspection Service (NAK). NAK Services tests seed potatoes and seed from cereals and grasses from all over the world. Table 4-5 shows the surveyed crops, development stage of the plant and seed treatment product. Substances of the seed treatment product, formulation, supplier and application ratio are listed as well in Table 4-5.

Table 4-5 Surveyed crops with corresponding development stage of the plant (BBCH-stage) during sampling, used seed treatment product with its formulation, supplier, concentration of active substance (a.s.) and application ratio (g a.s./100 kg seed).

Crop	BBCH-	Product	Formulation	Supplier	Concentration	Application ratio
	stage				of a.s. (g	a.s./100 kg seed)
brown bean	85-89	Apron XL <sup>a</sup>	ES	Syngenta	339.2 g/l metalaxyl-M	13.6
		TMTD 98% <sup>b</sup>	WP	Bayer CropScience	98% thiram	196
chicory	85-89	Flowsan Ultra	i FS	Taminco	485 g/l thiram	1.2 <sup>d</sup>
maize	85-89	Mesurol FS 50	00 FS	Bayer CropScience	500 g/l methiocarb	500
		Flowsan FS	FS	Taminco	530 g/l thiram	63.6 <sup>d</sup>
onion seed	81-89	Mundial	FS	BASF Belgium	500 g/l fipronil	10 <sup>d</sup>
		Apron XL <sup>a</sup>	ES	Syngenta	339.2 g/l metalaxyl-M	17
		TMTD 98% <sup>b</sup>	WP	Bayer CropScience	98% thiram	392
rye	85-89	Celest	FS	Syngenta Crop Protection	500 g/l fludioxonil	100
spelt	85-89	Redigo	FS	Bayer CropScience	100 g/l prothioconazol	e 10
spring barley	85-89	Fungazil mlf 5	50 <sup>c</sup> LI	AgroDan	50 g/l imazalil	5
sugar beet	85-89	Poncho beta	FS	Bayer CropScience	53.3 g/l beta-cyfluthrin	ı 8 <sup>d</sup>
					400 g/l clothianidin	60 <sup>d</sup>
		Flowsan Ultra	i FS	Taminco	485 g/l thiram	6 <sup>d</sup>
triticale	85-89	Redigo	FS	Bayer CropScience	100 g/l prothioconazol	e 10
winter barley	85-89	Redigo	FS	Bayer CropScience	100 g/l prothioconazol	e 10
		Argento	FS	Bayer CropScience	50 g/l prothioconazole	10
					250 g/l clothianidin	50
	14-16	Redigo	FS	Bayer CropScience	100 g/l prothioconazol	e 10
		Argento	FS	Bayer CropScience	50 g/l prothioconazole	10
					250 g/l clothianidin	50
winter wheat	85-89	Celest	FS	Syngenta Crop Protection	500 g/l fludioxonil	100
	14-16	Celest	FS	Syngenta Crop Protection	500 g/l fludioxonil	100

BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale); ES = emulsion for seed treatment; FS = suspension concentrate for seed treatment; LI = liquid; WP = wettable powder; <sup>a</sup>(Syngenta, 2013); <sup>b</sup>(SATEC, 2011); <sup>c</sup>(JMPR, 1977); <sup>d</sup>g a.s./100 000 seeds.

### 2.1.3 Beeswax and bee bread trial

Beeswax and bee bread of five different hives at different locations were examined for the presence on various PPPs. Beeswax is a natural wax produced by honey bees of the genus *Apis*. The wax is formed into scales by eight wax-producing glands in the abdominal

segments of four through seven of worker bees, who discard it in or at the hive. The hive workers collect and use it for comb structural stability, to form cells for honey storage and larval and pupal comfort and protection within the beehive. Chemically, beeswax mainly consists of esters of fatty acids and various long-chain alcohols (Brown, 1981). Studying beeswax is particularly interesting because most of the PPPs and acaricides are fat soluble, non-volatile and persistent and so easily accumulate herein. These chemicals also resist the wax melting temperature. Therefore, they can accumulate for decades as it is a common beekeeping practice to recycle wax almost continuously in the form of foundations on which bees will construct a complete comb (Ravoet et al., 2015). Bee pollen is the pollen ball that has been packed by worker bees into pellets. Bee bread consists of pollen that has been stored by the bees in the cells of the honeycomb. This is wetted with glandular secretion and covered by a layer of honey. It is a source of protein, fats, microelements and vitamins for the bees (Bogdanov, 2015).

The first three hives (1, 2, 3) were located in the city of Roeselare. The fourth and fifth hive (4, 5) were located in the countryside of Moorslede. Crops around these hives were carrots, leek, cauliflower (Brassica oleracea var. botrytis subvar. cauliflora) and maize. One honeycomb was taken at random from each hive and cooled after collection. In order to investigate if the presence of PPP residues affects the honey bee colony health, an additional important note is that no living bees were present in the hives 1, 2 and 4. Besides the honeycombs from different hives, two other samples of beeswax were also examined. These samples were already melted into different beeswax plates. One sample was bought in a store and was a collection of beeswax from different honeycombs, which was melted into a beeswax plate. The other sample contained beeswax collected over a period of ten years (of hives in Moorslede), which was also melted into one beeswax plate. Three randomly selected samples of food-free beeswax (25 cm<sup>2</sup>) were collected from each of the seven beeswax samples by using disposable spatulas and placed in resealable freezer bags. All beeswax samples were stored at -21°C before residue analysis. Subsequently, three samples of bee bread (± 5 g) were taken at random from each of the five honeycombs by using disposable spatulas. These samples were placed in resealable plastic tubes and stored at -21°C before residue analysis.

#### 2.2 Extraction methods

A description of the persistence and translocation of PPPs in plants after seed treatment was made by determining the residue levels on different crops and in different plant development stages. The analysis included an extraction of the active substances from the plant material followed by a quantitative determination of the residue level. Recovery values were determined as well. These values demonstrate the efficiency of the extraction method and should be close to 100%. A known amount of the product is added to a sample that undergoes the entire exctraction procedure. The amount of the residue level found in the

sample is then compared with the original amount added to the sample in order to calculate the efficiency (%) of the method.

# 2.2.1 QuEChERS method

Fludioxonil, imazalil, prothioconazole, metalaxyl-M, methiocarb, methiocarb sulfone and methiocarb sulfoxide were extracted by using the QuEChERS method. QuEChERS stands for Quick, Easy, Cheap, Effective, Rugged and Safe, all kind of properties that describe this sample preparation method. This method is often used in the PPP residue analysis from plant resources which have a high water content. Low water content of the plant material can be corrected by adding water to the commodity. Further, PPPs dissolve again after which the extraction can take place (Kowalski and Cochran, 2015).

The QuEChERS method consisted of several steps. First, 10 g of plant material was weighed into a centrifuge tube of 50 ml. If the water content of the plant material was too low, only a few grams of plant material were weighed and the tube was supplemented with water to obtain 10 g. Second, 15 ml of acetonitrile was added to the centrifuge tubes whether or not spiking these first with 1 mg/l active substance. Third, the tubes were manually shaken for one minute. Next, salts were added (6 g MgSO<sub>4</sub> and 1.5 g NaOAc) and the tubes were shaken for five minutes. Subsequently, the tubes were centrifuged for five minutes at 10000 rpm. Finally, the upper liquid layer present in the tube was transferred to an autosampler vial (Agilent Technologies, 2013).

If sufficient plant material was present, only the underground plant biomass was included in the sample preparation for the non-systemic substance of fludioxonil. This was due to the fact that no translocation of non-systemic active substances is expected in the plant. Prothioconazole, imazalil and metalaxyl-M are systemic substances through which the entire plant was included in the sample preparation. The non-systemic methiocarb was also treated as a systemic active substance as it belongs to the group of the carbamates. Certain substances of this group have some systemic properties. Earlier analysis already showed that metabolites of methiocarb (methiocarb sulfone and methiocarb sulfoxide) are transported in the plant (EFSA, 2006). If sufficient plant material was present, a distinction was made between the different plant parts for all substances with systemic properties during the sample preparation.

# 2.2.2 Determination of CS<sub>2</sub>

The extraction method of thiram was based on the principle that dithiocarbamates are broken down to  $CS_2$  in an acid environment. The produced amount of  $CS_2$  was collected in a solution of diethanolamine (DEA), copper(II) acetate and ethanol 96% (colour reagent). The yellow-brown complex formed was quantified by spectrophotometry. In this way, the produced amount of  $CS_2$  could be determined by comparing it with a standard series of known amounts of  $CS_2$  (Coresta, 1978; Caldas et al., 2001). A 500 ml three neck flask was heated by means of an electric heating element. The third neck was connected to a diaphragm pump whereas a tube was placed on the first neck to pump air into the mixture. On top of the third neck, a reflux condenser was placed which was connected to two spiralshaped wash bottles in series. During the sample preparation, plant samples were cut into large pieces and mixed to a homogeneous mixture. Further, 50 g of this mixture was weighed into a three neck flask and 130 ml of an acid stannous chloride solution was added. The first wash bottle contained 30 ml NaOH (6.5%) whereas the second wash bottle contained the colour reagent. The colour reagent was obtained by dissolving 12 mg of copper(II) acetate and 25 g DEA in ethanol (96%) and bringing the volume to 250 ml. Once the distillation equipment was set up, the water cooling, the diaphragm pump and the heater were turned on. The mixture was boiled for 30 minutes. If a discolouration occurred after 30 minutes in the wash bottle with the colour reagent, the contents of this bottle was transferred into a 50 ml volumetric flask and rinsed with ethanol (96%). The volume was adjusted to 50 ml with ethanol (96%) and the absorbance was measured at 435 nm by using a UV-VIS spectrophotometer. Finally, the absorbance value was compared with the absorbance of a control solution.

#### 2.2.3 Extraction of beeswax

Niell et al. (2014) described a simple variation of the QuEChERS method which was used for the analysis of PPP residues in beeswax. The study described a multi-residue analysis based on the QuEChERS method. By making some modifications to the already described QuEChERS method (2.2.1), acceptable recovery values were obtained for a large number of PPPs present in the beeswax (Niell et al., 2014). First, 2 g of beeswax was weighed into a 50 ml centrifuge tube, 10 ml of acetonitrile was added, and the tube was closed and placed in a water bath at 80°C. After the wax had melted, the tube was shaken vigorously for 10 to 15 seconds and placed back into the water bath to melt again. The procedure was repeated four times. Then, the tube was left to cool to room temperature and put into the freezer at -18 °C for at least two hours for precipitation of the wax. An aliquot of the extract was transferred into a PP single-use centrifugation tube, which contained 25 mg of primary-secondary amine (PSA) and 25 mg of C18 sorbent. The tube was shaken vigorously for 30 seconds and centrifuged for five minutes at 5000 rpm. Finally, the cleaned extract was transferred into a screw-cap vial.

### 2.2.4 Extraction of bee bread

Bee bread was analysed based on a method described in a study of Kasiotis et al. (2014). First, 3 g of bee bread was weighed into a 50 ml centrifuge tube and 7 ml of water was added. Subsequently, 11.5 mL of acetonitrile and 3.5 ml of hexane were added. The tube was shaken for a few seconds and salts were added (6 g MgSO<sub>4</sub> and 1.5 g NaOAc). The tube was shaken for two minutes and centrifuged for five minutes at 5000 rpm. 7 to 8 ml of the acetonitrile layer was transferred into a PP single-use centrifugation tube which contained 25 mg of PSA and 25 mg of C18 sorbent. The tube was shaken again for two minutes and centrifuged for five minutes at 5000 rpm. 5 ml of the extract was passed through an Extra Bond  $C_{18}$  cartridge 200 mg which was preconditioned with 3 ml of acetonitrile and 3 ml of water under vacuum. Rinsing with an additional volume of 3 ml of acetonitrile, collection of the organic extracts and evaporation to dryness under a vacuum resulted in the dry concentrate which was reconstituted in 1 ml of an acetonitrile/water (10:90) solution.

### 2.3 LCMS-MS analysis

Extraction of the active substances from the plant matrix was followed by the quantification of the PPP residues. In this study, a LCMS-MS device was used. The LCMS-MS system consisted of a Waters Acquity Series UPLC instrument coupled to a Waters Xevo TQD MS/MS system. To separate the active substances, a Waters BEH  $C_{18}$  column with dimensions 100 mm x 2.1 mm and a particle size of 1.7  $\mu$ m was used. The quantified substances with accompanying masses of parent and daughter ions which were examined with the LCMS-MS system are listed in Table B-9 of Appendix B. All vials including extracted active substances were injected directly into the device, except those of the beeswax samples. Before injecting the beeswax sample into the LCMS-MS device, the pH of these samples was quickly lowered to ca. 5 by adding a 5% formic acid solution in acetonitrile (10  $\mu$ l/ml extract).

## 2.4 Data analysis

Data processing was performed using SPSS 23.0 to compare the residue levels observed in the samples of the greenhouse trial. First, normality and homoscedasticity (equality of variances) should be checked when performing a parametric test. The normality of the results was examined by using a Shapiro-Wilk Test (n < 50) and the Modified Levene's test was used to determine the homoscedasticity. If normality and equal variances could be identified, the various residue levels were compared by using an independent t-test. When normality could not be determined, a non-parametric test was used. The non-parametric test used was the Mann-Whitney U test during which several p-values were calculated in order to determine significant differences.

# **Results and discussion**

# 3.1 Greenhouse trial

# 3.1.1 PPP residue trial during growth of maize

Methiocarb residues from coated seed were monitored during a trial in maize plants. Taking into account the maximum authorised application rate of Mesurol FS 500 on maize seed (1 l product/100 kg seeds), the expected amount of methiocarb on one seed is 1.43 mg and 1.56 mg for Telexx and Ronaldinio maize respectively (2.1.1.1). Unlike the calculated methiocarb residue, the seed of the Ronaldinio variety contained on average 0.107 ( $\pm$  0.004) mg methiocarb/seed while Telexx seed contained 1.337 ( $\pm$  0.207) mg methiocarb/seed. The average recovery value of seed analysis was 98.23%. The variability (relative standard deviation) between samples of Ronaldinio seed was 4% and 16% between samples of Telexx

seed. The quantity of methiocarb present on Telexx seed was higher than the amount on Ronaldinio seed. Furthermore, the amount of active substance found on the seed of Ronaldinio maize was much lower than expected. This can be explained by the fact that probably a lower application rate of Mesurol FS 500 was applied on Ronaldinio seed compared to the recommended dose. Another possible explanation for this difference in amount of active substance on the seed, is the difference in seed treatment quality between Ronaldinio and Telexx maize. Differences in seed treatment quality between the two varieties could also be determined visually (Figure 4-7). Seeds are transported after treatment during which seed coating can be damaged. The extent to which the coating disappears from the seeds, is dependent on the quality of the treatment. The quality of the coating on the seed was lower for the Ronaldinio seed. The coating released more easily by friction compared to the Telexx seed.



Figure 4-7 Untreated maize seed (left) vs. treated maize seed: Ronaldinio (centre) and Telexx (right).

Samples were taken from the 4<sup>th</sup> to 5<sup>th</sup> leaf stage for the analysis at which the foliage and roots of the plant were examined separately. Sampling of the 3<sup>rd</sup> to 4<sup>th</sup> leaf stage was not included in this analysis since the mass of the plants was quite small at this stage. A minimum weight was required to perform the analysis which meant that many plants were necessary to examine this particular stage of development. Due to the restricted test area, a limited amount of plants were sown.

### 3.1.1.1 Methiocarb residues in maize

Figure 4-8 illustrates the residue levels of methiocarb found in Ronaldinio and Telexx maize. A distinction was made between foliage and roots of Telexx maize. The average residue levels are shown with their corresponding standard deviation (SD). The results in Figure 4-8 show a decline between the different leaf stages. The methiocarb residues decreased as the plant developed. A significant decrease in methiocarb residues was found between the 4<sup>th</sup> to 5<sup>th</sup> and 7<sup>th</sup> leaf stage (p = 0.05) for Ronaldinio maize. Telexx maize showed a significant decrease between the 3<sup>rd</sup> to 4<sup>th</sup> and 6<sup>th</sup> leaf stage, between the 3<sup>rd</sup> to 4<sup>th</sup> and 7<sup>th</sup> leaf stage and between the 3<sup>rd</sup> to 4<sup>th</sup> and 6<sup>th</sup> leaf stage (p = 0.05). The decrease between the 3<sup>rd</sup> to 4<sup>th</sup> and 4<sup>th</sup> to 5<sup>th</sup> leaf stage as well as between the 4<sup>th</sup> to 5<sup>th</sup> and 6<sup>th</sup> and 7<sup>th</sup> leaf stage was not significant (p = 0.083; p = 0.248 and p = 0.083 respectively). The residue levels of methiocarb found in the

roots and foliage of Telexx maize showed significant differences in all different leaf stages (p = 0.05).



Figure 4-8 Average residue levels of methiocarb ( $\pm$  standard deviation) found in the entire plant  $\blacksquare$  of Ronaldino maize (A), the entire plant  $\blacksquare$  of Telexx maize (B), the foliage  $\blacksquare$  of Telexx maize (C) and the roots  $\blacksquare$  of Telexx maize (D) (n = 3).

#### 3.1.1.2 Methiocarb sulfoxide residues in maize

In Figure 4-9, the average residue levels of methiocarb sulfoxide are shown with their corresponding standard deviation. These results indicated that the methiocarb sulfoxide reduced as the plant developed. The initial deposit varied between the two maize varieties, but the residue levels showed a quite similar progress. A significant difference was found between the 4<sup>th</sup> to 5<sup>th</sup> and 7<sup>th</sup> leaf stage for Ronaldino maize (p = 0.05). The differences between the other samplings were not significant (p > 0.05). All the leaf stages of Telexx maize showed significant differences in residue levels of methiocarb sulfoxide, except between the 6<sup>th</sup> and 7<sup>th</sup> leaf stage (p = 0.127). Figure 4-9C illustrates that residue levels of methiocarb sulfoxide were mainly present in the roots of Telexx maize.



Figure 4-9 Average residue levels of methiocarb sulfoxide ( $\pm$  standard deviation) found in the entire plant  $\blacksquare$  of Ronaldinio maize (A), the entire plant  $\blacksquare$  of Telexx maize (B) and the foliage  $\blacksquare$  and roots  $\blacksquare$  of Telexx maize (C) (n = 3).

### 3.1.1.3 Methiocarb sulfone residues in maize

The average residue levels of methiocarb sulfone found in Ronaldinio and Telexx maize are illustrated in Figure 4-10 with their corresponding standard deviation. The residue levels showed a peak in the 6<sup>th</sup> leaf stage of the plant in both maize varieties. Figure 4-10C indicates that this peak was due to a significant increase of residue level in the foliage during the 6<sup>th</sup> leaf stage (p = 0.05). The residue levels of methiocarb sulfone in the roots significantly decreased in the 6<sup>th</sup> leaf stage compared to the 4<sup>th</sup> to 5<sup>th</sup> leaf stage (p = 0.05).

Both the increase in residue level between the 4<sup>th</sup> to 5<sup>th</sup> leaf stage and 6<sup>th</sup> leaf stage as well as the decrease of methiocarb sulfone residues between the 6<sup>th</sup> and 7<sup>th</sup> leaf stage of Ronaldinio maize were significant, i.e. p = 0.05 and p = 0.046 respectively. The decrease between these last leaf stages in the foliage of Telexx maize was not significant (p = 0.275). Figure 4-10C also illustrates that methiocarb sulfone mainly occured in the foliage and disappeared rather quickly in the roots. In the 7<sup>th</sup> leaf stage no methiocarb sulfone was found in the roots of Telexx maize.



Figure 4-10 Average residue levels of methiocarb sulfone ( $\pm$  standard deviation) found in the entire plant  $\blacksquare$  of Ronaldinio maize (A), the entire plant  $\blacksquare$  of Telexx maize (B) and the foliage  $\blacksquare$  and roots  $\blacksquare$  of Telexx maize (C) (n = 3).

# 3.1.1.4 Discussion

The parent methiocarb metabolises both in the soil as well as in the plant. Bowman and Beroza (1969) added methiocarb to apples, pears and maize plants to test gas and liquid chromatography analytical methods. The methylthio group of methiocarb was oxidised in plant samples to sulfoxide and sulfone with hydrolysis to the corresponding methylthio phenol, methylsulfoxide phenol and methylsulfone phenol (Figure 4-11). In studies of Abdel-Wahab et al. (1966) and Kuhr and Dorough (1976), methiocarb was injected into bean plants and oxidised to form sulfoxide. The sulfoxide itself was oxidised, more slowly, to the sulfone. In samples taken 24 hours after injection, about 20% of the applied dose was recovered as parent methiocarb, indicating a rapid metabolism. In a study conducted by the European Food Safety Authority (EFSA, 2006), labelled <sup>14</sup>C-methiocarb was applied to the soil at a rate of 1.12 kg a.s./ha to lettuce, tomato seedlings (Solanum lycopersicum), mature tomato plants and rice seed (Oryza sativa) simulating a seed treatment. Uptake of radioactivity was rapid in both lettuce and tomato plants. A continued increase in uptake of radio-labelled material was observed during the study time. On characterisation of the organic-extractable radioactivity in lettuce, tomato and rice two major components were identified as methiocarb and methiocarb sulfoxide between 1 and 35 days after application. Up to 19% of the total <sup>14</sup>C-residue (TRR) was recovered as methiocarb whereas up to 52% of TRR as methiocarb sulfoxide. Methiocarb sulfone residues were minor (max 2% TRR).



Figure 4-11 Metabolism of methiocarb in soil (s), plants (p) and in microsomal preparations from mammals and insects (mi): methiocarb (1), methiocarb methylthio phenol (2), methiocarb methylsulfoxide phenol (3), methiocarb methylsulfone phenol (4), methiocarb sulfoxide (5), methiocarb sulfone (6) and N-hydroxymethyl derivative (7).

The results obtained by performing the greenhouse trial indicated that methiocarb residue levels were always higher in all leaf stages compared to the residue levels of its metabolites. These results did not correspond to the results of EFSA (2006) at which lettuce, tomato plants and rice were examined. The difference in results may be due to the method in which methiocarb was applied. EFSA (2006) applied methiocarb to the soil whereby methiocarb was more quickly degraded to its metabolites. Soil degradation occurs faster than degradation after seed treatment since the substance is more spread in the soil. Residue levels found in the samples in this greenhouse trial showed a high variability between samples, ranging from 9-93%, 11-62%, 23-73% and 3-43% in the entire plant of Ronaldinio maize, the entire plant of Telexx maize, and in the foliage and roots of Telexx maize respectively (Figure 4-8, 4-9 and 4-10). Figures 4-8 and 4-9 illustrate that the residue levels of methiocarb in Ronaldinio maize were 10 to 20 times higher than the residue levels of methiocarb sulfoxide up to the 6<sup>th</sup> leaf stage. In the 7<sup>th</sup> leaf stage the residue level of methiocarb was only twice as higher than the residue level of methiocarb sulfoxide. Residue levels of methiocarb in Telexx maize were 10 to 30 times higher than the residue levels of methiocarb sulfoxide in all the leaf stages. Residue levels of methiocarb sulfoxide and methiocarb sulfone were illustrated in Figures 4-9 and 4-10. In the 3-4<sup>th</sup> leaf stage, the residue level of methiocarb sulfoxide in Ronaldinio maize was 10 times higher than the residue level of methiocarb sulfone. However, this ratio decreased during the following leaf stages and residue levels of methiocarb sulfoxide were even 40 and 70 times lower than the residue levels of methiocarb sulfone in the 6<sup>th</sup> and 7<sup>th</sup> leaf stage respectively. When the entire plant of Telexx maize was analysed, the residue levels of methiocarb sulfoxide were 40 to 50 times higher than the residue levels of methiocarb sulfox of methiocarb sulfoxer, the ratio decreased in the later leaf stages where the residue levels of methiocarb sulfoxide were ratio decreased in the later leaf stages where the residue levels of methiocarb sulfoxide were twice as high than the residue levels of methiocarb sulfoxer.

The uptake of methiocarb depends on both concentration and the solubility of the metabolites. Methiocarb has a solubility in water of 27 mg/l (20°C) and an octanol-water partition coefficient of 3.18 (expressed as log P at pH 7 and 20°C) (Lewis and Green, 2011). No exact values for the solubility or octanol-water partition coefficient of methiocarb metabolites were found in literature. A conclusion on the plant residue definition for risk assessment and monitoring was unable to be reached in the peer review procedure conducted by EFSA (2006). Studies investigated by EFSA (2006) indicate that low levels of methiocarb residues are found in plants. Therefore, a low risk is anticipated for bees. The experts' meeting for residues concluded that there is currently insufficient information available to have a firm view on the residue definition and proposed that either all phenol metabolites should be included in the plant residue definition and hence more information on appropriate residue trials is needed or alternatively that the toxicity of the phenol metabolites should be further addressed (EFSA, 2006). However, the results of the guttation trial (3.1.2) and the more polar structures of the metabolites indicate that these metabolites have a higher water solubility than methiocarb. Due to the expected higher solubility, metabolites are more easily absorbed by the plant and can achieve a higher residue level than methiocarb in mature crop plants when the concentration in the soil moisture is sufficiently high.

The results of the greenhouse trial where a distinction was made between the roots and the foliage of the maize plant illustrated that methiocarb mainly occurred in the roots of the plant. Only some small portions were found in the foliage. In consecutive sampling, the residue levels decreased with 50% indicating a rapid metabolism of methiocarb in the plant. Abdel-Wahab et al. (1966) and Kuhr and Dorough (1976) indicated that only 20% of the applied dose of methiocarb was recovered 24 hours after injection. However, methiocarb was not injected into the plant when performing the greenhouse trial. On the other hand, the results of Abdel-Wahab et al. (1966) and Kuhr and Dorough (1976) also pointed out that methiocarb undergoes a rapid metabolism in the plant.

The metabolite methiocarb sulfone was especially located in the foliage of the plant according to the obtained results (3.1.1.3). The residue levels of methiocarb sulfone in the

roots were low and decreased as the plant developed. In the 7<sup>th</sup> leaf stage, residue levels were no longer found in the roots. The decrease was due to the growth of the plant on the one hand and through hydrolysis of the component to methiocarb methylsulfone phenol on the other hand. In the 6<sup>th</sup> leaf stage, the residue levels of methiocarb sulfone indicated a peak. This peak was also observed in the foliage of the plant as shown in Figure 4-10C. The difference with the previous leaf stages was that the total leaf area of the plant strongly increased and the erectophile position of the leaves changed into a more planophile position (Loomis and Williams, 1969; Lemeur and Blad, 1974; Vandelanotte, 2009). Because of these changes the photosynthetic capacity of the plant increased as the leaves absorbed more light. The increased photosynthetic capacity was associated with an increased upward transport of the substances dissolved in water through the xylem sap to the leaves. Given that the leaves absorbed more light, the temperature of the leaves increased as well. Methiocarb sulfone and methiocarb sulfoxide were both transported through the xylem sap to the leaves. The presence of methiocarb sulfoxide in the leaves in combination with the increased temperature, increased the oxidation of methiocarb sulfoxide to methiocarb sulfone. This increased oxidation process explains the peak in the course of methiocarb sulfone. The residue levels of methiocarb sulfoxide were especially located in the roots of the plant. The residues of this component decreased through the investigated leaf stages.

This experiment was conducted under optimal growth conditions in a greenhouse. Various studies already illustrated the discrepancy between results of laboratory and field experiments (Harned and Tortora, 1986; Schmuck et al., 2001). When performing a field study, residue levels can be influenced by ambient conditions and variability between samples is usually higher compared to tests conducted in the laboratory. Due to the limited available literature concerning the fate of methiocarb and its metabolites in the plant, it is difficult to make strong statements about these substances. However, the results of this greenhouse trial illustrated that residue levels of methiocarb rapidly decrease into the plant not only by plant growth but also due to metabolism into its metabolites. Due to higher systemic properties (higher polarity due to incorporation of oxygen), these metabolites are more easily translocated throughout the plant than methiocarb. Therefore, higher residue levels of these metabolites can be present in mature crop plants compared to methiocarb.

#### 3.1.2 Guttation trial

The guttation phenomenon is affected by a number of factors such as humidity, temperature, growth stage, water stress, root depth and soil water potential. Moreover the insecticide residues in guttation fluid exhibit wide variability due both to factors affecting guttation as a phenomenon and to formulation, metabolism within the plant, application methods, adjuvant, solubility of the active substance and plant species (Tapparo et al., 2011). This guttation trial was conducted in order to verify if methiocarb and its metabolites are translocated from the seed to guttation drops and if they could be quantitative determined. Factors affecting guttation were not further investigated in this study. During

the guttation trial, guttation drops were collected daily after emergence of the seedlings. Further, these drops were examined on the presence of methiocarb and its metabolites. Methiocarb is classified by the World Health Organization (WHO) as a highly toxic product, wherein the  $LD_{50}$ -values of the component are shown in Table 4-6.

Table 4-6 Toxicity values of methiocarb (µg/bee) for the honey bee (Apis mellifera).

Exposure	LD <sub>50</sub> (µg/bee)	Reference		
acute, oral	0.47	Sanchez-Bayo and Goka (2014)		
acute, contact	0.23	Paranjape et al. (2014)		
LD Lathel Deep studieb 500/ of the test energies source mentality in a single deep				

 $LD_{50}$  = Lethal Dose at which 50% of the test species cause mortality in a single dose.

Toxicity values of methiocarb sulfone and methiocarb sulfoxide for the honey bee were not available in literature. Table 4-7 shows the residue levels of methiocarb and its metabolites in the guttation drops.

Table 4-7 Residue levels (mg/l) of methiocarb, methiocarb sulfoxide and methiocarb sulfone found in the guttation drops collected on consecutive days during the  $1^{st}$  to  $3^{rd}$  leaf stage of the plant (n = 1).

Days after sowing	Methiocarb (mg/l)	Methiocarb sulfoxide (mg/l)	Methiocarb sulfone (mg/l)
9	0.185	3.679	< LOD
10	0.111	4.394	0.105
11	0.051	3.937	0.047
12	0.001	2.858	0.076

Table 4-7 illustrates that residue levels of methiocarb sulfoxide in the guttation drops were higher than those of methiocarb. This is due to the expected higher water solubility of the metabolite and the rapid metabolism of methiocarb to its metabolites. The higher solubility facilitates the translocation of the component through which it is transported to the leaves via the water transport in the xylem sap. The results of the previous described greenhouse trial (3.1.1) indicated that residue levels of methiocarb sulfone were repeatedly lower than the other two components. The results of the guttation trial illustrated that upward of the second sampling the residue levels of methiocarb sulfone were about as high as the residue levels of methiocarb. Again, this indicates a higher solubility of the component and an improved translocation of the component in the plant compared to methiocarb. Only toxicity values of methiocarb could be obtained from literature and were compared with the residue levels found in the guttation fluids. Analogous to the study of Girolami et al. (2009), a 5 µl water uptake for the bee was assumed. Table 4-8 shows based on the obtained results of Table 4-7, the absorbed residue levels of methiocarb, methiocarb sulfoxide and methiocarb sulfone by a bee while consuming guttation fluids. Comparing the amounts listed in Table 4-8 with the acute oral LD<sub>50</sub>-value of methiocarb for the honey bee (Table 4-6), the residue levels in the samples never exceeded the  $LD_{50}$ -value of 0.47 µg/bee. Girolami et al. (2009) found exposure values for bees of 0.060 μg thiamethoxam, 0.115 μg clothianidin and 0.235 µg imidacloprid per bee (application rates of 1 mg/seed, 1.25 mg/seed and 0.5 mg/seed respectively). Despite the higher application rate per seed (1.43 mg methiocarb/seed), residue levels of methiocarb found in the guttation drops of this study were much lower. Because guttation is affected by several factors that cause a high variability both in its intensity and in the insecticide content, further experiments are needed to better understand the phenomenon and the consequent risk assessment for honey bees. Furthermore, it is important to clarify in laboratory and field studies to what extent bees are attracted to the guttation drops and whether they are able to change their behaviour and the intensity of their flights when water sources are contaminated. Further research of the Bee Protection Group (ICPPR) also focuses on the risk assessment related to guttation drops (1.4.1).

Table 4-8 Absorbed residue levels ( $\mu$ g/bee) of methiocarb, methiocarb sulfoxide and methiocarb sulfone by a bee while consuming guttation fluids (5  $\mu$ l, Girolami et al., 2009) from plants during the 1<sup>st</sup> to 3<sup>rd</sup> leaf stage (n = 1).

Days after sowing	Methiocarb (µg/bee)	Methiocarb sulfoxide (µg/bee)	Methiocarb sulfone (µg/bee)
9	0.00092	0.01839	-
10	0.00055	0.02197	0.00052
11	0.00026	0.01968	0.00024
12	0.00005	0.01429	0.00038

## 3.2 Monitoring

Table 4-9 indicates the results of the residue analysis of the monitoring. All active substances of the seed treatment are shown with their corresponding recovery values, crop and development stage of the plant.

Table 4-9 Monitoring results of the residue analysis ( $\mu$ g/kg plant material ± standard deviation) with corresponding recovery values (%), crop and development stage of the plant (BBCH-stage) for each active substance (n = 3) of the seed treatment.

Active substance	Recovery (%)	Crop (part)	BBCH-stage	Residue level
				$(\mu g/kg plant material \pm SD)^a$
fludioxonil	69.39	rye	85-89	< LOD
		winter wheat (roots)	14-16	20.0 ± 13.8
		winter wheat	85-89	8.2 ± 9.6
imazalil	40.46	spring barley	85-89	37.3 <sup>b</sup>
prothioconazole	9.97	spelt	85-89	< LOD
		triticale	85-89	< LOD
		winter barley (roots)	14-16	< LOD
		winter barley (foliage)	14-16	< LOD
		winter barley	85-89	< LOD
methiocarb	98.94	maize (roots)	85-89	2.0 <sup>b</sup>
		maize (lower leaves)	85-89	< LOD
		maize (top leaves)	85-89	< LOD
		maize (ear)	85-89	< LOD
methiocarb sulfone	105.45	maize (roots)	85-89	< LOD
		maize (lower leaves)	85-89	32.9 ± 4.4
		maize (top leaves)	85-89	46.5 ± 10.0
		maize (ear)	85-89	< LOD
methiocarb sulfoxide	94.34	maize (roots)	85-89	3.9 ± 2.7
		maize(lower leaves)	85-89	0.7 ± 0.6
		maize (top leaves)	85-89	< LOD
		maize (ear)	85-89	< LOD
metalaxyl-M	93.38	brown bean	85-89	< LOD
		onion seed	81-89	< LOD
thiram	-	brown bean	85-89	< LOD
		chicory	85-89	< LOD
		onion seed	81-89	< LOD
		sugar beet	85-89	< LOD

BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale); <sup>a</sup>LOQ =  $0.1 \mu g/kg$  for all substances except for prothioconazole,  $5 \mu g/kg$ ; <sup>b</sup>SD is not listed since the residue level was obtained in only one sample.

### 3.2.1 Fludioxonil

According to the study of the National Registration Authority for Agricultural and Veterinary Chemicals (2000), fludioxonil is an immobile component and microbial degradation in the soil is quite limited. Furthermore, fludioxonil is stable to hydrolysis and degradation mainly

takes place through photolysis when the component is in solution or to the surface of the soil. Monitoring of the residue levels observed in winter wheat (Table 4-9) confirmed that fludioxonil is a stable component, which is difficult to degrade. On the other hand, no residues were detected in rye (< LOD; Table 4-9). These results can be explained by the various soil texture of the fields where the crops were located. The soil texture of the field with winter wheat had a clayey texture whereas the field with rye had a sandy texture. PPPs are better preserved in a clay soil and are less present in the soil solution compared to a sandy soil (Trautmann et al., 2012; Gardner, 2016). PPPs are less available for degradation by photo- or hydrolysis through which higher residue levels are possible in a clay soil. Furthermore, soil leaching occurs slower in a clay soil compared to a sandy soil which will slow down the transport of the PPPs away from the seed (Johnston, 2001). As a result, fludioxonil is a persistent substance and can remain in the underground parts of the plant up into the mature stage. This persistence was also shown by the spray records of winter wheat (Appendix B, Table B-7) at which fungicides were only used in later development stages of the crop.

#### 3.2.2 Imazalil

Imazalil is moderately soluble in water (184 mg/l) and is very stable to hydrolysis. Photodegradation occurs relatively rapidly and soil degradation is very slow under aerobic conditions. Further, the component is guite immobile in the soil and is not expected to volatilise (US EPA, 2005). Due to these properties and by taking into account the systemic activity of imazalil, residues can be expected in the plant. Cheng et al. (1994) examined wheat grown from imazalil treated seeds. Residues of imazalil and its two metabolites, R14821 and R42639, were determined. Imazalil was the major component detected in straw grown from wheat treated seeds. In another study, barley seeds treated with a dose of <sup>3</sup>Himazalil were sown in soil. Plants were harvested after one and three weeks. Soil and plant parts were analysed for radioactivity. Most of the radioactivity was present in the soil directly around the seed coats. After three weeks, the green parts of the plant contained only 6% of the radioactivity that had originally adhered to the seeds (FAO, 1978). In the monitoring results, a residue level of 37.3 µg/kg plant material was found in only one of the two samples making it difficult to arrive at a decision on the average residue level of imazalil that was expected in the mature stage of spring barley (BBCH 85-89; Table 4-9). Based on the results of Table 4-9 and those of the studies described above, residues of imazalil can be present in the mature stage of the plant. In 1978, the Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization already indicated that imazalil is quite fast present in the soil solution after which it is absorbed by the plant. The uptake will continue for quite a long time because of the stability and immobility of imazalil in the soil. This leads to possible residues in the mature stage. This conclusion was further enhanced since no fungicides were used in the spray records (Appendix B, Table B-3).

## 3.2.3 Prothioconazole

The entire plant of spelt and triticale was used in the residue analysis of prothioconazole, but no residues were detected (< LOD). For winter barley, two samplings took place at which the first occurred during the mature stage of the plant (BBCH 85-89). The analysis of this stage was done on the entire plant and also here no residues were detected (< LOD; Table 4-9). The second sampling of winter barley happened several weeks after emergence (BBCH 14-16) and in the analysis of these samples a distinction was made between above- and underground plant parts. However, after analysis again no residues of prothioconazole were detected both in the above- and in the underground plant parts (< LOD). In a study of the Australian Pesticides and Veterinary Medicines Authority (2007), cereals of treated seed contained no prothioconazole either. The spray records (Appendix B, Table B-5 and B-6) show that the crops were treated a few more times with fungicides which explains the nonpersistence of prothioconazole. However, prothioconazole degrades rather quickly to prothioconazole-desthio which is quite persistent in soil and moderately mobile. The desthio component is stable to hydrolysis and is very slowly degradable in aerobic soils. Photolysis of the desthio component in solution also occurs very slowly (Ambrus, 2008). In this study, this matter was not further investigated. Furthermore, the recovery values of the prothioconazole residue analysis in spelt, triticale and winter barley were quite low because of which no clear conclusion could be made (Table 4-9).

## 3.2.4 Methiocarb

Methiocarb has different degradation metabolites which have systemic properties into the plant. Due to these systemic properties, different plant parts were analysed separately. The results indicated that in mature crop plants methiocarb was almost completely converted to the metabolites methiocarb sulfone and methiocarb sulfoxide. Further, methiocarb sulfone proved to be the major component in the aboveground parts of the plant. Both the lower leaves as well as the top leaves (along with a part of the stem) contained a higher residue level of methiocarb sulfone. The major component in the roots of the plant was methiocarb sulfoxide whereas methiocarb occured only once and no residues of methiocarb sulfone were found (Table 4-9). Discussion of these components is described in the paragraph of the greenhouse trial (3.1).

### 3.2.5 Metalaxyl-M

Due to the systemic nature of the component metalaxyl-M, the residue analysis was performed on different plant parts. Distinction was made between the above- and underground parts of the plant for onion seed. The brown bean plant was divided into roots, foliage and beans. None of the investigated plant parts contained residue levels of metalaxyl-M (< LOD; Table 4-9). This observation was consistent with the results from a study by Singh et al. (1986). Uptake of metalaxyl through roots, leaves and seed, its translocation and distribution in different plant parts and persistence following seed
application were studied in pearl millet (*Pennisetum glaucum*) using <sup>14</sup>C-metalaxyl. Both uptake and efflux of metalaxyl by pearl millet seeds were complex and compartmentalised. Distribution inside the seed was not uniform. A major part of applied fungicide remained within the treated plant part, particularly after seed and foliar applications. Metalaxyl was ambimobile inside the plant and was found to get accumulated at apex and margins of leaf blade. No metalaxyl could be detected in grains harvested from plants grown from metalaxyl treated seeds. When metalaxyl is used as seed treatment, a major part of the fungicide is lost through diffusion from seed into the soil during germination or remains in seed parts that are later shed to the ground.

### 3.2.6 Thiram

Thiram belongs to the dithiocarbamates which are not stable. Contact with acidic plant juices degrades dithiocarbamates rapidly and they decompose into CS<sub>2</sub> and the respective amine (Dasgupta et al., 2012). Several studies already examined the persistence of thiram. Womer and Balba (1979) indicated that wheat seedlings (five weeks old) growing in a sandy loam soil from thiram treated seed (334 mg/kg seed) still contained residues, i.e. the residue level of thiram was 0.019 mg/kg. Harned and Tortora (1986) grew soybean, cotton (Gossypium) and wheat plants in a greenhouse and in the field from seed treated with <sup>14</sup>Cthiram at various seed treatment rates per crop, i.e. 1.03 mg thiram/kg seed for soybeans, 1.4 mg thiram/g seed for cotton and 1.3 mg thiram/g seed for wheat. The results of the study were expressed in <sup>14</sup>C-concentrations. After 30 days, <sup>14</sup>C-residues were found in both the greenhouse and open field test at which the indicated residue levels were in most cases lower in the open field test. The study also examined mature plants revealing that when they were grown on open field, no residues were found into the soybean and cotton plants. However, wheat plants contained residues at concentrations of 0.298 and 0.822 mg<sup>14</sup>C/kg dry weight of plant tissue for the chaff and leaves respectively in the greenhouse test. In the open field test, the residue levels of <sup>14</sup>C expressed as thiram were lower. No specific information was given about the ambient and soil conditions of both the open field and greenhouse test. The fact that thiram degrades more rapidly in acidic soils and in soils high in organic matter, can explain the difference in residue levels found in wheat. The above described studies indicate that thiram is rapidly decomposed to CS<sub>2</sub> and that residue levels are quite low in mature crop plants. This explains why residue levels of thiram were not detected for chicory, onion seed, sugar beet and brown bean in the monitoring (Table 4-9).

#### 3.3 Beeswax and bee bread trial

Both beeswax and bee bread of five different hives were examined for the presence of various PPPs. A first step of the analysis consisted of a multi-residue screening to determine the present PPPs. Second, a quantification of the present PPPs was carried out with a corresponding determination of the recovery values.

# 3.3.1 Bee bread

Simon-Delso et al. (2014) investigated bee bread of various hives. The hives were situated at locations on the border between Flanders and Wallonia (Belgium). A distinction was made between hives with healthy colonies and hives with colonies with well identified problems. Disorders included dead colonies or colonies in which part of the colony appeared dead, or had disappeared, weak colonies, queen loss or problems linked to brood and not related to any known disease. Results showed that only in a very small portion, up to seven samples of the 108 tested bee bread samples, PPPs were detected. The substances found were boscalid, captan, coumaphos, fenpropimorph, iprodione, pyraclostrobin, pyrimethanil, tau-fluvalinate, thiophanate-methyl and zoxamide. Only boscalid, captan, iprodione and thiophanate-methyl could be quantified, i.e. 0.68, 1.90, 0.90 and 0.38 mg/kg bee bread respectively. Johnston et al. (2014) examined residues of PPPs in hives originating from several European countries. The most common PPPs in bee bread were products to control the Varroa destructor, i.e. amitraz (24% of the samples), tau-fluvalinate (24% of the samples) and coumaphos (12% of the samples). Some other detected active substances, at low frequency (4 to 8%), were the fungicides cyprodinil, carbendazim, chlorpyrifos, fludioxonil, boscalid, dimethomorph, fenhexamid, folpet and tebuconazole. No residues of the three neonicotinoids whereupon restrictions were imposed in Europe (i.e. imidacloprid, clothianidin and thiamethoxam), were detected in the bee bread. The above described studies indicated that different groups of products were observed in bee bread. However, the frequency at which residue levels occurred in bee bread was quite low. This amounted to 6.5% for the study of Simon-Delso et al. (2014). In the study of Johnston et al. (2014), amitraz and coumaphos were two of the three most common products in bee bread. These two products are no longer authorised in Belgium. Authorisations of amitraz and coumaphos were withdrawn in 2004 (in 2006 as veterinary medicinal product in beekeeping) and 2009 respectively (Ravoet et al., 2015).

In this study, no PPPs were observed after screening the bee bread of five different hives. The low frequency at which PPPs occur in bee bread is possibly due to microbiological processes – like lactic acid fermentation – taking place in the cells of the comb where the bee bread is stored over the winter. Fresh pollen is high in moisture and protein and, especially when brought into the hive – which stays around an internal temperature of 37°C – becomes an ideal environment for mould growth. The bees' digestive fluids, however, are rich with lactic acid bacteria (Vásquez and Olofsson 2009), which come to dominate the pollen substrate when it is packed together and sealed from the air with honey. The bacteria metabolise sugars in the pollen, producing lactic acid and lowering the pH (Mattila et al. 2012). Kye et al. (2009) observed that lactic acid bacteria can degrade organophosphorous insecticides by fermentation. These bacteria use organophosphate as a source of carbon and phosphorus. Microbiological processes may have resulted in degradation of some PPPs originally present down to levels below detection limits (Gilliam, 1979; Johnston et al, 2014). However, there is a need of further study about the degrading effects of fermentation on the other chemical PPPs. Moreover, it might be interesting to determine the contribution of

each species in the degradation of PPPs during the fermentation period. Components produced from degraded PPPs must be identified and evaluated as well.

#### 3.3.2 Beeswax

Table 4-10 illustrates the active substances found in beeswax as well as the corresponding recovery values, the sample on which a residue was found, the average residue levels ( $\mu$ g/kg beeswax ± SD) and residue levels cited from literature.

Various groups of active substances were observed in the beeswax samples (Table 4-10). Boscalid, cyprodinil, piperonyl butoxide and trifloxystrobin were detected just like in the study of Simon-Delso et al. (2014). The low recovery values for hexythiazox and piperonyl butoxide (Table 4-10) indicated that the extraction method used for these components can be further optimised. These low values made it difficult to make statements about these substances. Residue levels of hexythiazox, i.e. 3.6 to 235.5 µg/kg beeswax, were found in all samples except in the beeswax of the 5<sup>th</sup> hive. Hexythiazox is used as an acaricide on apples and grapes. A high risk of this substance to bees cannot be excluded for all representative uses because of the potential adverse effects on bee brood (EFSA, 2010b). Higher residue levels of piperonyl butoxide were found in all samples (except in the 5<sup>th</sup> hive) compared to Ravoet et al. (2015) and ranged between 13.0 to 136.8 µg/kg beeswax. Piperonyl butoxide is authorised for topical use in cows, sheep, goats and horses with no maximum residue level (MRL) required in food from these animals (Commission Regulation (EU) No 37/2010). In theory, piperonyl butoxide could be allowed to be used in beekeeping upon prescription of a veterinarian. The use of this PPP in beekeeping cannot be excluded: it is sometimes used in insect and bee repellents which are on the market for use in beekeeping. Some authors claim that piperonyl butoxide is enhancing the toxicity of fluvalinate to control Varroa destructor (Hillesheim et al., 1996). Piperonyl butoxide has the status of 'not a PPP' under Regulation (EC) No 1107/2009; but is a synergist. The provisions of Regulation (EC) No 396/2005 are not applicable.

Table 4-10 Active substances and their PPP group (insecticide = In, fungicide = Fu, acaricide = Ac, synergist = Syn) with corresponding average residue levels ( $\mu$ g/kg beeswax ± standard deviation, n = 3) and recovery value (%, n = 7) found in beeswax.

Active	PPP	Residue level in literature	Recovery	Sample	Residue level
carbofuran	In	(μβ/ κβ beeswax)	82.69	1 <sup>st</sup> wax plate <sup>d</sup>	(μg/ kg beeswax ± 3D) 1.4 <sup>j</sup>
				2 <sup>nd</sup> wax plate <sup>g</sup>	5.0 <sup>j</sup>
boscalid	Fu	290 (Simon-Delso et al., 2014)	60.97	1 <sup>st</sup> hive <sup>a</sup>	10.9 <sup>j</sup>
		12 (Ravoet et al., 2015)		2 <sup>nd</sup> hive <sup>b</sup>	16.4 ± 6.9
				3 <sup>rd</sup> hive <sup>c</sup>	< LOQ
				4 <sup>th</sup> hive <sup>d</sup>	9.0 ± 7.2
				5 <sup>th</sup> hive <sup>e</sup>	23.9 ± 11.7
				1 <sup>st</sup> wax plate <sup>f</sup>	13.9 ± 0.3
				2 <sup>nd</sup> wax plate <sup>g</sup>	15.6 ± 2.4
cyprodinil	Fu	< LOQ <sup>i</sup> (Simon-Delso et al., 2014)	92.27	1 <sup>st</sup> hive <sup>a</sup>	7.8 <sup>j</sup>
				2 <sup>nd</sup> hive <sup>b</sup>	64.3 ± 54.5
				3 <sup>rd</sup> hive <sup>c</sup>	21.5 ± 3.8
				4 <sup>th</sup> hive <sup>d</sup>	3.8 ± 2.9
				5 <sup>th</sup> hive <sup>e</sup>	1.7 ± 0.4
				1 <sup>st</sup> wax plate <sup>f</sup>	7.6 ± 0.7
				2 <sup>nd</sup> wax plate <sup>g</sup>	2.3 ± 0.3
azoxystrobin	Fu	-	59.60	2 <sup>nd</sup> hive <sup>b</sup>	1.3 <sup>j</sup>
				4 <sup>th</sup> hive <sup>d</sup>	14.2 ± 15.5
				5 <sup>th</sup> hive <sup>e</sup>	1.7 ± 0.7
				1 <sup>st</sup> wax plate <sup>f</sup>	1.4 <sup>j</sup>
pyraclostrobin	Fu	-	59.41	4 <sup>th</sup> hive <sup>d</sup>	19.9 <sup>j</sup>
				5 <sup>th</sup> hive <sup>e</sup>	$1.1 \pm 0.4$
trifloxystrobin	Fu	< LOQ <sup>i</sup> (Simon-Delso et al., 2014)	43.95	2 <sup>nd</sup> wax plate <sup>g</sup>	4.1 ± 1.1
hexythiazox	Ac	-	34.91	1 <sup>st</sup> hive <sup>a</sup>	181.3 <sup>j</sup>
				2 <sup>nd</sup> hive <sup>b</sup>	194.2 <sup>j</sup>
				3 <sup>rd</sup> hive <sup>c</sup>	32.3 ± 3.5
				4 <sup>th</sup> hive <sup>d</sup>	3.6 <sup>j</sup>
				1 <sup>st</sup> wax plate <sup>f</sup>	235.5 ± 11.6
				2 <sup>nd</sup> wax plate <sup>g</sup>	43.4 ± 4.9
piperonyl	Syn	10 (Ravoet et al., 2015)	14.64	1 <sup>st</sup> hive <sup>a</sup>	41.2 <sup>j</sup>
butoxide		< LOQ <sup>i</sup> (Simon-Delso et al., 2014)		2 <sup>nd</sup> hive <sup>b</sup>	40.12 ± 11.7
				3 <sup>rd</sup> hive <sup>c</sup>	13.0 ± 3.1
				4 <sup>th</sup> hive <sup>d</sup>	43.2 ± 10.8
				5 <sup>th</sup> hive <sup>e</sup>	< LOQ
				$1^{st}$ wax plate <sup>f</sup>	136.8 ± 16.3
				2 <sup>nd</sup> wax plate <sup>g</sup>	76.7 ± 1.5

<sup>a</sup>1<sup>st</sup> beehive from the centre of Roeselare; <sup>b</sup>2<sup>nd</sup> beehive from the centre of Roeselare; <sup>c</sup>3<sup>rd</sup> beehive from the centre of Roeselare; <sup>d</sup>1<sup>st</sup> beehive of the countryside in Moorslede; <sup>e</sup>2<sup>nd</sup> beehive of the countryside in Moorslede; <sup>f</sup>wax plate from a store; <sup>g</sup>wax plate consisting of melted beeswax plates collected over 10 years; <sup>h</sup>LOQ: 0.5 µg/kg beeswax (except for boscalid and hexythiazox: 2.5 µg/kg beeswax); <sup>i</sup>LOQ: 100 µg/kg beeswax; <sup>j</sup>SD is not listed since the residue level was obtained in only one sample.

Furthermore, various fungicides were detected in the beeswax samples as illustrated in Table 4-10. Fungicides can have an impact on the bee colony through changes to existing microflora in their food stocks as well as those in their intestinal canal. Several studies already demonstrated the potential changes in microbial composition of the microflora (Anderson et al, 2011;. Yoder et al, 2013.). These changes may give rise to dysbiosis, which is a disturbed balance of the intestinal flora (Sartor, 2008). This balance can be disturbed in such a way that the unfavourable microflora gain the upper hand and will negatively affect the bees. Pettis et al. (2013) showed that fungicides, such as pyraclostrobin, which was also found in this study, have an impact on the health of the bee. Honey bees exposed to this component showed an increased probability of Nosema infection. The same effect was observed for chlorothalonil. Boscalid is a fungicide distributed on plants. It can be applied in a large number of crops, particularly in fruit and vegetables growing in open air. The residue levels of boscalid, i.e. 9.0 to 23.9 µg/kg beeswax, matched with the results of Ravoet et al. (2015). However, Simon-Delso et al. (2014) found a residue level of 290 µg/kg beeswax based on only one sample with an LOQ-value of 100  $\mu$ g/kg beeswax. Under these high LOQ, boscalid was still detected in 22.2% of the samples. The quantified residue level gives a distorted view of the average residue level of boscalid in hives. According to studies conducted by US EPA (2010), boscalid is slightly toxic to honey bees via both oral ( $LD_{50}$  > 165.96  $\mu$ g/bee) and contact (LD<sub>50</sub> > 200  $\mu$ g/bee) exposure routes (acute). The residue levels found here (Table 4-10) were much lower than the toxicity values, so no lethal residue levels of boscalid were present in the samples. In addition, synergism with other active substances like insecticides are possible and increase the toxicity for honey bees (Simon-Delso et al., 2014). Cyprodinil, also a fungicide especially used in the cultivation of apple and pear trees and ornamental trees and shrubs, was quantified in all samples while in Simon-Delso et al. (2014) this substance could only be detected (< LOQ; Table 4-10). Azoxystrobin is active against a broad spectrum of fungi and can be used in the cultivation of cereals, most vegetables, fruit, potatoes and ornamental plants. Azoxystrobin was quantified in the wax of one beehive located in Roeselare, in the wax of the beehives located in Moorslede and in a wax plate coming from a store. Pyraclostrobin is used against fungi and rust on lawns, grapes and cereals and could only be quantified in the beeswax of the 4<sup>th</sup> and 5<sup>th</sup> hive. No residue levels of both azoxystrobin as pyraclostrobin in beeswax were found in literature. Trifloxystrobin, used on numerous crops, was detected in Simon-Delso et al. (2014), but quantification was not possible (< LOQ). In this study, a residue level of 4.1 µg/kg beeswax was found in the wax plate consisting of melted beeswax plates collected over ten years.

The highly toxic active substance carbofuran is used against soil insects in crops as maize and beet but is no longer authorised in the EU since 2007. Residue levels of carbofuran were found in the wax plates, i.e. 1.4 and 5.0  $\mu$ g/kg beeswax. The acute, contact LD<sub>50</sub>-value of carbofuran is 0.16  $\mu$ g/bee which is highly toxic to bees. The LOQ-value (0.5  $\mu$ g/kg beeswax) was also higher than the LD<sub>50</sub>-value as a result of which exposure to lethal doses cannot be excluded. Ravoet et al. (2015) examined beeswax of ten hives located in Flanders on the presence of PPPs. All samples contained PPPs and the most common substances were

coumaphos and tau-fluvalinate. Both products were used to control the Varroa destructor. Fluvalinate was used for both agricultural and beekeeping purposes. In Belgium, it was a widely used acaricide in the first years after the Varroa destructor mite established in 1984. However, it was abandoned by most beekeepers once fluvalinate-resistant mites were found all over Europe. Besides, a veterinary medicinal product with fluvalinate as active substance is no longer registered for apicultural use in Belgium since 2008. The PPP coumaphos is or was solely used in beekeeping and hence point to a beeswax contamination caused by apicultural practices. It is difficult to say whether it represents a recent or historic contamination. Since 2009, coumaphos is no longer authorised in Belgium. The beeswax samples also contained piperonyl butoxide (10% of the samples) and boscalid (20% of the samples) and were found at residue levels of 10 µg/kg and 12 µg/kg beeswax respectively. In France, a field survey over a period of three years (2002-2005) was conducted to study honey bee colony health in relation to PPP residues found in the colonies. Tau-fluvalinate and coumaphos were the two most frequently found residues in the French beeswax samples (Chauzat et al., 2009). Both tau-fluvalinate and coumaphos were not detected in the beeswax samples surveyed here (Table 4-10).

Just like in three other Belgian studies, no traces of neonicotinoids were found in beeswax (Nguyen et al., 2009; Simon-Delso et al., 2014; Ravoet et al., 2015). As mentioned before, neonicotinoids are a class of neuro-active insecticides that are among the culprits of bee mortality, and the subject of debate in Europe and beyond. So far only few of the studies that were undertaken succeeded in determining their presence in beeswax (Mullin et al., 2010; Yanez et al., 2013), though these insecticides are frequently found in other matrices (honey bees, pollen, honey; Mullin et al., 2010) and can cause adverse effects at ppb-levels (Blacquière et al., 2012). Not one of all detected substances is authorised in the treatment of seeds in Belgium. The consulted Belgian study only detected the seed treatment product iprodione (Simon-Delso et al., 2014). This substance was also included in the multi-residue analysis of this research but could not be identified. Products used in the treatment of seed are rarely detected in Belgian studies of beeswax. A lot of products, including iprodione, are also used as foliar or soil applications. It is quite unlikely that the origin of the products detected in the beeswax originated from seed treatment. Based on the residue levels of active substances found in the different hives, no direct link between hives with healthy and dead colonies and the amount of residues found in beeswax was observed.

Several fungicides were found in the beeswax of the hives studied here. Limited research is already conducted on the lethal and sub-lethal effects that these products exert on bees. These products deserve analysis for their specific toxicity, individually or in synergy with other substances or pathogens or their extensive exposure given their large scale and/or repeated use. Furthermore, data on the potential for synergistic mechanisms of PPPs within the hives and how they could affect honey bee populations are lacking. Research looking at additive and synergistic effects between multiple PPPs is clearly needed. Various studies already indicated the interactions between two chemicals (especially neonicotinoids).

Seed treatment

However, exposure data demonstrate that bees are often exposed to several PPPs over a period of time. Data are required to determine the effects of such long-term low level exposure to multiple PPPs on the health and functioning of honey bee colonies.

Beeswax contamination primarily affects the brood due to its direct contact with the brood cell wall. This developmental exposure to PPPs in brood combs will affect larval development (Wu et al., 2011) but also post-emergence fitness and performance of adult workers and queens (Pettis et al., 2004; Wu et al., 2012; Collins and Pettis, 2013; Rangel, 2013). Moreover, transfer from the beeswax matrix to the stored honey was experimentally demonstrated for PPPs (Wallner, 1995) and sulphonamides (Reybroeck et al., 2010). Hence, even without any recent environmental exposure, newly deposited honey can become contaminated by historical pollution of the beeswax. Given the contamination of beeswax, one may consider to take action in the field, aimed at lowering the PPP contamination of beeswax in Belgian apiaries, as stated before in other countries (Pettis et al., 2004).

## Conclusion

Seed treatment is considered to be an environmentally friendly form of chemical crop protection. The use of treated seeds also has some disadvantages which are all related to the coating of the seed. Residues of systemic active substances can be present in the guttation fluid, plant pollen and nectar of seed treated plants. In the last few years, this emission has resulted in bee losses in several countries and contamination of surface water. The uptake, translocation and persistence vary greatly among various products. In this study, these three factors were examined for various crops with special focus on the investigation of methiocarb residues in maize.

Various PPPs used in the treatment of seeds are quite persistent. Some of them can still be found in the mature stage of the plant. The seed treatment product methiocarb metabolises very quickly in the plant, which was observed by performing the PPP residue trial of maize (3.1.1). Because of the growth of the plant, dilution of active substances occurs, as well as degradation. The results of this trial indicated that residue levels of methiocarb quickly decreased into the plant. If the entire plant was included in the residue analysis, the residue levels of both methiocarb and methiocarb sulfoxide decreased continuously. The residue levels of methiocarb sulfone showed an irregularity. An increase of the methiocarb sulfone residues was observed in the 6<sup>th</sup> leaf stage caused by an increased photosynthetic capacity and an increased oxidation process of methiocarb sulfoxide to methiocarb sulfone. The results also showed that residue levels of methiocarb sulfone were mainly located in the upper plant parts and residue levels of methiocarb sulfoxide more in the lower plant parts. Both components are persistent since they were also found in mature plants. No residue levels of methiocarb sulfoxide were found in the upper plant parts of maize, only residue levels of methiocarb sulfone. The systemic properties of the metabolites of methiocarb were further demonstrated by means of the guttation trial. Both components as well as the parent methiocarb were found in the guttation fluids. However, methiocarb occurred at a very low concentration taking into account the quantity on the seed. This indicated a limited systemic activity. Residue levels of methiocarb sulfone were found at the same magnitude as methiocarb (0.1 mg/l) whereas residue levels of methiocarb sulfoxide occurred at a factor 30 to 40 higher (3.0 to 4.0 mg/l). Since residue levels of both metabolites in the roots of the plant were remarkably lower than those of methiocarb, their systemic activity is much higher. The observed residue levels of methiocarb in the guttation fluids revealed no lethal doses for bees.

Quantification of fludioxonil, imazalil and two metabolites of methiocarb, i.e. methiocarb sulfone and methiocarb sulfoxide was possible during the monitoring, showing that these substances are persistent. Fludioxonil is a non-systemic component that is difficultly degraded in the soil and in the plant. Moreover, fludioxonil is also immobile which increases the persistence of this product. Imazalil is a persistent active substance, just like fludioxonil. Once in the soil, imazalil is very immobile and degradation is quite slow. These two characteristics of imazalil as well as it being a systemic active substance ensure that residue levels of imazalil can be detected in mature crop plants.

The bee bread contained little to no PPPs. Beeswax however, appeared to be more contaminated with PPPs. Different groups of products were detected in beeswax, i.e. fungicides, insecticides and additives. In some beeswax samples, residue levels of insecticides were potentially lethal, e.g. carbofuran. Previous studies indicated that several fungicides, such as pyraclostrobin, may adversely affect the bee's health by making them more susceptible to infection. Fungicides play a possible role in the massive bee mortality as well. No traces of neonicotinoids were found in beeswax and not one of all detected substances is authorised in the treatment of seeds in Belgium. All observed substances are the result of foliar or soil applications in surrounding fields.

The results of this chapter illustrate that several active substances, used as seed treatment products, are translocated through the plant. These substances can end up in pollen, nectar and guttation fluids through which pollinators, e.g. bees, can be exposed. However, a lethal dose for bees was never registered in the guttation fluid during this study. Furthermore, not a single one of all detected substances in bee bread and beeswax is authorised in the treatment of seeds in Belgium. Based on the obtained results, the risk of exposure to pollinators by means of seed treatment is supposed to be very low for the crops and active substances investigated in this study. Within Belgium, it would be interesting to extend the work performed concerning seed treatment. Additional studies should cover other crops and seed treatment products to investigate whether this issue is limited to some specific cases or is more widespread within the seed treatment industry. Further studies at national level should be performed on beeswax (both beeswax on the market as beeswax used by beekeepers) to characterise residue levels of PPPs and veterinary medicines in beeswax since not yet many details are known about this topic.

A lack of knowledge on PPPs used for non-agricultural purposes still exists and especially actors responsible for non-agricultural PPP use (D) are still not sufficiently known (**RQ 7**). In order to provide an answer on this research question, **Chapter 5** describes an identification and allocation of non-agricultural use of PPPs in Belgium. Subsequently, one of the identified key players – i.e. non-professional PPP users – is further investigated to estimate if the non-professional use of PPPs (D) exert pressure (P) on operators, aquatic organisms and bees (**RQ8**). Knowledge about home and garden use of PPPs by amateur gardeners in Flanders is quite limited. **Chapter 5** investigates the current PPP handling behaviour by Flemish amateur gardeners (D-R) (**RQ9**) in order to verify if the estimation of the pressure exerted by non-professional PPPs (P) can be improved by having extra insights in the practice (D-R) (**RQ10**).

This chapter has been compiled from:

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**Fevery, D., Houbraken, M., Spanoghe, P., 2016.** Pressure of non-professional use of pesticides on operators, aquatic organisms and bees in Belgium. Science of The Total Environment. 550, 514-521.

**Fevery, D., Houbraken, M., Spanoghe, P., (in preparation).** Current pesticide handling behaviour by amateur gardeners in Flanders (Belgium).

# Abstract

The first part of this chapter describes the identification and allocation of non-agricultural use of PPPs in Belgium conducted in the framework of a pilot study financed by Eurostat, since a lack of knowledge on PPPs used for non-agricultural purposes still exists. The non-agricultural use of PPPs in Belgium was identified and divided into 17 categories (green areas, golf courses, etc.). Every single category corresponds to a specific use, i.e. areas where PPPs are applied, and includes a panel of actors responsible for applying PPPs (= interlocutors) which were identified as well. Given the complexity of relations between interlocutors and categories of PPP use in Wallonia and Flanders, a ranking of the 17 categories according to their importance in terms of PPP use and data accessibility was set up based on expert judgment. This expert judgment combined with statistical tools revealed

the key players in the non-agricultural use of PPPs in Belgium. Private companies of parks and gardens (for public or private institutions), private companies and amateur gardeners are the interlocutors who require a combination of administrative data (for amateur gardeners) with inquiries for all kind of private companies in order to collect data on the non-agricultural use of PPPs. Subsequently, one of the key players in the non-agricultural use of PPPs was further investigated, i.e. non-professional users and more precisely amateur gardeners which were identified in the first part. Whereas professional use of PPPs is more regulated, this is less conventional for non-professional use as only usage advice can be given. The results of the first part of this chapter suggest to use sales figures (administrative data) of non-professional PPPs to determine non-professional PPP use. Based on these results, an attempt was made in the second part of this chapter to estimate the pressure of non-professional use of PPPs on operators, aquatic organisms and bees in Belgium. Both sales figures and three exposure models were used and a classification in non-professional use was made based on type of PPP, application method and on intensity of nonprofessional use. In general, both total usage (kg) and pressure of PPPs decreased for the period 2005 to 2012 due to efforts made by the government and industry. Special attention should be paid to aerosol spray applications and the non-professional use of insecticides. Furthermore, the use of non-professional sales figures is more reliable than usage figures of non-professional PPPs, as the latter are dependent on the representativeness of the surveyed public. However, non-professional sales figures could be supplemented with more detailed and specific information by performing a survey to investigate the current PPP handling behaviour of non-professional users. Knowledge about home and garden use of PPPs by amateur gardeners in Belgium is limited, so the use of PPPs by amateur gardeners in Flanders (Belgium) was investigated based on a questionnaire. The questionnaire was kept concise in order to have the highest response rate possible and was conducted by means of three different survey methods. Survey results illustrated that knowledge of the different hazard symbols on the PPP label is still insufficient. The challenge remains to better inform amateur gardeners by providing easier to understand PPP labels and to provide advice on how to deal with PPPs both in terms of safety for humans and the environment. Most amateur gardeners wear personal protective equipment during preparation and application of PPPs. However, 100% protection should definitely be pursued. About 30% of the respondents still pour rinse water into the sink, a practice that needs to be avoided. The amateur gardener can reduce the PPP pressure on the environment by consciously dealing with surpluses and rinse water of the used PPPs. The results of this chapter also illustrate the importance of collecting additional data by a combination of administrative data with questionnaires in order to be able to estimate non-agricultural PPP use more precisely.

Non-agricultural use

## Introduction

Plant protection products are useful in many professional non-agricultural settings in Belgium, including gardens, parks, public spaces, sport fields and outdoor leisure areas. They also help the functioning of transportation corridors such as road shoulders, airport runways and railway tracks, as well as industrial sites and drainage infrastructure. Furthermore, nonprofessional areas of ornamental plants and lawns also need protection against harmful pests and diseases. Householders and amateur gardeners, known as non-professional users, use products to protect plants, to grow fruits and vegetables and to control weeds that damage paths and drives. Aesthetic reasons for keeping parks, paths, farmyards and gardens free of weeds and plant pathogens also determine PPP use (Spliid et al., 2004; CIEH, 2015; ECPA, 2015). There could be losses from the non-agricultural treated areas to aquatic environments or to other surrounding areas. Surfaces, for example, are often constructed to encourage surface run-off to avoid flooding or for rapid penetration of water: this can result in contamination or damage of nearby wells, ditches, hedges, ground water or sewage systems. Plant protection products applied to golf courses can potentially move to urban areas and also disposal of PPP waste can cause problems. Consequently, the use of PPPs in non-agricultural settings may lead to different environmental issues from their agricultural use. Therefore, it is important to subject non-agricultural use to a separate consideration (Spliid et al., 2004). Unnecessary or abundant use of non-agricultural PPPs and also accidental release can put the health of humans, other organisms, plants and the environment at risk (Rushton and Mann, 2009). Simple but essential safety steps can be applied in order to ensure that non-agricultural PPPs are stored and used safely. Before this can be established, actors responsible of non-agricultural use of PPPs in Belgium have to be identified. In addition, the effective use of non-agricultural PPPs should also be linked to the identified users.

### 1.1 Identification and allocation of non-agricultural PPP use

A lack of knowledge on PPPs used for non-agricultural purposes still exists, so an indent was introduced in Section 6 of Annex II of Regulation (EC) No 1185/2009. According to this indent, Eurostat (Directorate-General of the European Commission located in Luxembourg) takes a lead in identifying the importance of professional non-agricultural use of PPPs. To avoid possible false conclusions on the non-professional and non-agricultural use of PPPs, Eurostat decided to carry out a pilot study on both professional and non-professional non-agricultural use of PPPs. As described in **Chapter 1**, non-professional use of PPPs includes all use where the user is a private person and the aim of the user is not to produce products for the market, i.e. the product is only used in private households. Conversely, professional non-agricultural use of PPPs includes all use of PPPs by private companies, public or semi-public institutions, non-governmental organisations, etc. Agricultural use covers the production of all agricultural products for own consumption is not included in agricultural production. In

Belgium, this study was realised in the framework of the Federal Pesticide Reduction Program as indicated in the federal action 10.4 in Annex I of the Royal Decree of 15 December 2013 (KB 15/12/2013). A consortium was set up between Ghent University (UGent) and Catholic University of Louvain (UCL) in order to promote exchanges and to share experience in collecting data on PPPs in Flanders (UGent) and in Wallonia (UCL). This estimation of non-agricultural use of PPPs in Belgium consists of a collection of Belgian statistics on the used quantities of PPPs including at least the names of active substances and their quantities (expressed in kilograms) used in a given year.

In Belgium, users of PPPs are divided into professional and non-professional users thanks to the 'Separation of approvals' (KB 10/01/2010) by the Federal Government. All PPPs in their commercial form are identified by an authorisation number that is specified in national sales figures. Amateur gardeners belong to non-professional users and are private people who do not produce any products for the market (Directive 2009/128/EC, Article 4). Among professional users, a distinction between agricultural and non-agricultural users will be made thanks to the implementation of the 'System of licensing certificates' for professional users and the recordkeeping of PPP use (Regulation (EC) No 1107/2009). 'System of licensing certificates' is called 'Phytolicence' in Belgium. This is a certificate delivered by the Federal Public Service Health, Food Chain Safety and Environment (FPS) that ensures that all professional use of PPPs is based on sufficient knowledge and proficiency, and that is regularly updated. The knowledge and proficiency required include the ability to recognize occurring problems, seeking long and short-term solutions among the alternatives to chemical pest control and the proper use of PPPs. Only those in the possession of the certificate will be able to buy products for professional use. The phytolicence is mandatory as of 25 November 2015 for all professional users, distributors and advisors. In the future, this system will provide information about the professional use of PPPs (FOD, 2015). Up until now, no uniform information about non-agricultural use of PPPs in Belgium was shared. Figure 5-1 illustrates the current situation regarding users of PPPs in Belgium.



Figure 5-1 Situation regarding users of PPPs in Belgium.

## 1.2 Pressure of non-professional PPP use

A PPP is only available and authorised as suitable for non-professional use when it carries a minimal risk of exposure to both operator and the environment (Grey et al., 2006; KB 10/01/2010). The profile of non-professional PPPs is hardly dangerous, i.e. (highly) toxic or corrosive products are not authorised for non-professional use (De Cock and Knaepen, 2008). Although the unit dose of an active substance used by a non-professional user can never be large, the contribution of non-professional users in the overall use of PPPs is nevertheless considerable as a result of the large number of operators. In 2004, 21.7% of all PPPs was used in non-professional settings in Belgium. In 2005, this amount even increased to 25.4% (Pissard et al., 2005; Van Bol et al., 2007; De Cock and Knaepen, 2008).

The use of PPPs varies between professional and non-professional users. A non-professional user is often not acquainted with the used PPPs or not able to deal with the PPPs in an effective way. A non-professional user often takes fewer precautions or does not read the instructions on the PPP well (Mostin, 2007; De Cock and Knaepen, 2008). An observational study in the United Kingdom found that few participants read the label of PPPs, that they often found it hard to understand and that compliance with instruction was low (Weale and Goddard, 1998). In general, non-professional users of PPPs are less cautious than professional agricultural users (De Cock and Knaepen, 2008; Rushton and Mann, 2009). Recent studies have generated data that identify potential dermal and inhalation exposure during the application of non-professional PPPs (Sanborn et al., 2004; Harrington et al., 2005; Grey et al., 2006; Lessenger, 2006; Rushton and Mann, 2009; Sanborn et al., 2012).

According to Harrington et al. (2005) potential exposure to non-professional PPPs is highest during mixing and loading, and this mainly at the height of the hands and chest. Furthermore, exposure during application is negligible compared to exposure during mixing and loading (van Hemmen, 1992). The product formulation (liquid, powder, granule, etc.) also influences the potential exposure to non-professional PPPs. The use of liquids for example may result in dermal contact, while the use of powders could cause inhalation exposure (Tyvaert et al., 1999). All of these elements can lead to an increase of the health risk for non-professional users (Waichman et al., 2007).

Due to the non-specificity of PPPs and losses during application, a portion of the applied PPP ends up in non-target areas, e.g. surface water (VMM, 2015). The quality of surface water is very important for aquatic life. Too high concentrations of PPPs may be toxic to aquatic organisms. Annex X of the Water Framework Directive specifies a number of priority substances (including some herbicides) that pose a risk to the aquatic environment (Directive 2008/105/EC). Surface water measurements by the Flemish Environment Agency (VMM) indicate that many active substances exceed the water quality standards in Flanders (Belgium), which can lead to acute or chronic effects on aquatic life. Especially herbicides prove to be problematic to aquatic life (VMM, 2015). The portion of PPPs by nonprofessional use in surface waters should be seen in perspective of the professional agricultural use. A study on household glyphosate use and its major metabolite aminomethylphosphonic acid (AMPA) in surface water drains illustrates the contribution of non-professional use. The study concludes that when glyphosate is used correctly, contribution from non-professional users of PPPs is very small compared to professional use (Ramwell et al., 2014).

Bumble bees and honey bees are wide-range pollinators. They are not only essential in ecosystems but also of crucial importance for seed and fruit production in many agricultural crops (Fuchs and Miller, 2004; Parmentier et al., 2014). Given their considerable importance, the apparent global decline of pollinators has led to growing concern (Ghazoul, 2005; Goka, 2010; Potts et al., 2010; Szabo et al., 2012; Parmentier et al., 2014). This decline seems to be a result of several causes, i.e. habitat degradation, pests and diseases, pollution and PPP use (Ghazoul, 2005; Mommaerts et al., 2010; Potts et al., 2010; Szabo et al., 2012; Whitehorn et al., 2012; Parmentier et al., 2014). Although PPPs have a negative impact on bumble bees at the individual or colony level, Szabo et al. (2012) determined that PPPs are not a main contributor to declines of these species when their entire ranges are considered. On the other hand, according to Mommaerts et al. (2010), certain concentrations of PPPs that are not lethal for bees can have a negative influence on their foraging behaviour. Especially neonicotinoid insecticides are known to negatively affect the foraging behaviour of bees (e.g. imidacloprid). These insecticides occur at trace levels in nectar and the pollen of crop plants (Whitehorn et al., 2012). Since 2013, non-professional use of neonicotinoid insecticides is prohibited (Commission Implementing Regulation (EU) No 485/2013).

#### 1.3 Current PPP handling behaviour by amateur gardeners

A study conducted in the United Kingdom investigated the relationship between various socio-demographic characteristics (age, gender, used quantities and application method). Age appeared to have a significant influence on the use of household PPPs: older generations have a significantly higher consumption. According to Steer and Grey (2006), higher educated people use more PPPs compared to less qualified people on average. Grey et al. (2006) stated that a higher socio-economic status is associated with an increased use of PPPs. This survey conducted in the UK on the use of amateur PPPs indicated that 82% of the households used 3 to 4 PPPs on average. Furthermore, the study showed that most PPPs were used in the garden (76%) followed by indoor use (57%). Insecticides in the home environment were the most commonly applied PPPs at 21% of total PPPs. In gardens, slug pellets and insecticides were used with an average of 4.5 applications per year. Fungicides were hardly used. The most commonly used active substances belonged to the pyrethroids, a group of insecticides. The most common weed killer ingredient was glyphosate (Grey et al., 2006). During the 1990s and early 2000s, more than 90% of PPP users from the UK and the United States (US) preferred chemical products above non-chemical alternative methods (Davis et al., 1992; Grieshop et al., 1992; Adgate et al., 2000; Grey, 2003; Grey et al., 2005). This amount is expected to decline in the US to about 75% (Whitmore et al., 1992; Donaldson et al., 2002; Berkowitz et al., 2003). On the other hand, the use of herbicides is expected to increase from 17% (Whitmore et al., 1992) to about 30% of the households (Feagan and Ripmeester, 1999; Donaldson et al., 2002; Grey, 2003; Grey et al., 2006; Steer and Grey, 2006).

As described in 1.2, professional PPP use differs from the non-professional use. An amateur gardener is often not acquainted with the used PPPs or not able to deal with the products in an effective way. According to Grey et al. (2006), the use of PPPs in garden and home environment could undoubtedly be a significant source of exposure for operators and bystanders. Wearing personal protective equipment – and especially gloves – reduce the risk of dermal exposure during mixing and loading (Harrington et al., 2005). Furthermore, occurrence of acute health effects is usually the result of accidental or deliberate misuse of PPPs (Rushton and Mann, 2009). A British study stated that PPPs are often not kept safely (Rushton and Mann, 2009). An American study revealed that the majority of PPPs (70%) are stored inside the house (excluding the basement) and mainly in the kitchen (45%) (Bass et al., 2001). Results of a study carried out by the Belgian Poison Control Centre indicated that 37% of the adults were orally exposed to PPPs during the period 2003-2006. Dermal and inhalation exposure to PPPs were subsequently 29% and 21% respectively. Adults are more exposed to PPPs than children whereas children are more exposed to biocides. From 2003 to 2006, the average oral exposure of PPPs to children was 84.4%. Therefore, PPPs should be kept out of reach of children at all times (Mostin, 2007; De Cock and Knaepen, 2008). In Flanders, the most frequent users of PPPs can be found in the professional agricultural sector. During the period 1990-2012, this sector was responsible for 80% of the PPP use

(MIRA, 2014). However, PPPs are of interest in professional non-agricultural situations. In the period 1990-2005, the non-agricultural use of PPPs (professional and non-professional) exceeded 20% of the total use in Flanders every year. Since 2005, the non-agricultural use of PPPs decreased to about 3% in 2012 (Peeters et al., 2010; MIRA, 2014).

Different sources of PPP information are available for amateur gardeners. The information required to safely use a product can be found on the PPP label. As mentioned before, these labels are sometimes not well understood by amateur gardeners (Weale and Goddard, 1998; Steer and Grey, 2006). In addition, an amateur gardener can also obtain information from family and friends or perform research on the internet. Today, internet is probably the most accessible source of information. However, it is important to be careful with this source of hyper information. The overall amount of available information is growing at an enormous rate and finding the correct information on the internet is becoming a fundamental problem. Plant protection products are available in both specialised garden centres, as well as in do-it-yourself shops and supermarkets. In Belgium, sellers in garden centres are licensed to sell PPPs and provide better advice than in supermarkets. As everyone will need a licence according to the sustainable use legislation, it is expected that all sellers will be aware of what they sell (De Cock and Knaepen, 2008).

Information according to the amateur use of PPPs can be collected in various ways. Face-toface interviews are appropriate to come into contact with the operator and are also quite accurate. The trained personnel can give an explanation to the questions, to achieve a fair and clear answer. Face-to-face surveys, however, are time-consuming and have a smaller geographic area than other data collection methods. Telephone interviews are less timeconsuming and therefore less expensive, for example by avoiding travel costs (Eurostat, 2008). On the other hand, it is increasingly difficult to reach people by phone the last years. A large number of landlines were replaced by mobile phones, making it harder to reach people haphazard. The busy lifestyle of the modern society also negatively affects the provision of information by telephone. Fewer people are willing to make time to respond to a survey (Louwen, 1992; Stoop, 2005; De Leeuw, 2010). Online surveys are inexpensive, time-saving and user-friendly (Bronner et al., 2014). A disadvantage of an online survey however is the low response rate, meaning that participation to the survey is low compared to the huge amount of people who are reached. Survey numbers can be increased to account for the reduction in participation in order to reach the desired number of responses (Eurostat, 2008). Another disadvantage of an online survey is that the reached target group is quite limited. Due to an increasing use of the internet as a communication and information tool, this problem is little by little solved (Fox et al., 2001; Salamonsen et al., 2002; Wright, 2005). Furthermore, these surveys are particularly open to misinterpretation with respondents leaving out specific information or even disregarding part of the survey (Eurostat, 2008).

Non-agricultural use

### 1.4 Objectives

Various studies focus on agricultural use of PPPs whereas research about non-agricultural use is often neglected. First, this chapter identified the non-agricultural users of PPPs in Belgium. Based on expert judgment, the identified non-agricultural users were ranked according to their importance in terms of PPP use and according to data accessibility. Key players were put forward to collect additional data in order to be able to estimate nonagricultural use of PPPs more precisely. Subsequently, one of the key players in the nonagricultural use PPPs was further investigated. Whereas professional use of PPPs is more regulated, this is less conventional for non-professional use as only usage advice can be given. Both administrative data as more detailed information by performing a survey were used to provide an indication of the non-professional PPP use. An attempt was made to estimate the pressure of non-professional PPP use on operators, aquatic organisms and bees in the second part of this chapter. Exposure of operators and the environment to nonprofessional PPP use was illustrated based on sales figures of non-professional PPPs. Furthermore, pressure of non-professional use of PPPs was calculated by using various indicators for the period 2005-2012. Third, knowledge about home and garden use of PPPs by amateur gardeners in Flanders is quite limited. The objectives of the third part were to determine the levels of knowledge, usage and awareness of amateur gardeners in Flanders based on a questionnaire conducted by means of three different methods.

# **Materials and Methods**

### 2.1 Identification and allocation of non-agricultural PPP use

#### 2.1.1 Identification of categories of non-agricultural PPP use and interlocutors

In order to establish a list of actors that contribute to non-agricultural use of PPPs in Belgium, all available information on PPP use on a Belgian level and more precisely in Wallonia and Flanders was collected. An identification of existing data on sales and use of PPPs was accomplished and a determination of known and unknown data was done as well. Furthermore, the results of previous studies on non-agricultural use of PPPs were analysed and potential professional users of PPPs were contacted. Each identified category of PPP use corresponds to a specific use, i.e. areas where PPPs are applied. These categories include a panel of actors responsible for applying PPPs (= interlocutors) which have been accurately identified as well.

#### 2.1.2 Allocation of non-agricultural PPP use

An expert meeting was organised to classify the identified categories of PPP use according to their importance in terms of PPP use and according to data accessibility. The consulted experts belonged to different institutions. One of the experts represented the government. The other experts belonged to the industry (6) and research institutions (2). All of these

experts have many years of experience in PPP sales. Every expert was asked to put all identified categories of PPP use in order of importance in terms of PPP use for the year 2014. The different categories were scored with numbers 1 to 17 (17 categories were identified); 1 indicating the highest importance of PPP use and 17 the lowest importance. Afterwards, the mean of all experts' scores were taken to obtain a ranking of all categories. This ranking helped to highlight the key players in the non-agricultural use of PPPs and to implement some tools (inquiries, data collection, etc.) that are indispensable to the estimation of nonagricultural PPP use. Each category of use includes different interlocutors which correspond to or are representative for the real users of PPPs. Since information on non-agricultural use of PPPs cannot only be obtained by categories of PPP use, interlocutors of PPPs should also be taken into account. Therefore, the order of importance of interlocutors related to the scores attributed by the experts on the importance of categories of use (in terms of PPP use) was also evaluated in Wallonia and Flanders for the year 2014. Based on advice given by Statistics Belgium, a list of interlocutors ranked by importance in terms of PPP use was established. First, the inverse of scores attributed by the experts on the importance of categories of use was calculated for each category. Furthermore, the categories of PPP use were identified for every interlocutor. The results were attributed for each category included in every interlocutor. Subsequently, the total sum of results from different categories of use included in every interlocutor was calculated. Finally, a ranking of results obtained for every interlocutor was made in descending order.

### 2.2 Pressure of non-professional PPP use

# 2.2.1 PPP sales figures

In order to calculate the pressure of non-professional PPP use, data concerning their use were collected. As described in **Chapter 2**, reliable data on usage of PPPs are critical for the development of indicators of the effects of PPPs on humans and the environment. Usage figures of PPPs cover all kinds of data on the actual use of PPPs by operators, but are not always quick and easy to produce. Sales figures of PPPs are relatively simple to collect and fairly inexpensive, but can give rise to confidentiality issues and restrictions on the release and use of data for commercial reasons (Fevery et al., 2015). As in Belgium no usage figures are available for non-professional PPP use, sales figures of 2005 to 2012 were provide an indication of the non-professional PPP use. Sales figures of non-professional PPPs were not available for 2006. Phytofar represents manufacturers and formulators of PPPs (phytosanitary or phytopharmaceutical products) in Belgium. Data included authorisation number, product name, size and number of packages, type of product and name and concentration of the active substances present in the product from which the use was determined. The sales figures represented 90% of the Belgian market of non-professional PPP use.

#### 2.2.2 Indicators

Use indicators are not adequate proxies for assessing pressure exerted by PPP use (Chapter 2). The toxicity of PPPs and their degradation time vary depending on the active substances, so a useful indicator has to take these characteristics into account (Barnard et al., 1997; Levitan, 2000; Van Bol et al., 2003; Tzilivakis et al., 2004; Stenrod et al., 2008). One kilo of one PPP can exert a completely different pressure than one kilo of another PPP. To quantify the risk of exposure to PPPs, it is necessary to weigh the use of PPPs to the acute and chronic toxicity coefficients for the various environmental compartments. Subsequently, an estimation of the exposure of humans and the environment to these risks should be made (Tzilivakis et al., 2004). The risk is estimated by the use of risk indices. A risk index (RI) is determined from the ratio of the calculated exposure (e.g. predicted environmental concentration (PEC)) and the effect standard, i.e. a toxicological reference dose (Equation 5-1). The overall pressure of non-professional PPP use for Belgium was determined by multiplying the risk index with the non-professional use of PPPs based on sales figures, as shown in Equation 5-2.

$$Risk index (RI) = \frac{Exposure}{Effect}$$
 Equation 5-1

Total pressure = risk index (RI) x use

Exposure of operators, aquatic organisms and bees to non-professional PPPs were determined by using exposure models. First, the French amateur garden model (UPJ) was used for the operator exposure (UPJ, 2005). Second, the pressure of non-professional PPP use on aquatic organisms in surface water was based on the Pesticide OCcupational and Environmental Risk indicator (POCER) (Vercruysse and Steurbaut, 2002; Vergucht et al., 2006) and third, a risk assessment model of non-target organisms was used to indicate the pressure on bees (Fytoweb, 2014). Toxicological reference doses from the database of Laboratory of Crop Protection Chemistry (UGent) were achieved for aquatic life. These data are in line with the new official data from the review program for PPPs in the European Union. The following sources for parameter values, ranked as function of importance, were used to create the toxicity database: the authorisation files of the European Union, the Footprint database (Lewis and Green, 2011) and the toxicity database of UGent supplemented with new products until 2009 including the information of the Tomlin Pesticide Manual (BCPC, 2006).

#### 2.2.2.1 Indicator operators

The UK-Predictive Operator Exposure Model (UK-POEM) database is based on a review of the data available on the exposure of PPP spray operators in the United Kingdom. The review indicated that several factors determine the dose absorbed by a spray operator. In the model, the possibility exists to include home garden spray applications and inhalation values for solid formulations. This model has a straightforward structure and is simple to

Equation 5-2

use. However, it includes insufficient information to estimate the pressure of nonprofessional PPPs on operators (EFSA, 2008b). In this study, the amateur (garden) model of 'Union des entreprises pour la Protection des Jardins et Espaces verts' (UPJ) was selected to calculate the risk indicator for the pressure on operators. UPJ sponsored a study to measure operator exposure when using non-professional garden application equipment. This study was conducted according to the Good Laboratory Practices in the South-east of France during autumn 2003 in order to measure the dermal and inhalation exposure of amateur gardeners during various mixing/loading and application tasks known as being representative of treatment activities in gardens. Based on the results of this study on nonprofessional use, a model to assess the exposure of amateur gardeners when applying PPP was developed (UPJ, 2005). The toxicological endpoint, Acceptable Operator Exposure Level (AOEL) and penetration factors, were provided by PPP companies, controlled and supplemented with data from the PPP database UGent.

$$RI_{operator} = \frac{Exposure}{AOEL}$$

Equation 5-3

AOEL = Acceptable Operator Exposure Level (mg/kg BW/day)

#### 2.2.2.2 Indicator aquatic organisms

The indicator used to determine the pressure of non-professional PPP use on aquatic organisms in surface water was based on the Pesticide OCcupational and Environmental Risk indicator (Vercruysse and Steurbaut, 2002; Vergucht et al., 2006). The indicator is based on acceptance criteria formulated in Annex VI of the European Council Directive 91/414/EC. POCER evaluates both human risk from occupational exposure to PPPs and risk to the environment from the use of agricultural PPPs. The assessment of the risk to aquatic organisms is determined by the indicator as described in Pussemier (1999) and Vergucht et al. (2006). The exposure was calculated based on Equation 5-4 and Equation 5-5. The endpoint for aquatic organisms is based on the Maximum Allowable Concentration, which has been described in **Chapter 2** (2.7).

$RI_{aquatic \ life} = \frac{PEC_{aquatic \ life}}{MAC}$	Equation 5-4
$PEC_{aquatic \ life} = PCOW \ x \ (1 - BFI)$	Equation 5-5

PEC<sub>aquatic life</sub> = Predicted Environmental Concentration (mg/l)
MAC = Maximum Allowable Concentration in the environment (mg/l)
PCOW = Predicted Concentration in Outflowing Water (mg/l)
BFI = Base Flow Index, fraction of water not directly linked to rainfall (0.5)

**Equation 5-6** 

## 2.2.2.3 Indicator bees

The risk assessment of bees was performed using a risk assessment model of non-target organisms created by the national government (Fytoweb, 2014), as shown in Equation 5-6. This model allows to evaluate the risk to non-target organisms according to the Uniform Principles. These principles were prepared according to the recommendations by European Directives (Directorate E-Food Safety, 2002a, 2002b). This first step of risk assessment makes it possible to determine if the product presents a risk to non-target organisms (birds, mammals, aquatic organisms, bees, earthworms and soil micro-organisms). The scenario for non-professional use includes drift values based on the drift study of Dekeyser et al. (2007). The lethal dose 50% values were provided by PPP companies, controlled and supplemented with data from the toxicity database of UGent.

$$RI_{bees} = \frac{Conc_{PPP} \ x \ HRD}{LD_{50}}$$

 $Conc_{PPP}$  = PPP concentration (g/kg) HRD = highest registered dose (g/m<sup>2</sup>) LD<sub>50</sub> = lethal dose 50% (µg/bee)

## 2.2.3 Classification of non-professional PPP use

A classification was implemented in the above described indicators to allocate the nonprofessional use of PPPs in Belgium more precisely. This classification was made based on product information and usage advice per non-professional PPP provided by PPP companies supplemented with expert judgment. The type of non-professional PPP and the application method as well as the type of operator were considered.

First, non-professional PPPs in this study were split into three groups, i.e. insecticides, fungicides and herbicides, since these types of PPPs are most commonly used for non-professional use. However, rodenticides, which belong to biocides, were also present. The pressure of these products was not included in this study. The use of iron sulphate to control mosses was included here as a herbicide, as this product was also sold in combination with other herbicides.

Second, the type of application method of the non-professional user varies compared to the professional user. Non-professional PPPs can be applied in various ways, e.g. by using a watering can, a pressure sprayer, a backpack sprayer, a ready-to-use sprayer or granules. In this study, the focus was mainly on manual and spray applications (Figure 5-2). Strewing of powders or granules is considered to be a manual application. Spray applications comprise trigger sprays and aerosol spray cans. Trigger sprays are dispensers turning a liquid into a spray. There are ready-for-use PPP trigger sprays and formulations that should be mixed and loaded in a plant sprayer. By turning around the nozzle of the plant sprayer, the spray distribution can be adjusted, which results in a spray with fine or coarse droplets. Aerosol

spray cans are pressure resistant containers from which a liquid is discharged under the pressure of a propellant. These cans are ready-to-use products.

Finally, the type of operator was divided into three categories which reflect the intensity of application by the non-professional user (Figure 5-2). An occasional operator uses especially PPPs for the treatment of ornamental plants on terrace, while an ordinary operator applies PPPs on lawns, driveways and small ornamental gardens. A non-professional user, which performs a PPP treatment on its lawn, vegetable garden, orchard, large ornamental garden and driveways, is called an intensive operator.



Figure 5-2 Type of application method (trigger, aerosol and manual) and operator type (occasional, ordinary and intensive) used in this study (Picture references: Cole-Parmer, 2016; Phytofar, 2014, Houbraken et al., 2014; Manfung, 2014; Lievens et al., 2014; Art Nouveau, 2016).

### 2.3 Current PPP handling behaviour by amateur gardeners

#### 2.3.1 Survey

The survey of amateur gardeners consisted mainly of multiple choice questions (Table 5-1; Appendix C). In the first part of the survey, the socio-demographic characteristics were investigated, i.e. gender, age group, social class, highest educational qualification and family composition. The postal code was questioned as well to monitor the scope of the survey and to indicate a possible correlation between PPP usage and region. Subsequently, a brief explanation of the term PPP in this context was given. Some questions were supported by

visual aids, for example pictures of PPPs or personal protective equipment. To the people who deliberately choose not to use PPPs, the question was asked why they made that choice. Besides a number of options, the respondent was also given the option to write down his own remarks. Places where PPPs are applied and the number of annual treatments were investigated. Furthermore, attempts were made to reveal which categories of PPPs are used: insecticides, fungicides or herbicides. Products intended for the destruction of mosses belong to herbicides. In this survey, moss killers were considered separately in order to find out the amount of both herbicides and moss killers. Because it is unclear if amateur gardeners are aware of what active substances they use, a number of common active substances were listed with the question on the kind of active substances they recognised. Furthermore, the respondent had the possibility to enter other substances, trade names or PPPs that did not appear in the list. Nowadays, non-chemical PPPs are quite popular. The survey examined if amateur gardeners also select this kind of PPPs. Places where PPPs are purchased were investigated as well. Additionally, the awareness of potential risks caused by the use of PPPs was investigated. Respondents were asked how they protect themselves when applying PPPs and if they follow the instructions on the label of the PPP. The question if an amateur gardener takes into account the pressure of PPPs on the environment into account, was assessed by asking how they handle potential surpluses or rinse water. Finally, the knowledge of the amateur gardener in relation to PPP residues on food (home-grown or bought in store) was examined. An additional question was whether the amateur gardener would adjust its PPP use in the future. Participating in the survey allowed the operator to reflect on his PPP habits. The survey was first subjected to a few test respondents. Afterwards, some improvements were made and a duration of 5 to 10 minutes proved to be the average time for participants to complete the survey. At the launch of the survey, the average duration of the survey was also mentioned.

The survey was conducted by means of three different methods: an online survey, face-toface interviews and telephone interviews. The online survey was distributed via different organisations and companies that were willing to participate in this study, e.g. the Flemish Environment Agency. The service of 'Qualtrics' was chosen as a platform to collect all the information. This is a user friendly medium that can be used to collect and process results. The second method, face-to-face interviews, consisted of approaching people in particular places and filling in the survey under supervision. The location can affect the profile of the participants. In this survey, the interviews were conducted at garden centres and supermarkets in the region of Ghent, Deinze and Dendermonde in Flanders. In this way, people were reached wherein the probability of the possession and application of PPPs were quite high. Telephone interviews were conducted as a third method. Random phone numbers were dialled, questions were asked and clarified where possible. Table 5-1 Questions on amateur gardeners' characteristics, handling, application and awareness related to PPP use.

Question
Social and demographic characteristics
What is your gender/age/postal code/profession/education
Is your profession related to agriculture
Do you have children
PPP handling
Do you use PPPs
How many times a year do you apply PPPs
Is the label on the PPP clear enough
Which amount of PPP do you use during an application
Which type(s) of protective clothing do you wear during application, mixing and loading of PPPs
Where do you buy PPPs
Which factors play a role in the purchase of PPPs
PPP application
How many different products do you use to control insects/mosses/weeds/fungi
Which products do you use
Do you use organic, non-chemical PPPs
Where do you use PPPs
How do you apply PPPs
How do you get rid of the leftover spray liquid
What do you do with rinse water after cleaning the spraying equipment
Amateur gardeners' awareness
How do you feel about residues of PPPs/PPP storage/label of PPPs
How do you remove any residues of PPPs on home-grown fruits and vegetables
Do you think, after completing this questionnaire, your PPP use will change

### 2.3.2 Data analysis

Data processing was performed using SPSS 20.0. Chi-square ( $\chi^2$ ) tests (p < 0.05) were used to compare the differences between the survey methods.

$$\chi^{2} = \sum \frac{(observed \ value - expected \ value)^{2}}{expected \ value}$$
Equation 5-7

# **Results and discussion**

### 3.1 Identification and allocation of non-agricultural PPP use

#### 3.1.1 Identification of categories of non-agricultural PPP use and interlocutors

Table 5-2 illustrates the 17 categories of non-agricultural use of PPPs and 23 interlocutors which were identified. The **first category**, **private areas**, is represented by different actors in Belgium, i.e. amateur gardeners and private companies of parks and gardens. As described in 1.1, amateur gardeners are non-professional users of PPPs. They also refer to individuals

who own pastures for animals (horses, sheep, goats, etc.). Moreover, private areas include areas belonging to private owners that may be open to the public. The second category, green areas, is divided into public and private green areas, and include parks, gardens and cemeteries. This category excludes green areas belonging to private owners that are open to the public (category 1). Several actors responsible for the maintenance of green areas were identified, i.e. municipalities, intermunicipalities, provinces, regions and private companies (of parks and gardens). The third category includes public and private road infrastructure. Road infrastructure refers to motorways, roads, streets, paths, etc. but does not refer to access pathways to public or private buildings which count as part of the infrastructure of buildings, as they belong to category 5. Individuals who maintain their private roads themselves are included in category 1. Actors involved in the use of PPPs on road infrastructure are municipalities, provinces, regions and private companies (of parks and gardens). Transport service areas (category 4) are maintained by several public and private companies that are responsible for trains, buses, underground, touristic tram- or train ways, tramways, planes in the common airports and aerodromes, boats in the ports, bicycles and railbikes. Public and private industrial areas belong to the fifth category. Furthermore, the tertiary sector (commercial, touristic, car park, etc.) is also a part of this group. Several actors are responsible for the maintenance of these areas, i.e. municipalities, intermunicipalities, provinces, regions and private companies (of parks and gardens). Stadium, race courses, tennis courts, etc. belong to the sixth category of public and private sport areas. Fédération Wallonie-Bruxelles, Public Social Assistance Centre and communities are also responsible for the maintenance of these areas besides the same actors (except regions) as in category 2 and 5. Golf courses (category 7) are closely related with sport areas. However, the treatment of PPPs differs in application frequency and in treated surface areas (except for sport areas for high performance). Moreover, golf courses are only private areas maintained by private companies. The eighth category groups military areas, including military fields and airports. Although, military areas are no public or private spaces, they were considered as public spaces in this study since the manager or person in charge of military fields is invited to respect notably the environmental law implemented in Belgium. Regions, Belgian Army, NATO, War Graves Commissions and private companies of parks and gardens maintain these areas. Leisure parks, adventure parks, campsites and miniature golfs form the ninth category (recreation areas) where public and private parks are distinguished as well. These areas are maintained by the same actors as in category 2 and 5. Actors of education are classified differently in Wallonia and Flanders; this is why they are regarded separately. Most schools in Wallonia and Flanders have their own gardeners or subcontract with private companies of parks and gardens (category 10). The type of education (free or official) does not influence the use of PPPs. Anyway, some schools do not use PPPs (only alternative control methods) and others use few PPPs (especially herbicides) on their areas. The Walloon legislation foresees to prohibit the use of PPPs on school areas from June 1, 2018 (AGW 11/07/2013), more specifically inside and less than 50 metres from schools. The Flemish legislation prohibited the use of PPPs on the areas of schools from January 1, 2015

(Decreet 8/02/2013). The eleventh category of public and private health care establishment areas, especially includes rest homes, hospitals, day-nurseries, childcare facilities, etc. Actors involved in the use of PPPs on these areas are the Public Social Assistance Centre, municipalities, provinces, intermunicipalities, communities and private companies (of parks and gardens). The twelfth category is formed by the areas belonging to the Public Social Assistance Centre and housing corporations. These social services include financial or medical help, housing, legal advice, etc. Municipalities, provinces, intermunicipalities, regions, private companies (of parks and gardens) and the Public Social Assistance Centre itself maintain these areas. Areas of public and private water, electricity, gas and phone companies belong to the thirteenth category. These areas are maintained by municipalities, intermunicipalities and private companies (of parks and gardens). The fourteenth category includes public and private forests and nature. A high proportion of private owners maintain their forest themselves, but without necessarily planning management. Their forests may be open to the public. The owners can sometimes rely on private guards and forest experts who ensure the supervision and manage the maintenance of forests. Those private owners are included in category 1. Several actors involved in the use of PPPs in forests and nature, were identified, i.e. municipalities, provinces, intermunicipalities, legal people under public Belgian law, Public Social Assistance Centre, church wardens, regions, federal public companies and private companies (of parks and gardens). Navigable waterways belong to the fifteenth category and are maintained by different waterway managers (public or private companies), each responsible for specific watercourses. The identified actors are municipalities, intermunicipalities, provinces, regions, Fédération Wallonie-Bruxelles, public interest organisations and private companies (of parks and gardens). The Royal Trust is a Belgian independent public body that manages numerous grounds, castles and other buildings that King Leopold II of Belgium gave to the Belgian state in 1900. Private gardening companies supervise weed control for some green areas belonging to this sixteenth category. Finally, public and private immovable heritage form the seventeenth category. In Wallonia, approximately 3350 real estate properties are classified as monuments, sites or archaeological sites. In Flanders, some 80000 real estate properties are classified as architectural heritage, historic parks and gardens, historic organs and World War Heritage. Immovable heritage also refers to cultural buildings, i.e. operas, theatres, etc. Some private owners have real estate properties that may be open to the public. Private owners who supervise weed control themselves are included in the group of private areas (category 1). Municipalities, provinces, intermunicipalities, Fédération Wallonie-Bruxelles, regions, federal public companies, public interest organisations, public interest foundations, Public Social Assistance Centre and private companies (of parks and gardens) were identified as actors involved in the use of PPPs on areas belonging to immovable heritage.

Links between use and interlocutors of PPPs are really diversified and complex. More precisely, 23 interlocutors were identified in Belgium, e.g. municipalities, provinces, regions, private companies of parks and gardens, etc. (Table 5-2). The identification of interlocutors for each category of PPP use shows that some important information on non-agricultural

PPP use can be given due to the help of the interlocutor and not only through the category of PPP use. For example, there are five interlocutors for the category of road infrastructure in Wallonia and Flanders. Road infrastructure is divided into public and private roads. In Flanders, public roads are maintained by municipalities, regions and private companies of parks and gardens whereas private roads are maintained by private companies and private companies of parks and gardens. In Wallonia, the same interlocutors are responsible for PPP use on the roads except for some provincial roads. Private companies in this study refer to non-profit associations, limited companies, limited liability companies, private limited companies, etc. Private companies of parks and gardens are specialised garden companies which maintain both public and private areas.

Category of PPP use	Interlocutor
1. private areas	<u>Private</u>
2. green areas	1. amateur gardeners
3. road infrastructure	2. private companies
4. transport service areas	3. private companies of parks and gardens
5. industrial areas	for private institutions
6. sport areas	<u>Public</u>
7. golf courses	4. municipalities
8. military areas	5. intermunicipalities
9. recreation areas	6. provinces
10. school areas	7. regions
11. health care establishment areas	8. federal public companies
12. areas of Public Social Assistance Centre and	9. communities
housing corporations	10. private companies of parks and gardens
13. areas of water, electricity, gas and phone	for public institutions
companies	11. Fédération Wallonie-Bruxelles
14. forests and nature	12. Belgian Army
15. navigable waterways	13. NATO
16. areas of the Royal Trust	14. War Graves Commissions
17. immovable heritage	15. Public Social Assistance Centre
	16. church wardens
	17. dioceses and religious congregations
	18. autonomous public institutions
	19. confessional schools
	20. non-confessional schools
	21. public interest organisations
	22. public interest foundations
	23. legal people under public Belgian law

Table 5-2 17 categories of non-agricultural use of PPPs and 23 interlocutors which were identified in Belgium.

Belgium is a federal state, comprised of communities and regions. This indicates that decision-making power in Belgium is not centralized but divided between the federal state, three communities and three regions. They are on an equal footing but have powers and responsibilities for different fields. Given the complex federal structure of Belgium, collecting

information to identify the different categories of PPP use and interlocutors is somehow difficult because of the lack of answers from some potential interlocutors and the implementation of new legal measures at federal and regional levels.

## 3.1.2 Allocation of non-agricultural PPP use

## 3.1.2.1 Ranking according to importance in terms of PPP use

Given the complexity of relations between interlocutors and categories of PPP use in Wallonia and in Flanders, the identified categories were ordered according to their importance in terms of PPP use. Table 5-3 illustrates the ranking of categories in terms of PPP use (for the year 2014) according to expert judgment. Private areas, green areas, transport service areas and road infrastructure are among the most important categories of PPP use in Belgium.

Table 5-3	Final	ranking	of	categories	in	terms	of F	PPP	use	obtained	after	discussion	with	several
experts in	2014	for Belgi	um											

Category of PPP use	Mean of scores
1. private areas	1.4
2. green areas	2.9
4. transport service areas	4.0
3. road infrastructure	4.4
5. industrial areas	4.9
6. sport areas	5.4
7. golf courses	6.8
9. recreation areas	8.4
13. areas of water, electricity, gas and phone companies	9.6
8. military areas	10.6
10. school areas	12.0
12. areas of Public Social Assistance Centre and housing corporations	12.7
14. forests and nature	13.1
11. health care establishment areas	13.6
17. immovable heritage	14.1
15. navigable waterways	14.7
16. areas of the Royal Trust	14.7

Information on interlocutors of PPPs helps to reveal the key players in the non-agricultural use of PPPs. Therefore, a list of interlocutors ranked by importance in terms of PPP use was established. All categories of PPP use were identified for each interlocutor. Table 5-4 illustrates the calculation for the interlocutor 'provinces' in Flanders. For this interlocutor, 9 categories of PPP use were identified, e.g. the category green areas. The sum of scores attributed by the experts on the importance of categories of use was for green areas 26 resulting in an inverse value of 3.85. The inverse of the sum of scores was attributed for each interlocutor were ranked in descending order (Table 5-5). According to this ranking, the trend observed

in Wallonia and in Flanders in terms of importance of PPP use related to the interlocutors is that private companies of parks and gardens (for public or private institutions) and private companies are considered as the largest users of PPPs. Amateur gardeners got a score of 8, which means that the use of PPPs can be considered as high. The ranking carried out by the expert panel was based on an ordinal ranking. This means that a category of use for example ranked fourth by the expert panel can use more PPPs than the category ranked fifth. This kind of ranking does not give an idea of used quantities of active substances. The ranking sets priorities and helps to make choices to go into more detail on the implementation of inquiries and to identify interlocutors for whom it seems necessary to collect data on used quantities of active substances.

The category of transport service areas includes nine different transport services. Given that some interlocutors can use PPPs for some transport services only, the maintenance of different types of transport with a large number of interlocutors has to be defined in order to quantify the order of importance of interlocutors related to the previous ranking of categories by experts. According to Statistics Belgium, a solution was to have a global estimation on the used quantities of active substances for each type of transport and to weigh obtained values for each type of transport. Based on the extrapolation of some obtained used quantities of PPPs for the different transport services in Belgium, 90% of used quantities of PPPs allocated to category 4 were attributed to trains and 10% to the other transport services.

Category of PPP use	Inverse of sum of scores (x 10 <sup>-2</sup> )
2) green areas	3.85
5) industrial areas	2.27
6) sport areas	2.04
9) recreation areas	1.32
10) school areas	0.93
11) health care establishment areas	0.82
12) areas of Public Social Assistance Centre and housing corporation	s 0.88
14) forests and nature	0.85
17) immovable heritage	0.79
total	13.73

Table 5-4 Calculation of scores for the interlocuto	'provinces' in Flanders.
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Table 5-5 List of interlocutors ranked by order of importance related to the scores attributed by experts on the importance of categories of PPP use in Wallonia and Flanders in 2014.

Wallonia		Flanders			
Interlocutor	Scores (x 10 <sup>-2</sup> )	Interlocutor	Scores (x 10 <sup>-2</sup> )		
private companies of parks and gardens for private institutions	26.12	private companies of parks and gardens for private institutions	25.37		
private companies of parks and gardens for public institutions	20.24	private companies	19.31		
private companies	20.07	private companies of parks and gardens for public institutions	19.19		
municipalities	18.43	municipalities	16.51		
provinces	16.99	regions	14.53		
intermunicipalities	14.73	provinces	13.73		
regions	12.17	intermunicipalities	13.43		
amateur gardeners	7.69	amateur gardeners	7.69		
Public Social Assistance Centre	5.37	Public Social Assistance Centre	5.37		
Fédération Wallonie-Bruxelles	4.51	federal public companies	4.13		
federal public companies	4.13	communities	3.79		
public interest organisations	1.63	Belgian Army	1.05		
Belgian Army	1.05	War Graves Commissions	1.05		
NATO	1.05	confessional schools	0.93		
War Graves Commissions	1.05	non-confessional schools	0.93		
dioceses and religious congregations	0.93	church wardens	0.85		
church wardens	0.85	legal people under public Belgian law	0.85		
legal people under public Belgian law	0.85	public interest organisations	0.79		
public interest foundations	0.79	public interest foundations	0.79		
autonomous public institutions	0.76	autonomous public institutions	0.76		

#### 3.1.2.2 Ranking according to importance in terms of PPP use, data accessibility and

#### legislative changes

Given the transposition of Directive 2009/128/EC which aims at reducing the risk of PPPs to human health and the environment, all Member States had to set up National Action Plans in which they set quantitative objectives, targets, measures, timetables and indicators. Legal texts in Wallonia and in Flanders were adapted in order to respect these requirements (especially Article 12). Based on expert judgment explained in 3.1.2.1 and advice of Statistics Belgium, a ranking was made to order the 17 categories of PPP use according to their importance in terms of PPP use, data accessibility and the legislative changes in 2014 and in 2020 for Flanders and Wallonia respectively. In Wallonia, the general banning of PPPs will be implemented as of June 1, 2019 in public spaces and as of June 1, 2018 in some specific areas (AGW 11/07/2013; Décret 10/07/2013). Some specific protection measures were defined for vulnerable groups (children, pregnant women, patients, the elderly, etc.) in order to avoid exposure to PPPs. In Flanders, on the other hand, the use of PPPs is already prohibited since January 1, 2015 for all public services (Besluit 19/12/2008; Besluit 15/03/2013; Decreet 21/12/2001; Decreet 8/02/2013). In both Flanders and Wallonia, PPPs can only be used under specific conditions after undergoing a specific procedure. In Flanders, many areas have a minimum use of PPPs, e.g. all areas accessible to the general public or to vulnerable groups which are not owned by the government or managed by a public service. This means that a reduction in PPP use in this field should be defined; PPPs may only be used in spots (not the surrounding area). Where possible, alternative nonchemical methods should be used and only authorised PPPs may be applied. Application requirements must strictly be respected as well. If an area is (re)constructed, the design should be tested against a control without PPPs. Therefore, in the design of some new areas, a PPP-free management should be taken into account. From the period 2014 to 2020, the use of PPPs will be prohibited in some identified areas (categories of PPP use) and a lot of identified interlocutors will not use PPPs anymore. Many of them will have already switched to alternative control methods. Also, the national quantities of active substances attributed to non-agricultural professional users will probably decrease over time and will be allocated to the remaining categories, involving that categories of PPP use that have been ranked as not important in terms of PPP use in 2014 might be considered as important in terms of PPP use in 2020. The transposition of Directive 2009/128/EC will influence the estimation on the used quantities of PPPs according to the years. For most categories, the used quantities of PPPs will decrease strongly over time. The targets and measures deriving from Directive 2009/128/EC will influence a huge change in the plant protection practices in the future.

Table 5-6 illustrates the ranking of categories of PPP use for the current situation (2014) and future situation (2020) in Wallonia and in Flanders related to the importance in terms of PPP use and data accessibility (always based on the expert judgment explained in 3.1.2.1). In both Flanders and Wallonia, some categories of PPP use are restricted to private areas in 2020 and category 1, private areas, is considered as a category important in terms of PPP use

in 2014 and 2020. For many categories, data on PPP use from private companies and private companies of parks and gardens (working for public or private institutions) are not readily accessible, but still quite important. The sold quantities of PPPs for non-professional use (based on authorisation number) are not equal to the used quantities because of the existing stocks. These existing stocks in the warehouses of amateur gardeners can be an important bias in the quantitative estimation of use by amateur gardeners belonging to the category of private areas (category 1). Most amateur gardeners store significant quantities of PPPs over time in their warehouse. The problem of existing stocks is highest when looking at annual figures but is quite limited on the medium term (3-5 years). The used quantities of PPPs are partly available for military areas (category 8). Only the quantities of PPPs used by the Belgian Army were obtained (including the stored quantities of PPPs) in the framework of this study. In Flanders, data are readily accessible for water companies (category 13). These companies are municipal, intermunicipal or provincial mixtures and belong to public services. They are obliged to record their use of PPPs (Besluit 19/12/2008; Besluit 15/03/2013; Decreet 21/12/2001; Decreet 8/02/2013). In Flanders, the use of PPPs in forests is restricted since 1990 (Decreet 13/06/1990). In special cases some deviations are approved to allow the use of PPPs. These data are readily accessible. In Wallonia, the use of herbicides, fungicides and insecticides is prohibited in forests (Décret 15/07/2008), except in some cases laid down by the Walloon Government since 2009 (AGW 27/05/2009). In Flanders, all areas maintained by private companies that are not accessible for the general public, have no ban on the use of PPPs. Areas maintained by private companies that are accessible for the general public, should have a minimum use of PPPs. Table 5-7 lists the eliminated categories of PPP use (2020) and the cause of elimination.

Table 5-6 Ranking of categories (Cat.) of PPP use for the current (2014) and future situation (2020) in Wallonia and in Flanders related to the importance in terms of PPP use and data accessibility (based on expert judgment).

Current situation (2014)						
	Wallonia		Flanders			
	Important	Not important	Important	Not important		
Data readily accessible	Cat. 1: private areas Cat. 7: golf courses Cat. 8: military areas		Cat. 1: private areas Cat. 2: green areas Cat. 3: road infrastructure Cat. 4: transport service areas Cat. 6: sport areas Cat. 7: golf courses Cat. 8: military areas	Cat. 12: areas of Public Social Assistance Centre and housing corporations Cat. 14: forests and nature Cat. 15: navigable waterways		
Data not readily accessible	Cat. 2: green areas Cat. 3: road infrastructure Cat. 4: transport service areas Cat. 5: industrial areas Cat. 6: sport areas Cat. 9: recreation areas	Cat. 10: school areas Cat. 11: health care establishment areas Cat. 12: areas of Public Social Assistance Centre and housing corporations Cat. 13: areas of water, electricity, gas and phone companies Cat. 14: forests and nature Cat. 15: navigable waterways Cat. 16: areas of the Royal Trust Cat. 17: immovable heritage	Cat. 5: industrial areas Cat. 9: recreation areas Cat. 13: areas of water, electricity, gas and phone companies	Cat. 10: school areas Cat. 11: health care establishment areas Cat. 16: areas of the Royal Trust Cat. 17: immovable heritage		
		Future situation (202	0)			
	Wallonia		Flander	s		
>	Important	Not important	Important	Not important		
Data readily accessible	Cat. 1: private areas		Cat. 1: private areas Cat. 7: golf courses			
Data not readily accessible	Cat. 4: transport service areas Cat. 5: industrial areas (private) Cat. 12: areas of housing corporations (private) Cat. 13: areas of water, electricity, gas and phone companies	Cat. 3: road infrastructure (private) Cat. 15: navigable waterways (private) Cat. 17: immovable heritage (private)	Cat. 2: green areas (private) Cat. 3: road infrastructure (private) Cat. 4: transport service areas (private) Cat. 5: industrial areas (private) Cat. 6: sport areas (private) Cat. 9: recreation areas (private) Cat. 13: areas of water, electricity, gas and phone companies (private)	Cat. 12: areas of Public Social Assistance Centre and housing corporations (private companies) Cat. 17: immovable heritage (private)		

Table 5-7 List of eliminated categories (C	Cat ) of PPP use in 2020 with corresponding	t cause of elimination for Wallenia and Elanders
	cally of FFF use in 2020 with corresponding	g cause of eminination for wanonia and handers.

Wal	llonia	Flanders				
Eliminated category	Cause of elimination	Eliminated category	Cause of elimination			
Cat. 2: green areas	Zero-use of PPPs as of June 1, 2019 in all	Cat. 2: green areas (public)	Zero-use of PPPs as of January 1, 2015 for all			
Cat. 6: sport areas	public areas and as of June 1, 2018 in parts	Cat. 3: road infrastructure (public)	public services. PPPs can only be used under			
Cat. 7: golf courses	of parks, gardens, green areas, sport and	Cat. 4: transport service areas (public)	specific conditions after undergoing a			
Cat. 9: recreation areas	recreation grounds which are used by the	Cat. 5: industrial areas (public)	specific procedure (Besluit 19/12/2008;			
	general public and which are not included in	Cat. 6: sport areas (public)	Besluit 15/03/2013; Decreet 21/12/2001;			
	public areas (AGW 11/07/2013; Décret	Cat. 8: military areas	Decreet 8/02/2013).			
	10/07/2013).	Cat. 9: recreation areas (public)				
		Cat. 10: school areas				
Cat. 10: school areas	Zero-use of PPPs as of June 1, 2018 in some	Cat. 11: health care establishment areas				
Cat. 11: health care establishment areas	specific areas (AGW 11/07/2013).	Cat. 12: areas of Public Social Assistance				
		Centre and housing corporations				
Cat. 3: road infrastructure (public)	Zero-use of PPPs as of June 1, 2019 in all	Cat. 13: areas of water, electricity, gas and				
<b>Cat. 4:</b> transport service areas (public)	public spaces (Décret 10/07/2013).	phone companies (public)				
Cat. 5: industrial areas (public)		Cat. 15: navigable waterways				
Cat. 8: military areas		Cat. 16: areas of the Royal Trust				
<b>Cat. 12:</b> areas of Public Social Assistance		Cat. 17: immovable heritage (public)				
Centre and housing corporations (public)						
<b>Cat. 15:</b> navigable waterways (public)						
Cat. 16: areas of the Royal Trust						
Cat. 17: immovable heritage (public)						
Cat. 14: forests and nature	The use of herbicides, fungicides and	Cat. 14: forests and nature	The use of PPPs in forests is restricted since			
	insecticides is prohibited, except in some		1990 according to the decree that has been			
	exceptional cases laid down by the Walloon		adapted in 2014 (Decreet 13/06/1990). In			
	Government since 2009 (AGW 27/05/2009;		special cases some deviations are approved			
	Décret 15/07/2008).		to allow the use of PPPs. These data are			
			readily accessible.			
Non-agricultural use

#### 3.1.3 Discussion

To collect more information on non-agricultural use of PPPs, data collection methods should be implemented. Some categories of non-agricultural use of PPPs do not require the implementation of a data collection method, since these groups are considered as not important in terms of PPP use by the expert judgment and data for that category are not readily accessible (Table 5-6). Emphasis is put on the categories considered as important in terms of PPP use and where data was made readily accessible, i.e. private areas, golf courses, and military areas. More specifically, a survey on the use of stocks for amateur gardeners should be implemented in order to have a better knowledge of the real used quantities of PPPs. Data on used quantities of active substances for golf courses can be obtained for Belgium by the Belgian greenkeeper's association (Greenkeeper, 2013). For military areas, quantities of PPPs used by private companies of parks and gardens could be obtained due to data collected in the record-keeping for PPP use which is imposed to all professional users (Regulation (EC) No 1107/2009). According to the requirements of Directive 2009/128/EC, the ranking of categories of PPP use based on the scores attributed by the expert panel and the calculation evaluating the degree of importance of different interlocutors, it appears that the implementation of inquiries makes sense for four major interlocutors of PPPs, i.e. amateur gardeners, private companies, private companies of parks and gardens (working for public or private institutions) and private companies for water, electricity, gas and phone. These interlocutors will stay important in terms of PPP use for the future too.

The estimation of non-agricultural use of PPPs has to consist of a collection of Belgian statistics on the used quantities of PPPs including at least the names of active substances and their quantities (expressed in kilograms) used in a given year. In order to pursue this aim of the pilot study, different proposals on the implementation of inquiries for the key interlocutors were developed with the assistance of Statistics Belgium. Data collection methods for each type of interlocutor were divided into three types of investigations for Wallonia and Flanders, i.e. exhaustive investigations, investigations into sample and administrative data. First, data on PPP use for amateur gardeners can be approached by sales figures of PPPs from FPS allocated to the amateur gardeners calculated thanks to the 'Separation of approvals' (KB 10/01/2010). Sales figures of PPPs correspond to administrative data. From a statistical point of view, the implementation of inquiries for amateur gardeners through 'investigation into sample' is considered as inappropriate in comparison with the advantages that could be gained. The added value of such an inquiry would be limited to stocks, regional distribution and consolidation of already obtained results. Second, the implementation of inquiries for private companies should be done by investigations into sample. In order to sample this group of users, an interesting tool would be the Central Balance sheet Office which collects annual accounts of almost all Belgian companies that pursue a professional activity in Belgium (Balanscentrale, 2014). If the focus is on data from the Central Balance sheet Office, it is essential to create groups and to set a

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minimum threshold for capital asset expressed in monetary terms for each group. The sample has to include 1% of Belgian companies. This 1% is a global rate and is the result of a trade-off between supposed representativeness (very rough estimate) and feasibility. The response rate is expected to be low (inferior to 10%). It is also important to determine who is in charge of the establishment of the inquiries, on what basis and whether this was authorised, legitimate and transparent. Inquiries should be in the form of paper questionnaires. Work with some organisations could be useful too. Third, the implementation of inquiries for private companies of parks and gardens should be performed by exhaustive investigations. All private companies of parks and gardens should be questioned. The Belgian Central Enterprise Databank (KBO/CBE, 2014) includes official company information like the type of enterprise (legal people, sole traders, foreign companies, etc.), the establishment units of enterprises (a place where, or wherefrom activities of the enterprise are exercised) and some general information (name, address, legal functions, etc.). For the inquiries, professional activities of each Belgian company could be really useful in order to identify the potential private companies of parks and gardens. In the Belgian Central Enterprise Databank, the professional activities are specified in the form of codes according to the statistical classification of economic activities in the European Community (NACE-BEL, 2014). The response rate is expected to be low. The inquiry should be anonymous and in the form of paper questionnaires or web surveys. The indication of annual turnover should be mentioned. Another option would be to implement a survey that is not anonymous. In this case, if the response rate is not sufficient, a reminder will be sent which would specify that the inquiry is anonymous. It is also important to determine who is in charge of the establishment of the inquiries, on what basis and whether this was authorised, legitimate and transparent. Fourth, the implementation of inquiries for private companies for the category of water, electricity, gas and phone companies should be performed by exhaustive investigations. The procedure to sample this group is similar to the recommendations developed for inquiries for private companies of parks and gardens. A weighting scheme (with extrapolation) should be implemented in function of the number of answers received. Furthermore, additional information about the Belgian professional users of PPPs will be provided by the phytolicence (FOD, 2015).

# 3.2 Pressure of non-professional PPP use

# 3.2.1 Use of PPPs by non-professional users

Figure 5-3 provides an overview of the total quantity of non-professional PPPs sold in Belgium over a period of 2005 to 2012. The total non-professional PPP use in Belgium was approximately 2 million kg PPPs per year for 2005-2007. A sharp decline is visible from 2008 to 2010, resulting in a non-professional PPP use of about 250000 kg per year in 2010-2012. The decline is largely due to a drop in the use of sodium chlorate (NaClO<sub>3</sub>) and iron sulphate (FeSO<sub>4</sub>) in PPPs for non-professional use, as illustrated in Figure 5-3. The non-professional use of PPPs other than sodium chlorate and iron sulphate has remained largely constant and even shows a slight increase.



Figure 5-3 Total use of PPPs  $\blacktriangle$  by non-professional users in Belgium for the period 2005-2012 (no data for 2006; without NaClO<sub>3</sub>  $\bigcirc$ ; without NaClO<sub>3</sub> and FeSO<sub>4</sub>  $\diamondsuit$ ).

# 3.2.2 Pressure of non-professional PPP use on operators, aquatic organisms and bees

The overall pressure of non-professional use of PPPs was calculated by multiplying the non-professional use of PPPs based on sales figures with the specific risk index of operators, aquatic organisms and bees. Figure 5-4 shows the evolution in percentage of the pressure of non-professional PPP use on operators, aquatic organisms and bees in Belgium for the period 2005 to 2012. Over 7 years, pressure on aquatic life due to non-professional use of PPPs was reduced by 20%, pressure on operators and bees was respectively reduced by 55% and 60%.

# 3.2.2.1 Pressure of non-professional PPP use on operators

Table 5-8 presents the pressure of non-professional PPP use on operators in Belgium by type of PPP, application and operator type. The use of insecticides indicated the highest pressure of non-professional PPP use on operators: it dropped by 25% in the period 2005 to 2009. Pressure due to the use of herbicides and fungicides also decreased by 80% in this period. The use of an aerosol spray can application method exerts the most influence on operators and is responsible for 89% to 94% of operator exposure. This pressure decreased by 55% over the period 2007 to 2012 and the pressure exerted by a manual application dropped by 30%. Intensive operators cause the highest pressure of non-professional PPP use on operators (94-97%). This pressure was reduced by 51% over the period 2005 to 2012.

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Figure 5-4 Decrease in percentage of the pressure of non-professional PPP use on operators  $\blacktriangle$ , aquatic organisms  $\blacksquare$  and bees  $\bigcirc$  in Belgium for the period 2005-2012 regarding the reference year of 2005 (no data for 2006).

Pressure (x 10 <sup>6</sup> )					Oper	ators			
		2005	<b>2006</b> <sup>a</sup>	2007	2008	2009	2010	2011	2012
type of PPP	herbicides	17.2	-	14.2	4.4	3.4	3.3	2.4	2.5
	fungicides	4.2	-	2.9	0.8	0.6	1.2	1.0	0.8
	insecticides	247.3	-	258.8	226.3	185.7	142.9	142.3	127.2
	total	268.7	-	275.9	231.5	189.7	147.3	145.7	130.5
type of application	aerosol	253.6	-	257.1	213.8	170.9	131.0	134.9	117.8
	manual	15.1	-	18.7	17.7	18.8	16.3	10.8	12.7
	trigger	0.0	-	0.0	0.0	0.0	0.0	0.0	0.0
	total	268.7	-	275.9	231.5	189.7	147.3	145.7	130.5
type of operator	occasional	9.8	-	6.1	2.5	2.7	2.0	1.8	3.7
	ordinary	5.4	-	10.7	3.9	3.0	3.1	2.3	2.4
	intensive	253.6	-	259.1	225.1	183.9	142.2	141.6	124.4
	total	268.7	-	275.9	231.5	189.7	147.3	145.7	130.5

Table 5-8 Pressure of non-professional PPP use on operator by type of PPP, application and operator in Belgium for the period 2005-2012.

<sup>a</sup>Sales figures of non-professional PPPs were not available for 2006.

#### 3.2.2.2 Pressure of non-professional PPP use on aquatic organisms

Table 5-9 shows the pressure of non-professional PPP use on aquatic organisms in Belgium by type of PPP, application and operator type. Pressure on aquatic organisms showed a decrease of 20% over the period and each type of PPPs (insecticides, herbicides, fungicides) showed a slight decrease as well. The use of herbicides exerts the highest pressure on aquatic organisms followed by insecticides and fungicides. However, the decrease in pressure in 2008 and the increased pressure in 2009 were largely attributable to the use of insecticides. The use of aerosol spray can applications mainly determines the pressure of non-professional PPPs on aquatic organisms. Manual and trigger applications show a much lower pressure. The decrease in pressure in 2008 is explained by a decline in aerosol spray can applications. The increase in 2009 was mainly due to an increase in manual applications. In 2005, the highest pressure (69%) of non-professional PPP use was caused by an ordinary operator. Pressure caused by an intensive operator was 30% and by an occasionally operator only 1%. 77% of the pressure of non-professional PPPs on aquatic organisms was induced by an ordinary operator, 22% and 1% by an intensive and occasional operator respectively.

Pressure (x 10 <sup>3</sup> )				Α	quatic o	rganism	S		
		2005	<b>2006</b> <sup>a</sup>	2007	2008	2009	2010	2011	2012
type of PPP	herbicides	9.4	-	9.4	8.8	9.5	9.6	9.2	8.6
	fungicides	0.4	-	0.4	0.4	0.4	0.3	0.1	0.1
	insecticides	3.8	-	3.6	1.8	4.7	4.0	2.5	2.4
	total	13.7	-	13.4	11.0	14.7	13.8	11.7	11.1
type of application	aerosol	11.7	-	11.6	9.6	10.5	10.1	9.3	8.7
	manual	1.9	-	1.8	1.4	4.1	3.6	2.4	2.3
	trigger	0.0	-	0.1	0.1	0.1	0.1	0.1	0.0
	total	13.7	-	13.4	11.0	14.7	13.8	11.7	11.1
type of operator	occasional	0.2	-	0.2	0.2	0.3	0.3	0.1	0.0
	ordinary	9.4	-	9.4	8.8	9.5	9.5	9.1	8.6
	intensive	4.1	-	3.8	2.1	4.9	4.0	2.5	2.4
	total	13.7	-	13.4	11.0	14.7	13.8	11.7	11.1

Table 5-9 Pressure of non-professional PPP use on aquatic organisms by type of PPP, application and operator in Belgium for the period 2005-2012.

<sup>a</sup>Sales figures of non-professional PPPs were not available for 2006.

#### 3.2.2.3 Pressure of non-professional PPP use on bees

Pressure of non-professional PPP use on bees by type of PPP, application type and type of operator is shown in Table 5-10. Total pressure on bees is almost entirely caused by the use of insecticides. An increase in insecticide use in 2010 provided an almost proportional increase in the overall pressure. Although, the pressure of the use of herbicides and fungicides decreased by 85% in the period 2005 to 2012. The decline of the pressure on bees is mainly due to a decreasing use of the active substance imidacloprid, followed by chlorpyrifos, sodium chlorate and spinosad (authorised from 2007), as illustrated in Figure 5-5. The highest pressure of non-professional use on bees is attributed to the aerosol spray can application method and decreased over the period 2005-2012. The pressure of manual and trigger applications decreased slightly. The pressure of non-professional PPP use by an intensive operator is the highest, followed by an occasional operator. The ordinary operator indicates the lowest pressure on bees.

Pressure (x 10 <sup>6</sup> )					Be	es			
		2005	<b>2006</b> <sup>a</sup>	2007	2008	2009	2010	2011	2012
type of PPP	herbicides	27.2	-	24.0	16.4	4.1	1.8	4.7	2.9
	fungicides	3.7	-	3.1	3.0	1.8	1.2	0.5	0.6
	insecticides	257.0	-	156.4	169.7	143.1	199.0	137.5	108.7
	total	287.8	-	183.6	189.1	149.0	202.0	142.6	112.3
type of application	aerosol	227.7	-	131.0	149.8	100.0	159.9	107.8	78.4
	manual	55.8	-	49.8	36.7	48.2	41.6	31.6	32.1
	trigger	4.3	-	2.8	2.6	0.8	0.5	3.2	1.7
	total	287.8	-	183.6	189.1	149.0	202.0	142.6	112.3
type of operator	occasional	34.8	-	72.3	59.8	48.4	32.7	49.5	36.7
	ordinary	24.2	-	21.8	14.3	2.7	1.7	4.6	2.8
	intensive	228.8	-	89.5	115.0	97.9	167.6	88.5	72.8
	total	287.8	-	183.6	189.1	149.0	202.0	142.6	112.3

Table 5-10 Pressure of non-professional PPP use on bees by type of PPP, application and operator in Belgium for the period 2005-2012.

<sup>a</sup>Sales figures of non-professional PPPs were not available for 2006.



Figure 5-5 Total pressure  $\blacktriangle$  of non-professional PPP use on bees and more specific of the active substances: imidacloprid  $\blacksquare$ , chlorpyrifos x ,sodium chlorate  $\bullet$  and spinosad  $\blacklozenge$  in Belgium for the period 2005-2012 (no data for 2006).

# 3.2.3 Discussion

Based on sales figures provided by PPP companies, the pressure of non-professional PPP use on operators, aquatic organisms and bees in Belgium was determined. In general, both total usage (kg) and total pressure of non-professional PPPs decreased for the period 2005 to 2012. In 2005, the total amount of PPPs sold in Belgium was approximately 8.4 million kg (FPS, 2012) of which 2 million kg (25%) was used in non-professional settings (Pissard et al., 2005; Van Bol et al., 2007; De Cock and Knaepen, 2008). The use of non-professional PPPs declined to 250000 kg in 2012 whereas the total use of PPPs in Belgium only decreased with 25% (FPS, 2012). The sharp decline in total use (kg) of non-professional PPPs is due to a decrease in use of two products, i.e. iron sulphate and sodium chlorate. Iron sulphate is especially used as moss killer and experienced a drop in use in the period 2005-2008. This decrease in use of iron sulfate may result from the fact that iron sulphate is no longer seen as a PPP in its own right. Furthermore, iron sulphate – not sold as a PPP – is easily available and at a lower price in many commercial areas. In addition, given the high price of iron sulphate, it may seem more attractive for individuals to focus on other alternative strategies (e.g. liming), which are less expensive than the use of PPPs. Some commercial products based on iron sulphate were also removed from the Belgian market (Lievens et al., 2012). Sodium chlorate was an extensively used weed killer within Belgium, up until 2009 when it was withdrawn after a decision made under terms of EU Regulations (No 1107/2009).

As mentioned before, one kilo of one PPP can exert a completely different pressure than one kilo of another PPP. To quantify the risk of exposure to PPPs, it is necessary to weigh the use of PPPs to the toxicity coefficients for the various environmental compartments. Therefore, toxicity coefficients of the active substances exert an influence on the results of the indicator (Chapter 2). An active substance can be less toxic to humans and the environment even if large amounts of the active substance are used, e.g. sodium chlorate and iron sulphate. These two products were especially available in granules, which leads to a lower exposure compared to other formulations (De Cock and Knaepen, 2008). The decrease in pressure of non-professional PPP use is hereby not proportional to the decrease in the overall use or the corrected usage. Despite the fact that a large amount of iron sulphate and sodium chlorate was used as non-professional PPPs in the period 2005-2009, it did not cause the overall drop in pressure. In the period 2009-2012, sodium chlorate and iron sulphate no longer influenced the total non-professional PPP use (Figure 5-3). On the other hand, the pressure of non-professional PPP use on operators, aquatic organisms and bees experienced a decline over the entire period (Figure 5-4). The reduced pressure is partly due to a change in the behaviour of the non-professional user by using products that are less toxic, but it is also influenced by an increasing use of safer application methods. Since 2015, the Belgian nonprofessional user can only buy PPPs intended for non-professional use. These PPPs are only available and authorised when they carry a minimal risk of exposure to both operator and the environment (KB 10/01/2010).

For the period 2005 to 2012, pressure of non-professional PPP use on operators, aquatic organisms and bees decreased by 55%, 20% and 60% respectively. Pressure of non-professional use on operators is highest for intensive operators. The use of insecticides causes the highest pressure on operators followed by the use of herbicides and fungicides. The use of aerosol spray can applications exerts the most influence on operators. Pressure of non-professional PPPs on aquatic life is mainly caused by the use of herbicides. The aerosol spray can application method causes the highest pressure whereas the trigger application

hardly effects the total pressure. The ordinary operator provides most pressure on aquatic species. Pressure of non-professional PPPs on bees is mainly caused by the use of insecticides, especially the active substance imidacloprid. The highest pressure on bees is attributed to the aerosol spray can application method and especially an intensive operator determines the pressure of non-professional PPPs on bees. Insecticides are used to kill insects but they also kill or harm other creatures in addition to those they are intended to kill. This explains why the use of insecticides exerts the pressure on operators and bees. Herbicides prove to be problematic to aquatic life (VMM, 2015) which corresponds to the obtained results (3.2.2.2). In this study, especially aerosol applications caused the highest pressure on operators, aquatic organisms and bees followed by manual and trigger applications. During application, aerosol sprays induce an aerosol cloud of very small to small droplets. Parts of these aerosol cloud can be inhaled by the operator. In addition to inhalation exposure, the operator can also be dermally exposed to the product. Residues may end up on the body while spraying. The aerosol cloud can also make contact with other organisms, e.g. bees. Furthermore, aerosol applications are difficult to confine to target site or pest. All these characteristics provide the high pressure on operators, aquatic organisms and bees by the use of an aerosol (3.2.2). During application of powders, exposure may occur through inhalation of atomised particles. Dermal exposure (esp. the hands) can occur during the treatment of desired areas. Residues are easily moved off the target by air movement or water and powders will not stick to surfaces as well as liquids. Granular formulations are similar to powders except particles are larger and heavier. Drift hazard is low and particles settle quickly. Granules cause little exposure to operator (only little dust), but they may be hazardous to non-target species especially waterfowl and other birds that mistakenly feed on the seed-like granules. In general, manual applications cause a lower exposure for operators, aquatic organisms and bees compared to aerosol applications. For many home and garden PPP applications, the best choice is to purchase a ready-to-use product in a trigger pump type of sprayer. Trigger applications are excellent for spot treatments and induce a lower exposure for operators, aquatic organisms and bees compared to aerosol and manual applications (3.2.2) (Bremmer et al., 2006; Fishel, 2010). The classification of operator type also reflects logical results. An intensive operator applies PPPs on a wide range of target areas which results in an increase of exposure for operators and bees. Lawns, driveways and small ornamental gardens are especially treated with herbicides which exert the highest pressure on aquatic organisms. These areas are maintained in this study by ordinary operators which results in an increase of exposure for aquatic organisms. In this study, classification of non-agricultural PPP use was based on product information and usage advice per non-professional PPP provided by PPP companies. In particular, the classification of operator type could be supplemented with more detailed and specific information by performing a survey to investigate the current PPP handling behaviour of non-professional users (1.3).

The use of sales figures includes an uncertainty regarding the time at which the pressure takes place. On the one hand, plant protection products purchased by non-professional

operators are not always completely utilised in the same year. On the other hand, products that were previously purchased may still be used in subsequent years. However, in this study the use of non-professional sales figures is more reliable to estimate the pressure of nonprofessional PPP use than usage figures of non-professional PPPs, as the latter are dependent on the representativeness of the surveyed public. The formulas used are based on a correct use of non-professional PPPs. It is not verified if a non-professional user actually follows the guidelines present on the package of the products (correct dose, distances from waterways, use of gloves and appropriate clothing, etc.) and applies the products correctly. Pressure by the incorrect use of PPPs can in this way be higher than estimated. The pressure of non-professional PPP use on aquatic organisms, operators and bees is expected to further decrease in the future due to changing legislation. For example, thanks to the 'Separation of approvals' (KB 10/01/2010), a distinction can be made between professional and nonprofessional PPPs. All PPPs in their commercial form are identified by an authorisation number that is specified in the national sales figures. This source of administrative data is more accurate to estimate the pressure of non-professional PPP use in the near future. Furthermore, the calculated pressure exerted on operators, aquatic organisms and bees could be put into perspective with that incurred by professional PPP users, such as farmers. These data are not yet available at Belgian level but would provide useful information to indicate the total pressure of PPP use on humans and the environment in Belgium.

# 3.3 Current PPP handling behaviour by amateur gardeners

A very low response rate was obtained by telephone interviews (n = 7). Due to the lack of valuable information, the focus of this study was on online surveys and face-to-face interviews.

# 3.3.1 Social and demographic characteristics

Table 5-11 illustrates the social and demographic characteristics of both online and face-toface respondents. 862 people were willing to complete the online survey, of which a small majority of women (57%). 45% and 38% of the respondents belong to the age groups of 21-40 and 41-60 years respectively. The number of people aged over 60 years was lower but still considerably high with 11% of the total number of respondents. The remaining 6% of the online survey was completed by people younger than 20 years. The respondents lived across Flanders, with peak concentrations in the provinces of West and East Flanders (Figure 5-6). One fourth of the online participants performed a profession that is related to agriculture. When performing the face-to-face interviews, 92 people were surveyed of which 58 men and 34 women. The average age of respondents was higher than those of the online survey. Only 1% was younger than 20 years; 24% had an age between 21 and 40 years. The age group of 41-60 years was represented by 46% whereas 29% of the respondents was older than 60 years. Mainly residences near the drop-off locations of the surveys were determined, i.e. Gent, Deinze and Dendermonde (Figure 5-6). For 85% of the respondents, the professional activity was not related to agriculture.



Figure 5-6 Geographical distribution of online (left) and face-to-face (right) respondents.

Table 5-11 Social and demographic characteristics of both the online and face-to-face respondents (gender, age and education).

Question	Variables	Onl	line survey	Face-to-fa	ce survey
		n	%	n	%
gender	male	374	43	58	63
	female	488	57	34	37
age	< 21 years	48	6	1	1
	21-30 years	235	27	10	11
	31-40 years	157	18	12	13
	41-60 years	331	38	43	46
	< 61 years	92	11	27	29
highest education	primary school	40	5	16	17
	secondary school	227	26	19	20
	high school	253	30	30	32
	university	337	39	28	30

n = number of respondents.

# 3.3.2 Amateur gardeners' knowledge and behaviour related to PPP handling

Table 5-12 illustrates the amateur gardeners' knowledge and behaviour related to PPP handling. 66% of the face-to-face respondents confirmed the use of PPPs as compared to only 47% during the online survey. Face-to-face interviews were conducted at garden centres. Since two different populations were surveyed, the results of the questionnaire were discussed separately, unless distinction made no difference. Several reasons were identified why no PPPs are used. The negative impact on the environment due to the use of PPPs was for the remaining participants in both the online (OS) and face-to-face (FTF) survey the main reason for not using PPPs at all. 'Possible risks for human health' and 'the use of PPPs is unnecessary', were also frequently mentioned. Both the results of the online and face-to-face survey showed that of all PPP users mainly men apply products (51% OS, 70% FTF). Amateur gardeners in Flanders do not apply more than 5 times per year (88% OS, 74% FTF). Furthermore, the PPP use increases according to age and shows a peak for the age group 41-60 years (45% OS, 49% FTF). Regarding education levels, it is noticed that higher educated people use more PPPs than people who achieved a secondary school diploma. The online survey indicated that amateur gardeners especially are clerks, students, executives

and retired people. The face-to-face interviews identified mainly clerks, self-employed and retired people.

Question	Variables	Online survey	Face-to-face survey
		n	n
How many times a year do	you apply PPPs?		
$\chi^2 = 10.7^{a}$	1-5 times	342	45
	5-10 times	46	10
	more than 10 times	11	6
Is the label on the PPP clear	r enough?		
$\chi^2 = 1.79$	yes	206	36
	no	38	8
	more or less	131	16
Which amount of PPPs do y	ou use during an application?		
$\chi^2 = 2.12$	amount indicated on the label	320	48
	more than indicated on the labe	l 13	2
	less than indicated on the label	15	4
	I determine the amount intuitior	nal 28	7
Which type(s) of protective	clothing do you wear?		
$\chi^2 = 29.8^{a}$	gloves	143	30
	hat/cap	23	10
	safety mask	11	5
	boots	156	29
	trousers	187	33
	goggles	47	4
	long-sleeves	109	21
	mouth mask	60	11
	rubber/latex gloves	164	12
	coverall	51	7
	none	27	0
	other	9	4
Where do you buy PPPs?			
$\chi^2 = 1.68$	garden centre	318	53
	supermarket	57	6
	do-it-yourself shop	83	13
	internet	4	0
	other	19	0
Which factors play a role in	the purchase of PPPs?		
$\chi^2 = 7.68$	advice from professional user	174	30
	recommended by friends or fam	ily 88	14
	information on internet	50	13
	product seems safe	45	4
	product seems easy-to-use	67	10
	product is used before	157	27
	product seems effective	73	12
	other	35	1

Table 5-12 Amateur gardeners' knowledge and behaviour related to PPP handling.

<sup>a</sup>Significant difference between survey methods: p < 0.01; n = number of respondents.

70% of amateur gardeners has no knowledge of the different hazard symbols on the PPP label. The information on the label of a PPP is clear enough for 56% of the respondents, more or less clear for 34% and not clear at all for 11%. The majority of PPP users apply the amount as indicated on the PPP label whereas only 3% of all respondents applies more. Almost all respondents protect themselves during both mixing and loading as well as during the application of PPPs. Especially gloves (rubber/latex/fabric) and/or trousers and boots are used as personal protective equipment. The survey results illustrated that PPPs are not always stored safely. 35% of the respondents stated that PPPs are kept out of reach of children and pets. Amateur gardeners buy PPPs especially in garden centres, only 1% buys them online. Especially advice of professional sellers and a good previous experience with the products are main factors influencing PPP purchasing habits.

# 3.3.3 Amateur gardeners' knowledge and behaviour related to PPP application

Table 5-13 illustrates the amateur gardeners' knowledge and behaviour related to PPP application. Plant protection products are mainly used on lawns and driveways followed by terraces, vegetable gardens and flowerbeds. Application of PPPs on hedges, indoor plants, orchards and in greenhouses are less frequent. Furthermore, the indoor use of PPPs is limited to a minimum (6%). Of all households, less than 3% uses on average more than 5 PPPs. Herbicides are most frequently used followed by insecticides and moss killers. Fungicides are applied less. A total of 52 active substances were identified by the online respondents whereas the face-to-face respondents only listed 10 active substances. Table 5-14 shows the top five most listed active substances according to the online and face-to-face survey. In both surveys, the most commonly used substances were glyphosate, fluroxypyr, pyrethrins, metaldehyde and MCPA. Glyphosate is used to kill weeds, especially annual broadleaf weeds and grasses known to compete with commercial crops (BCPC, 2006). Approximately 30% of the amateur gardeners in Flanders use glyphosate in their households. Furthermore, 12-14% of the households use fluroxypyr to control broadleaf weeds and woody brush (BCPC, 2006). Pyrethrins, used in 8-11% of the cases, are used to control a wide range of insects and mites. Metaldehyde is commonly used as a PPP against slugs, snails and other gastropods, and the use of MCPA controls annual and perennial weeds (BCPC, 2006). These active substances are used less compared to glyphosate. 77% of the PPP users in the face-to-face interviews as well as 80% of the online respondents were able to name the active substances and products they use. 39% of the amateur gardeners apply biological alternatives, of which especially slag traps are frequently used. Other alternatives such as nematodes, parasitoid wasps, gall midges, beetles, etc. are rarely used. Plant protection products can be applied in different ways. The results showed that more than 50% of the respondents (56% OS, 52% FTF) opt for the use of a pressurised knapsack sprayer or granules (e.g. slug pellets). Despite the fact that ready-to-use products are easier to handle and user-friendly, they are used less (14% OS, 8% FTF). Surpluses of PPPs are either stored for later use (44% OS, 23% FTF) or sprayed empty on plants (41% OS, 59% FTF). Only a small part of the respondents will take the PPP surpluses to a collection point (7% OS,

13% FTF). Rinse water of cleaning the application equipment is by more than 50% of the amateur gardeners applied to plants. However, one third of the respondents still pours the rinse water into the sink.

Question	Variables C	Online survey	Face-to-face survey
		n	n
Where do you use PPPs?			
$\chi^2 = 10.9$	lawn	214	36
	greenhouse	31	2
	hedge	63	6
	flowerbed	100	23
	vegetable garden	105	23
	terrace	131	18
	driveway	216	36
	orchard	29	9
	indoor plants/indoors	49	12
How do you apply PPPs?			
χ <sup>2</sup> = 9.36	ready-to-use sprayer	112	9
	trigger sprayer	48	8
	watering can	86	18
	pressure sprayer	94	20
	pressurised knapsack sprayer	235	32
	granules	204	27
	other	5	0
How do you get rid of the	leftover spray liquid?		
$\chi^2 = 20.9^{a}$	throw it away in the sink	2	3
	spray it on the treated object	154	36
	save it for later use	163	14
	bring it to a collection point	26	8
	other	8	0
What do you do with rinse	e water?		
$\chi^2 = 5.18$	throw it away in the sink	106	20
	spray it additionally onto the treated o	bject 197	36
	bring it to a collection point	10	3
	other	46	2

Table 5-13 Amateur gardeners' knowledge and behaviour related to PPP application.

<sup>a</sup>Significant difference between survey methods: p < 0.01; n = number of respondents.

Table 5-14 Top 5 most used active substances according to online and face-to-face survey with their respective percentage (%) of use.

Active substance	PPP group	Online survey (%)	Face-to-face survey (%)
glyphosate	herbicide	32	30
fluroxypyr	herbicide	12	14
pyrethrins	insecticide	8	11
metaldehyde	molluscicide	5	10
МСРА	herbicide	3	3

# 3.3.4 Amateur gardeners' awareness to PPP residues

34% of amateur gardeners take the possible presence of PPP residues on their food (both home-grown or bought in store) into account. 8% of all respondents indicated no action to reduce residues. However, the majority of PPP users wash (45%) or peel (33%) their food to limit the number of PPP residues. Finally, 84% of all respondents indicated not adjusting its PPP use after completing the survey.

# 3.3.5 Discussion

Three different survey methods were used to carry out the survey, i.e. telephone interviews, online survey and face-to-face interviews. First, a very low response rate was obtained by telephone interviews. Although a large number of people can be addressed, it is quite difficult to convince someone to participate in the survey via telephone. The disadvantages of telephone interviews, such as unwillingness, were observed (Louwen, 1992; Stoop, 2005; De Leeuw, 2010). Second, online surveys are inexpensive, time-saving and user-friendly (Bronner et al., 2014). Furthermore, respondents can complete the survey at their own pace and check some questions. For example, the online respondent had the opportunity to take a look which kind of products are stored at home or perform research on the internet. This was not possible during the face-to-face interviews. A disadvantage of the online survey is that the reached target group is quite limited. 862 people were willing to complete the online survey, of which only 47% use PPPs. These surveys are also open to misinterpretation with respondents, leaving out specific information or even disregarding part of the survey (Eurostat, 2008). Some answers did not respond to the question. Clarification of the answers or questions was not possible during this kind of survey. Consequently, these answers had to be deleted. Third, face-to-face interviews are appropriate to come into contact with the amateur gardener and are also quite accurate. The trained personnel can give an explanation to the questions in order to achieve a more fair and clear answer. On the other hand, the words or actions of the interviewer can intentionally influence respondents to answer in a particular way. Face-to-face surveys, however, are time-consuming and also cover a smaller geographic area than other data collection methods.

At least half of all respondents use PPPs and usually not more than five times per year. Age appears to affect the use of PPPs, i.e. the number of PPP users increases by age. Furthermore, highly educated people apply more PPPs than people with a secondary school diploma. These results are in accordance with the studies of Steer and Grey (2006) and Grey et al. (2006). Higher educated people have higher incomes and are more likely to own their home or to belong to non-manual social classes compared to less frequent users. The effect of education may reflect a greater knowledge and awareness of PPPs leading to a higher reported usage. The strong housing effect may in part reflect non-homeowners passing the responsibility for treating problems to the property owners leading to a lower family usage. The profession itself exerts no unambiguous influence on the PPP use. According to Grey et al. (2006), on average 3 to 4 products per household are used. Most PPPs were used in the

garden, followed by indoor use. The Flemish amateur gardeners use on average not more than 5 PPPs per year, whereas more than three guarters of all respondents use less than 5 PPP applications per year. Previous studies already revealed that labels on PPPs are not read well or are not completely understood by amateur gardeners (Weale and Goddard, 1998; Grey et al., 2006; Mostin, 2007; De Cock and Knaepen, 2008). This survey confirmed that 70% of the respondents has no knowledge of the different hazard symbols on the label of PPPs. The label is still not clear enough for at least 40% of the respondents. Not understanding the PPP label can lead to an increased health risk (Waichman et al., 2007). It remains a challenge to better inform the amateur gardener by providing easier to understand PPP labels and to provide advice on how to deal with PPPs both in terms of safety for humans and the environment. Harrington et al. (2005) stated that potential exposure to amateur PPPs is highest during mixing and loading, and this mainly at the height of the hands and chest. However, almost all respondents protect themselves with gloves and/or trousers and boots during both mixing and loading as well as during the PPP application. Details with regard to the re-use of gloves are not known. This can be a source of contamination and may pose a larger problem compared to not wearing any gloves. The study of Rushton and Mann (2009) indicated that PPPs are often not stored securely, which was also observed in this survey. The increasing popularity of online purchases is not yet noticeable in the PPP industry, as only 1% of amateur gardeners buys PPPs online. Furthermore, the internet proves to be an important source of information and will probably increase in the near future. The control of correct PPP information on the internet will therefore be important as well. Amateur gardeners especially rely on the advice given by professional sellers who also have the best knowledge of the PPPs (De Cock and Knaepen, 2008).

Plant protection products are mainly used on lawns and driveways whereas the use of PPPs indoors is limited to a minimum. In 2006, especially insecticides were used in UK households, whereas fungicides were hardly used (Grey et al., 2006). On the other hand, the use of herbicides in households has increased (Whitmore et al., 1992; Feagan and Ripmeester, 1999; Donaldson et al., 2002; Grey, 2003). In Flanders, fungicides are applied less whereas herbicides are the main PPPs with glyphosate as the most commonly applied active substance. Knowledge of the active substances and PPPs proves to be quite high. Only 20% of all respondents was unable to mention or indicate at least one product. Striking fact here is that in the online survey 52 active substances were identified compared to only 10 active substances in the face-to-face interviews. During the online survey, the respondents had the opportunity to take a look which kind of products are stored at home. This was not possible during the face-to-face interviews. The identified active substances were especially products that belong to the list of authorised PPPs (Regulation (EU) No 656/2011, Annex III). However, some unauthorised active substances were listed as well, i.e. phoxim, permethrin, disodium-EDTA, brodifacoum and butocarboxim. The insecticide phoxim is toxic to bees and birds and has been prohibited for use in the European Union since 2007. Permethrin is prohibited as PPP since 2000. Despite its non-selectivity and toxicity to fish and other aquatic organisms, it is allowed as biocide. Disodium-EDTA is used as moss killer in Belgium. Since 2005, brodifacoum is prohibited as PPP, but it is still allowed as biocide to kill rodents. Also butocarboxim, used as an insecticide, is no longer authorised in the EU since 2002. However, the frequency in which these substances are listed, is rather small. In general, amateur gardeners in Flanders use only authorised products. Flemish amateur gardeners still prefer chemical PPPs over biological alternatives as in the US and the UK (Savage et al., 1981; Grieshop and Stiles, 1989; Davis et al., 1992; Grieshop et al., 1992; Adgate et al., 2000; Grey, 2003). Ready-to-use products are easier to use and reduce the potential exposure during the application of PPPs (Grey et al., 2006; FOD, 2009). Despite these benefits, the Flemish amateur gardeners still prefer to use a pressurised knapsack sprayer due to the fact that larger areas can be treated at once. Furthermore, the fact that amateur gardeners are still not familiar with ready-to-use products and the high price for these diluted products also plays a role. Surpluses and rinse water are mainly applied to plants, which is in accordance with the Code of Good Agricultural Practice (Tyvaert et al., 1999). About 30% of the respondents still pours rinse water into the sink. This way, households may contribute to the emission of PPPs and these discharges may negatively affect the environment (VMM, 2015). Furthermore, Directive 2008/105/EC lists a number of priority substances that pose a certain risk to the aquatic environment, including some herbicides. Surface water measurements of the VMM indicated that many active substances exceed the water quality standards in Flanders, especially herbicides prove to be problematic to aquatic life (VMM, 2015). Amateur gardeners should be continuously reminded to the effects that their actions may have on the environment. Practically all PPPs are purchased in garden centres, do-it yourself shops and supermarkets.

A study conducted by TNS Opinion & Social (2010) at the request of the European Food Safety Authority showed that 72% of the Belgian consumers have concerns about residues of PPPs on fruit and vegetables. In Flanders, 34% of all respondents take the possible presence of PPP residues on fruits and vegetables into account. 8% of all respondents indicated not to act in order to reduce residues. However, the majority of PPP users wash or peel their food to limit the number of PPP residues.

# Conclusion

The estimation of non-agricultural use of PPPs in Belgium highlights the complexity and diversity of actors involved in chemical control. Overall, 17 categories of PPP use were identified. These categories correspond to areas where PPPs are applied. Each category includes a panel of actors responsible for applying PPPs (= interlocutors) which were identified as well. Given the complex federal structure of Belgium, collecting information for identifying the different PPP use and interlocutors is somehow difficult because of the lack of answers from some potential interlocutors and the implementation of new legal measures at federal and regional level. Based on expert judgment, a ranking of the 17 categories according to their importance in terms of PPP use and data accessibility was set

up. A collection of Belgian statistics on the used quantities of PPPs including at least the names of active substances and their quantities (expressed in kilograms) used in a given year are required to estimate non-agricultural use of PPPs in Belgium. To achieve this, four interlocutors that are relevant in terms of importance of non-agricultural PPP use should be investigated, i.e. amateur gardeners, private companies, private companies of parks and gardens and private companies for water, electricity, gas and phone. The focus on these key-actors can be made through the implementation of different types of inquiries: investigations into sample for private companies, exhaustive investigations for private companies of parks and gardens and for private companies for water, electricity, gas and phone. The inquiries should be combined with data collection for amateur gardeners obtained by sales figures of PPPs from FPS.

Subsequently, one of the key players in the non-agricultural use of PPPs was further investigated, i.e. non-professional users and more precisely amateur gardeners. Whereas professional use of PPPs is more regulated, this is less conventional for non-professional use as only usage advice can be given. Both administrative data as more detailed information by performing a survey were used to provide an indication of the non-professional PPP use.

In the second part of this chapter, an attempt was made to estimate the pressure of nonprofessional use of PPPs on operators, aquatic organisms and bees in Belgium based on sales figures and by using three exposure models. The use and pressure on operators and the environment of non-professional PPPs in Belgium declined strongly for the period 2005 to 2012. This reduced pressure is the result of efforts made by the government and industry, such as the replacement of more toxic PPPs by less toxic PPPs and by the introduction of safer application methods. In the future, it remains a challenge for the industry to bring PPPs on the market that are less toxic but still just as effective. In addition, the focus should still be on safe application methods and advice should be given on how to deal with PPPs both in terms of humans and the environment. Based on the results of this part, special attention should be paid to aerosol spray can applications and the non-professional use of insecticides (especially imidacloprid). Furthermore, the use of non-professional sales figures is more reliable than usage figures of non-professional PPPs, as the latter are dependent on the representativeness of the surveyed public.

However, non-professional sales figures could be supplemented with more detailed and specific information by performing a survey to investigate the current PPP handling behaviour of non-professional users. This was further investigated in Flanders by means of three different survey methods. Survey results illustrated that the knowledge of the different hazard symbols on the label of PPPs is still insufficient. At least 40% of the respondents indicate that the PPP label is still not clear enough, which can lead to an increased health risk. It remains a challenge to better inform the amateur gardener by providing easier to understand PPP labels and to provide advice on how to deal with PPPs both in terms of safety for humans and the environment. Most amateur gardeners wear

personal protective equipment during preparation and application of PPPs. However, 100% protection should definitely be pursued. About 30% of the respondents still pour rinse water into the sink, a practice that needs to be avoided. The amateur gardener can reduce the pressure on the environment by consciously dealing with surpluses and rinse water of the used PPPs. Only few people rely on the internet to acquire information regarding PPPs so far. People especially trust on the tailor-made advice of a professional seller when they purchase PPPs. Also reassuring is the fact that in general only authorised active substances are used.

Conducting inquiries for the targeted interlocutors shall efficiently contribute to the estimation of non-agricultural use of PPPs and will serve as a basis for the definition of the guidelines of the European methodology. At the same time, these results will facilitate the transition to the changes expect from Directive 2009/128/EC for the estimation on the used quantities of PPPs over time, involving an expected reduction in the used quantities of active substances for most categories of PPP use. The targets and measures deriving from this directive will lead to a major change in plant protection practices in the near future. The results of this chapter also illustrate the importance of collecting additional data by a combination of administrative data with inquiries in order to be able to estimate non-agricultural use of PPPs more precisely.

# Chapter 6 General discussion, conclusions and future challenges

The aim of this doctoral dissertation is to present the work that has been done to improve the estimation of PPP use in Flanders in order to respect the requirements stipulated in Directive 2009/128/EC. These usage estimates are used to perform risk assessments to indicate the pressure of PPPs on human health and the environment in the framework of policy decisions (e.g. MINA-plan). The theoretical work performed in this doctoral dissertation was supported by experimental work. This chapter starts with a general discussion and conclusion of the main results from this work by answering the research questions described in **Chapter 1** and ends with some suggestions for future research.

# General discussion and conclusions

The structure of this thesis follows the DPSIR-framework, a widespread analysis framework within environmental reporting. 'D' stands for Driving forces, 'P' for Pressure, 'S' for State, 'I' for Impact and 'R' for Response.

**RQ1** Do total PPP use estimates (D) based on sales figures differ from estimates based on usage registration?

Databases should be complemented with new, more accurate information and research results to ensure the transparency of the applied data and to avoid misinterpretations of the policy makers. Total PPP use estimates can be determined by means of sales or usage figures. Sales figures of PPPs are relatively simple to collect and fairly inexpensive. However, sales figures give rise to confidentiality issues and restrictions on the release and use of data for commercial reasons (i.e. for non-professional PPP sales figures provided by industry). Sales figures contain no information about the crop on which they are applied, timing, regional variation in use, dose applied, number of applications to the crop or percentage of crop treated. Usage estimates cover all kinds of data on the actual use of PPPs by operators, but are not always quick and easy to produce. Use estimates differ from national sales figures as a certain number of PPPs are purchased in a given year, but are not necessarily applied in that year due to the non-presence of a particular disease or pest. Meteorological factors determine the development and intensity of pests and diseases resulting in a lower or higher PPP use. The amount of PPPs sold is also affected by predictive models and early warning systems providing information on the possible occurrence of a particular pest or disease and information concerning its prevention and control. The difference between sales and use estimates can also be explained by economic reasons, such as budgets that need to be spent in one year or commercial actions recommending certain products which results in a stock of PPPs. Furthermore, the distribution of PPP use among crops established by using usage estimates instead of sales figures of PPPs is based on an approach that is per definition more reliable.

To estimate total agricultural PPP use on farms, farm accountancy data are a useful tool. The purpose of FADN is to collect technical and economic data on the agricultural and horticultural holdings in Belgium through a network for the collection of accountancy data in order to answer the requirements of the European legislation. Regulation No 79/65/EEC imposes the Member States to create a network for the collection of accountancy data on the incomes and business operation of agricultural holdings in the European Community. In other terms, the accountancy data from FADN are used to estimate and monitor the profitability trend of agricultural and horticultural holdings in Belgium. Since 2006, the annual use of PPPs of some 700 holdings in Flanders is monitored yearly by the Flemish FADN. In Wallonia, the Walloon FADN has collected data on usage of PPPs since 2002. The number of holdings in the yearly samples of the Walloon FADN accounts on average 500 holdings (Lievens et al., 2014). A limitation of this methodology concerns the fact that FADN is based on a voluntary approach of the farmer but there is no obligation for farmers to participate in the survey of FADN. Furthermore, the main objective of FADN is to evaluate the income of agricultural holdings and not to produce PPP use estimates. The survey does not cover all agricultural holdings in the region but only those, due to their size, could be considered commercial.

**RQ2** Does the approach based on usage registration narrow the gap between risk indicators (P) and reality (S)?

Use indicators (expressed in kg a.s.) are not adequate proxies for assessing pressure exerted by PPP use. A useful indicator takes both the toxicity of PPPs and their degradation time into account as these characteristics vary depending on the active substances. One kilo of one PPP can exert a completely different pressure than one kilo of another PPP if their toxicity is taken into account. The risk of exposure to PPPs is quantified by weighting the use of PPPs to the toxicity coefficients for the various environmental compartments. Therefore, toxicity coefficients of the active substances exert an influence on the results of the indicator. An active substance can be highly toxic to humans and the environment even if only a small amount of the active substance is used. The pressure exerted by PPP use on humans and the environment is hereby not proportional to changes in the overall use or the corrected usage. In general, it was observed in this thesis that estimated use of PPPs based on accountancy data is more useful compared to sales figures in order to perform risk assessments. By using sales figures, products that are no longer sold, are not taken into account in the risk calculations. However, if these products are still used in practice, they can still exert pressure on human health and the environment. Despite its limitations, sales figures are a useful tool to adjust and improve surveys on PPP use and to produce national estimates of PPP usage.

**RQ3** What is currently the behaviour of farmers in Flanders related to PPP handling and application (D-R)?

In order to better inform farmers how to consciously deal with PPPs, data collection related to PPP handling and application of Flemish farmers is recommended by means of face-to-face surveys. These surveys are time-consuming and also cover a smaller geographic area than other data collection methods. However, face-to-face surveys are appropriate to come into contact with the farmer and are also quite accurate. The trained personnel can give an explanation to the questions in order to achieve a more honest and clear answer.

The results of the face-to-face surveys indicated that differences in meteorological factors influence the range of pest, disease and weed problems, require control, or affect the ability of the farmer to apply the PPP under suitable conditions. Further, pest pressure may be higher during warm weather resulting in higher inputs to some crops, e.g. during the summer than in the winter. The crop type also influences PPP usage, some crops e.g. potatoes are more susceptible to various pests and diseases. In general, all surveyed farmers applied the amount as indicated on the PPP labels. Wearing PPE – and especially gloves and coveralls - reduces the risk of dermal exposure during mixing and loading and is therefore highly recommended. Almost all operators surveyed in 2013 protected themselves with gloves, coveralls and boots during mixing and loading. During PPP application, gloves were hardly used due to the use of a closed cab sprayer. However, also during PPP application, cleaning the sprayer and performing other work activities in the field, gloves and coveralls have to be worn. Especially when using PPPs which pose large risk to humans (e.g. diquat), both operators and re-entry workers have to wear full PPE. Few farmers indicated the use of buffer strips, however, authorisations for most Belgian PPPs containing active substances that are known to pose risk to aquatic organisms stipulate a minimum buffer and/or drift reduction. Further, most farmers still used a classical spray technology instead of a driftreducing technology.

RQ4 Can risk assessments (P) be improved by having extra insights in the practice (D-R)?

Especially while using a multi-risk indicator – like POCER – which requires more input data, it is appropriate to provide additional information. Performing a survey to investigate the current PPP handling behaviour of farmers helps to improve these risk calculations. For example by pairing POCER with a farm level questionnaire, calculated RIs are more accurate. Compared to earlier sector and regional level implementations, the refined RIs concur more closely with the individual farmer's actual crop protection practices and make a better starting point for discussion between farmers. Both case studies described in this thesis illustrate how responsive modifications in crop protection practices can reduce human and environmental risk. Risk indicators can also serve as decision support tools, because simulations of management changes or substitution between PPPs can help farmers to decide on the most sustainable alternatives.

#### RQ5 Seed treatments: a guide to sustainable use?

Seed treatments are an environmentally friendly way of using PPPs as the amount used can be very small compared to leaf or soil spraying. However, the use of treated seeds also has some disadvantages which are all related to the coating of the seed. The determination of the amount of PPPs used in seed treatment is difficult since no figures are publicly available in Belgium. In this thesis, seed treatment data were calculated by using various assumptions. Only a fraction representing the average number of seed treatments across the total crop could be obtained by means of experts of industry (Phytofar). These figures could only be illustrated by an example due to the confidentiality of the obtained data. Recent incidents of bee losses were related to the application of seeds treated with PPPs. Due to these issues, it is useful to collect data from the use of PPPs during the treatment of seeds as well. These data can be provided by manufacturers of seeds coated with PPPs. Nowadays, this is not yet possible because of confidentiality reasons.

RQ6 Does the use of seed treatment exert pressure (P) on pollinators (e.g. honey bees)?

Pressure exerted by the use of PPP treated seeds on bees deserves the highest priority for investigation. Residues of systemic active substances used for seed coating can be present in the guttation fluid, plant pollen and nectar of seed treated plants. Recent incidents of bee losses in several countries, among other things, seemed to be the result of this emission. The uptake, translocation and persistence vary greatly among seed treatment products. Various PPPs used in the treatment of seeds are quite persistent. Some of them can still be found in the mature stage of the plant. In order to verify if the use of seed treatment products exert pressure on pollinators, a system analysis of seed treatment was conducted. Since 2013, the use of neonicotinoids is limited in Belgium. Therefore, the focus was on seed treatment products other than neonicotinoids. Results of this thesis illustrate that several active substances, used as seed treatment products, are translocated through the plant. In this thesis, a lethal dose for bees was never registered in the guttation fluid and not one of all detected substances in bee bread and beeswax is authorised in the treatment of seeds in Belgium. The risk of exposure to pollinators by means of seed treatment in Belgium is supposed to be very low for the crops and active substances investigated in this study. Additional studies should cover other crops and seed treatment products to investigate whether this issue is limited to some specific cases or is more widespread within the seed treatment industry. Further studies at national level should be performed on beeswax as well.

**RQ7** Which actors are responsible for the non-agricultural use of PPPs (D) in Flanders/Belgium?

Due to the implementation of new legal measures at federal and regional level, the use of PPPs is prohibited in some non-agricultural areas in Belgium from the period 2014 to 2020. A lot of identified actors responsible for applying PPPs, will not use PPPs anymore. Many of

them switch to alternative control methods. The national quantities of active substances attributed to non-agricultural professional users will probably decrease over time and will be allocated to other remaining categories. To collect information on non-agricultural use of PPPs, four interlocutors should be further investigated, i.e. amateur gardeners, private companies, private companies of parks and gardens and private companies for water, electricity, gas and phone. These interlocutors will stay important in terms of PPP use for the future too. The focus on these key-actors can be made through the implementation of different types of inquiries: investigations into sample for private companies, exhaustive investigations for private companies of parks and gardens and for private companies for water, electricity, gas and phone. The inquiries should be combined with data collection for amateur gardeners obtained by governmental sales figures of PPPs.

RQ8 Does the use of non-professional PPPs (D) exert pressure (P)?

As described in **Chapter 2**, the results of this thesis suggest to utilise accurate PPP usage estimates to determine the pressure exerted by PPP use on humans and the environment. However, PPP sales figures are also a useful tool to adjust and improve surveys on use of PPPs. For example, the use of non-professional sales figures is more reliable than usage figures of non-professional PPPs, as the latter are dependent on the representativeness of the surveyed public. These sales figures were used to calculate the pressure exerted by non-professional PPPs on operators, aquatic organisms and bees. In general, the pressure exerted by non-professional PPP use reduced for the studied period. This reduced pressure is the result of efforts made by the government and industry, such as the replacement of more toxic PPPs by less toxic PPPs and by the introduction of safer application methods. In the future, the challenge remains for the industry to bring PPPs on the market that are less toxic but still just as effective.

**RQ9** What is the current PPP handling behaviour by non-professional users (D-R), e.g. amateur gardens?

Information according to the PPP handling behaviour of non-professional users can be collected by means of online and face-to-face surveys. Unlike face-to-face surveys, online surveys are inexpensive, time-saving and user-friendly. Respondents can complete the survey at their own pace and check some questions. A disadvantage of an online survey is that the reached target group is quite limited. These surveys are also open to misinterpretation with respondents, leaving out specific information or even disregarding part of the survey. Some answers in the online survey conducted in this thesis, in fact, did not respond to the question. Clarification of the answers or questions is not possible during this kind of survey. Hence, a suitable method is selected depending on the required information. Especially to perform risk assessments for PPPs and to improve models of operator cumulative exposure, data collection by means of face-to-face surveys is recommended.

Survey results illustrated that especially the knowledge of different hazard symbols on the label of PPPs is still insufficient. Most amateur gardeners wear PPE during application and preparation of PPPs. However, 100% protection should definitely be pursued. Reassuring is the fact that in general only authorised active substances are used. Anyhow, the focus should still be on better informing the non-professional user by providing easier to understand PPP labels and to provide advice on how to deal with PPPs both in terms of safety for humans and the environment. Non-professional users of PPPs should be continuously reminded to the effects that their actions may have (e.g. disposal of surpluses and rinse water).

**RQ10** Can the estimation of the pressure (P) exerted by non-professional PPP use be improved by having extra insights in the practice (D-R)?

The results of this thesis illustrate the importance of collecting accurate data by a combination of administrative data with questionnaires in order to be able to estimate data on PPP use more precisely. When using only administrative data, assumptions – like the correct use of PPPs – are made. It is not verified if an operator actually follows the guidelines present on the package of the products (use of PPE, application rate, etc.) and applies the products correctly. Pressure by the incorrect use of PPPs can in this way be higher than estimated.

# **Future research**

Sales figures of PPPs are definitely a useful tool to produce national PPP usage estimates and especially to estimate non-professional PPP use. In order to perform risk assessments, however, use estimates are recommended. Risk is uncertainty that 'matters' (Harwood et al., 1999) meaning that risk calculations result in a better view of actual PPP use and its related pressure on human health and the environment. Data collection on agricultural PPP use on farms was in this study obtained by means of farm accountancy data (FADN). Due to some limitations of this method, the use of crop management record sheets – the most commonly used recording system in Belgium - is suggested to collect estimates of agricultural PPP use on farms. Operators of food businesses that produce or harvest crop products should record data on the use of PPPs and biocides at least seven days after execution. These records are preserved for five years for crops intended for human and animal consumption. When necessary, this mandatory information should be made available immediately. The sheets are filled in on a field-by-field basis by the operator and provide information on date of application, applied product, product dose (per ha), treated area and operator (farmer or spray contractor), date of sowing/planting, fertilisation activities and date of harvest (KB 13/07/2014). Using this source of administrative data will definitely improve PPP use estimates.

Nowadays, policy makers use simple indicators to evaluate trends, e.g. the Seq-indicator. This indicator requires limited data input and provides an easy tool for environmental policy planning. However, such indicators have their limitations, especially when applied in a legislative framework. Careful consideration is necessary within such a framework and a more case by case approach is advisable. Furthermore, the quality of the input data has shown to be of great importance. These simple indicators can provide a good first tier approach, whilst keeping in mind the limitations of these tools. Multi-risk indicators – such as POCER – have the advantage that risk can be assessed for multiple components such as human exposure and other environmental compartments. Dependent on the quality of the underlying models and calculations, the output of more complex indicators should be closer to the real situation. However, such indicators are often too complex and data-intensive which reduces their applicability on a regional scale. Although these limitations of multi-risk indicators, their use would provide insights on PPP use at local and sector levels, e.g. at farm level.

POCER is put forward to use as a farm level indicator in Belgium. Firstly, it is adapted to the Belgian context (PPP legislation, cultivation conditions, etc.) in which the sustainability of the farms is evaluated. Secondly, the indicator can be used as a decision-making tool to choose between alternative PPPs with respect to their potential pressure on humans and the environment. Thirdly, for a certain mode of application of a PPP, the environmental areas that need special attention can be identified. Fourthly, it can also be used as a tool to evaluate the effect of measures to reduce the exposure of PPP operators and workers and to reduce emissions to the environment. Finally, it can be used to assess the pressure on human health and the environment of all PPP applications related to a crop within a year and to evaluate alternative cropping systems. This thesis emphasises the usefulness of combining POCER with a farm level questionnaire. By means of face-to-face surveys extra insights in the farmers' practice are obtained in order to refine POCER calculations. These refined risks concur more closely with the individual farmer's actual crop protection practices. Face-to-face surveys reveal the knowledge, awareness and attitude towards PPP use, which are considered to be the driving factors behind the human actions. When performing a survey to obtain insights in the crop protection practices, it is vitally important that those farmers selected in the sample are fully aware of what is required in order to be able to collect accurate information throughout the process. For future face-to-face surveys it is also essential that participants chosen for the study are those that have already participated in collaborative work. Wherever possible, farmers with existing detailed records should be chosen as this ensures the data collected are complete and will reduce the amount of their time to help with the survey. Knowing that detailed records are in place ensures that visits can be made at any time of the day and at a time most suitable for individual farmers. Furthermore, by collecting this kind of information public authorities are aware of how mitigation measures are implemented and enforced in practice. Effective mitigation measures have to be put in place to reduce exposure to PPPs and should definitely be pursued. In general, the finest insights in agricultural PPP use can be obtained by combining administrative data (FADN or other PPP recording systems) with questionnaires on the behaviour of farmers related to PPP handling and application. In this way, the loss of information of operators who are not consciously dealing with PPPs is avoided.

An additional important note is that both seed treatment with PPPs and non-agricultural PPP use should be taken into account in environmental policy planning in order to obtain an overall view on PPP use and its related pressure on human health and the environment. This thesis illustrated the importance of knowledge on PPPs used for seed treatment and non-agricultural purposes on both Flemish as Belgian level. Although the research on seed treatment and non-agricultural PPP use conducted in this thesis represents a step forward to produce more accurate information to ensure the transparency of the applied data, there are still some fields in which improvement is possible and additional research is required.

Several experiments with treated seeds were performed to describe translocation and persistence of PPPs in the plant after seed treatment. However, the availability of seed treatment data remains a bottleneck for extending PPP usage estimates. The crop protection industry owns data from seed treatment products conducted as part of the registration dossiers, but unfortunately these data are not publicly available. Cooperation of public authorities with these industry task forces would greatly facilitate data access. Further, this thesis did not investigate residue levels of seed treatment products in pollen and nectar. There are few published data of systemic PPP residues found in pollen and nectar but only for seed treatments of some crops, e.g. maize and sunflower. Hence, extrapolation to other crops was not considered appropriate. In the EFSA guidance document on risk assessment of PPPs on pollinators (2013), worst-case assumptions were considered for the development of the risk assessment schemes due to the limited available datasets on residue levels in pollen and nectar. Furthermore, no standard test protocols are available. Therefore, the only way to refine risk assessments is a more precise estimate of the potential exposure in nectar and pollen. The currently ongoing EFSA project 'Collection and analysis of pesticide residue data for pollen and nectar' further investigates this topic which should definitely be taken into account into future risk assessments. In general, a lack of knowledge on toxicity values of various active substances and its metabolites still exists. Regarding toxicity to honey bees, residue levels of methiocarb sulfone and methiocarb sulfoxide found in the guttation drops studied here could not be compared to toxicity values. It is necessary to complement toxicity databases with new, more accurate information and research results to ensure the transparency of the applied data. Therefore, it remains a challenge for public authorities to encourage industry of making toxicity values publicly available.

An innovative study in the framework of the Eurostat project on non-agricultural use of PPPs was conducted in the collaboration with FPS and the Catholic University of Louvain. Belgium appeared to be further evolved in the field of data collection and reporting concerning PPP use compared to other consortium members. Furthermore, this research on the non-agricultural PPP use paired with knowledge about agricultural use of PPPs presented an overall image of PPP use in different sectors in Belgium, and more precisely in Flanders and

Wallonia. Focus on the key-actors in non-agricultural PPP use should be preserved in future research as these interlocutors will stay important in terms of PPP use for the future too.

In general, future research on PPP use should be conducted by means of cooperation between Flemish and Walloon institutes (both research institutions as public authorities) in order to promote exchanges and to share experience in collecting data on PPPs in Flanders and in Wallonia. Furthermore, another recommendation for public authorities is the setup of a database which validates the approval status of a product per country and the rate of application (maximum and minimum). Although it would require a significant cost and continuous investment the use of this database would be quite beneficial. Regular checks on the quality of the data during data collection and more frequent progress discussions with all partners should also be made to improve the collection of PPP usage data.

Another interesting topic for further investigation is the use of tank mixes. The risk of wetting agents or other adjuvants added separately to the tank mix have to be considered. Information on the toxicity of adjuvants is currently lacking. Regulation (EC) No 1107/2009 also requires that adjuvants undergo a strict authorisation procedure before they can be placed on the market. Due to this, information should become available to sum the risk exerted by adjuvants and co-formulants with those of active substances. Information on potential cumulative effects of formulations or tank mixes of multiple active substances is currently also lacking and cannot be taken into account.

In the future, the PPP industry should focus on bringing PPPs on the market that are less toxic but still just as effective. Further, safe application methods and easier to understand PPP labels should also be put forward. In addition, public authorities and industry should continue working together to further enhance knowledge and awareness of the public related to PPP use and its possible effects, e.g. the advisory council of NAPAN (which was up to 2015 the advisory council of FPRP). In this council, public authorities display their projects and achievements related to sustainable use of PPPs in order to gather remarks and suggestions from different stakeholders.

Training of farmers, spray operators and advisors should be continued. Wearing full PPE should definitely be pursued. Concerning farmers, the use of gloves, coveralls and boots is recommended to reduce the risk of dermal exposure of PPPs during PPP preparation and application, cleaning the sprayer and performing other work activities in the field. Non-agricultural users – such as amateur gardeners – only have access to garden products (unless they are in the possession of the Phytolicence certificate). These garden products carry a minimal risk of exposure to both operator and the environment. However, especially gloves should be worn at all times. Few farmers surveyed in this thesis indicated the use of buffer strips and drift-reducing spray technology. Therefore, farmers should be notified why these measures have been introduced by public authorities and its importance to pursue them (e.g. by means of discussion in small farmer groups). In addition, public authorities should also be open to discussion about the feasibility of some mitigation measures. Furthermore,

farmers and all other operators of PPPs should be continuously reminded to the effects that their actions may have on the environment. Practices like pouring rinse water into the sink should be avoided. Specific advice should be given on how to deal with surpluses and rinse water both in terms of safety for humans and the environment.

# Personal data

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# **Professional career**

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#### Main topic:

The use of plant protection products in Flanders and risks for humans and the environment

- data processing related to the non-agricultural use of PPPs (e.g. in the context of the Eurostat project)
- data processing related to the use of PPPs in agriculture in Flanders (e.g. in the framework of the EFSA project and DISCUSS)
- reporting the section PPPs of MIRA (Flanders Environment Report, VMM)
- carrying out risk assessments and risk evaluations of PPPs

# Other topics:

- collaboration in the service of the lab
- educational support

# Education

2010 – 2012	Master in Bioscience Engineering, option Environmental Technology Faculty of Bioscience Engineering, Ghent University, Belgium
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# **Publications**

#### International journals with peer review

<u>Fevery, D.</u>, Peeters, B., Lenders, S., Spanoghe, P., 2015. Adjustments of the pesticide risk index used in environmental policy in Flanders. PLoS One. 10(6), 21 pp.

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# Other

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# **Scientific activities**

# **Oral presentations at conferences**

2015 Pilot study on estimating non-agricultural use of pesticides in Belgium 67<sup>th</sup> International Symposium on Crop Protection (Ghent, Belgium)

# Poster presentations at conferences

2015	Seed treatment in Flanders
	67 <sup>th</sup> International Symposium on Crop Protection (Ghent, Belgium)
2014	Different methodologies for data collection on non-agricultural use of pesticides
	66 <sup>th</sup> International Symposium on Crop Protection (Ghent, Belgium)
	Introducing the DISCUSS indicator set in farmers' discussion groups
	66 <sup>th</sup> International Symposium on Crop Protection (Ghent, Belgium)
2013	Agricultural and non-agricultural use of chemical crop protection products
	65 <sup>th</sup> International Symposium on Crop Protection (Ghent, Belgium)
# **Appendix A**

#### 1. Survey questionnaire

#### Survey forms

Form 1 (Cropping details) was used to provide background information on the farm. It included details on the location and size of the farm and provided a summary of all cropping on the farm. It allowed the surveyor to have an overview of the type of questions that would need to be asked as the interview progressed. Contrary to the previous study (Glass et al., 2012) all crops, rather than only the selected crops were included in the study if these had been sprayed by the principal spray operator. Form 2 (Farm business details) allowed the surveyor (and the farmer) to organise how the interview was going to be structured. It allowed the surveyor to establish whether more than one farm was managed by the farmer as well as the number of spray operators, spray decisions, the number of workers and the range of worker activities carried out on the farm. This allowed the surveyor to estimate the length of time the visit was likely to take and how the most accurate and pertinent data could be collected. Form 3 (PPP application) was the most critical form of the survey and included detailed records of the crops, the areas grown, the dates, rates and methods of application of all PPPs applied to an individual field. It also contained information on the operator, the sprayer, nozzles, start time of spraying and duration of spraying. In some cases not all information was available and a default value of 99 or UN (unknown) was recorded to indicate where data were not available. On each farm one or more fields were selected as environmental fields (ERA fields). From the ERA field additional information was collected on the field margins and, where available, historical PPP usage data for the last five years were collected as well. The intention of the study was to collect data on the principal spray operator. In order to minimise the time taken for the collection and processing of the data and the time spent by the farmer or grower in providing the data, only data relating to the principal spray operator activities were collected. The only exception to this was on the ERA field where all PPP applications made to the field were recorded. Form 4 (Spray operator details) was used to collect information on the principal spray operator on the farm. In some cases, details for other spray operators were collected as well. It included personal details on the operators and their behaviour relating to mixing and loading, cleaning the sprayer and use of PPE. Form 5 (Spraying equipment details) was used to collect information on all of the sprayers, primarily farm owned, used on the farm. It included information on the manufacturer's name (where applicable), sprayer details including sprayer speeds, boom sizes and tank capacities, the sprayer type, PPP filling systems and the nozzle type and use. Form 6 (Principal spray operator - work activity details) was used to collect information on the work conducted by the principal spray operator on the farm. In particular, the data were required to add to the non-dietary exposure data already collected in **Form 3**. Especially work related activities that took the principal operator into the crop (drilling and filling the seed drill, pruning, picking, etc.) were included wherever possible. Other activities, particularly those during which the farmer was in a tractor or combine, such as ploughing, cultivating the field and harvesting were not as important as it was felt that these did not take the farmer into the crop. Earliest return dates were recorded wherever possible.

EFSA SURVEY 2013														
	FORM 1 - CROPPING DETAILS													
REFERENCE NUMBER	COUNTRY	REGIO	N	SURVEY										
SIZE GROUP	DATE OF VISIT	SURVE	YOR	FUTURE SURVEY										
CROPS		REA (HA)	OPEF	ATOR NUMBER										
01														
02														
03														
04														
05														
07														
08														
09														
10														
11														
12														
13														
14														
15														
16														
17														
18														
20														
21														
22														
23														
24														

FORM 2 - EFSA FARM BUSINESS DETAILS - 2013
Ref.
MULTIPLE FARMS
a) Including the sampled farm how many separate farms do you manage?
b) What is the total cropping area of all farms (ha)
bb) For what percentage of the sampled farm have detailed records been collected?
<b>OPERATORS</b>
c) Excluding contractors how many PPP operators do you have on the farm?
d) What percentage of spraying is carried out by contractor?
e) If a contractor is used please record their name(s) and address ('s) below.
Former Agricultural cont Suray contractor Other
Farmer Agricultural cont. Spray contractor Other
SPRAY DECISIONS
f) Do you use an agronomist or professional advisor to advise on pesticide usage (Y/N)
BUFFER STRIPS
g1) is there a permanent watercourse next to any news on your farm? (1/N)
g2) is there a temporary watercourse (in y unches) next to any next to any next so in your farm? (Y/N)
g() Do you use windbreaks to prevent drift on any fields on your farm? (Y/N)
g5) Do you use in-crop buffer strips to prevent drift on any fields on your farm? (Y/N)
INTEGRATED PEST MANAGEMENT
h) Do you consider yourself to be practising Integrated Pest Management on the farm? (Y/N)
If the answer is Yes, please indicate below the range of IPM practices you follow (use Y or N):
a) Crean rotation (b) Selection of resistant variation (c) Use of manitoring trans
a) Crop rotation b) Selection of resistant varieties c) Use of momoring traps
d) Use of biological control agents e) Use of predictive models/early warning systems
f) Maintaining & increasing populations of g) Selection of pesticides to minimise risk to
beneficial parasites & predators beneficial parasites & predators
Other IPM practices:

E	FSA	SUR	VEY 2013					FO	RM 3	3 -	P	ES	T		[ <b>D</b> ]	E	AP	PP	LICA	<b>TIO</b>	N	FORM
RE	FERENC	E	FIELD CROP				CROP	UMBEI	R 🗌		]			SUR	VEY	YE	AR			FIE	LD	AREA (HA)
SAI INT SIN	MPLED TENDED GLE/MU	FARM ( USE ULTIPL	Y/N) CROP COV IR% PB E (S/M)	/ER /TB%																		
Yr	Date	Crop Stage	Product	Meth app	Vol (l)	RR	R: Amnt	ate Units	Spray Round	Аг	ea T	reat	ed (	Ha)		OP	SP	NZ	Start	No. of Hours	BU	Remarks
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Pes	ticides in	clude in	secticides, acaricides, nematic	ides, mol	luscicide	es, fu	ingicides	, herbici	des, gro	wth	regu	lato	rs, s	eed t	reat	men	ts, se	oil st	erilants	, biologic	al co	ontrol, adjuvants,

E	FSA S	SUR	VEY 2013		FORM 3 (ENV) - PESTICIDE APPLICATION FORM																
RE	FERENC	E	FIELD CROP			CROP NUMBER							SUR	VEY	YE	AR					
SAI INT SIN	MPLED   ENDED GLE/MU	FARM ( USE ULTIPL	Y/N) CROP COV IR% PB E (S/M)			CO! CO! CO! CO!	MBINAT MBINAT MBINAT MBINAT	FION 1 FION 2 FION 3 FION 4			<u></u>	ELD		FIL				r (m)			
Yr	Date	Crop Stage	Product	Meth app	Vol (l)	RR	R: Amnt	ate Units	Spray Round	Аге	ea Ti	reate	ed (Ha)		OP	SP	NZ	Start	No. of Hours	BU	Remarks
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EFSA - SPRAY OPERATOR DETAILS - 2013	FORM 4 - OP1
Please complete a separate OP1 for each spray operator on the farm (including spray contractor	ors)
Holding Ref Number Operator Number	
Age       Gender       Percentage of all spraying undertaken         Full time       Part time       Are you spraying on any other farms?         Years of spraying experience       If Yes, what percentage of all spraying is on the same	Yes No
Relationship to the holding: Farm owner/tenant Farm employee (National) Farm employee (Migrant)	Contractor
SPRAYING QUALIFICATIONS AND TRAINING RECEIVED Do you have a nationally recognised spraying certificate? Please indicate the type and year of the most recent training?	Yes No
Year Training type (Theory, Practical or Both)	
MIXING & LOADING (Including filling with water)       Sprayer       Sprayer       Num       Hrs       Num         1) What is the average time (in hours) spent on each load?       2) On average how many times would you do this in a day?	yer Sprayer Hrs Num Hrs
3) What type of PPE do you wear when mixing liquids?       P	PE 3     PPE 4     PPE 5       PE 3     PPE 4     PPE 5
APPLICATION (Please record the PPE used for each method of application – if unchanged record the PPE 1 I) What type of PPE do you wear?	cord AL) PPE 2 PPE 3
3) What type of PPE do you wear?	PPE 2         PPE 3
5) What type of PPE do you wear?       M_APP       PPE 1         6) How many times would you use these before disposal/washing?	PPE 2         PPE 3
CLEANING THE SPRAYER       Sprayer       Sprayer         1) What is the average number of times the sprayer would be cleaned each year and the average time (in hrs) spent cleaning each sprayer?       Sum Times Hrs       Num Times H	Sprayer Irs Num Times Hrs
2) What type of PPE do you wear? 3) How many times would you use these before disposal/washing?	

EFSA - SPRAY OPERATOR DETAILS - 2	013	FORM	4 (ACT)	) - OP1
Please complete a separate OP1 for each spray operator on the farm	(including spra	y contractors	5)	
Holding Ref Number Operator Number				
WORK ACTIVITIES (Please record the PPE used for each work re related codes and enter these codes under the WORK heading)	lated activity – J	olease refer t	o the list of wo	ork
a) What type of PPE do you wear? b) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	PPE 2	<b>PPE 3</b>
c) What type of PPE do you wear? d) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	<b>PPE 2</b>	<b>PPE 3</b>
e) What type of PPE do you wear? f) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	<b>PPE 2</b>	<b>PPE 3</b>
g) What type of PPE do you wear? h) How many times would you use these before disposal/washing?	WORK	PPE 1	<b>PPE 2</b>	PPE 3
i) What type of PPE do you wear? j) How many times would you use these before disposal/washing?	WORK	PPE 1	<b>PPE 2</b>	<b>PPE 3</b>
k) What type of PPE do you wear? l) How many times would you use these before disposal/washing?	WORK	PPE 1	<b>PPE 2</b>	<b>PPE 3</b>
m) What type of PPE do you wear? n) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	<b>PPE 2</b>	<b>PPE 3</b>
o) What type of PPE do you wear? p) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	<b>PPE 2</b>	<b>PPE 3</b>
q) What type of PPE do you wear? r) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	PPE 2	<b>PPE 3</b>
s) What type of PPE do you wear? t) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	PPE 2	<b>PPE 3</b>
u) What type of PPE do you wear? v) How many times would you use these before disposal/washing?	WORK	<b>PPE 1</b>	<b>PPE 2</b>	PPE 3

EFSA – SPRAYING EQUIPMENT DETAIL	LS – 2013 FORM 5 - SP1
Please complete a separate SP1 for each sprayer and photograph the sprayer Holding Ref Number	Sprayer Number
Manufacturers name & model	Ownership
Is this sprayer tested as part of a sprayer testing scheme?	Yes No
<b>SPRAYER DETAILS</b> 1) What percentage of all spraying on the sampled farm is carried out 2) On how many farms is this sprayer used? 3) What is the typical speed at which you spray (kmph)?	t with this sprayer?
4) Tank capacity (l): Main 5) Boom wid Auxiliary 6) Boom heig Hand wash 6a) Measure	th (m) 7) Age (years) ght (m) 7a) Cab type d, estimated or irrelevant? (M/E/I)
SPRAYER TYPE - Using the codes provided in the instructions pl	ease record the relevant sprayer code
8) Other sprayer type – Including modified equipment - <i>Please includ</i>	le as many details as possible
PESTICIDE FILLING SYSTEMS USED: record %       Direct         Suction lance       Closed transfer system	t pour Induction bowl Other
<b>NOZZLE TYPE &amp; USE:</b> - Please record the set (a unique comb (FF), air-inclusion (AI), twin-jet (TJ), hollow cone (HC), make, pr months) for each set of nozzles used on the sprayer.	ination of type, make and pressure) type (flat fan ressure (bar), replacement frequency (number of
SET TYPE TRADE NAME/MANUFACTURER	PRESSURE (BAR) REPLACEMENT TIME

EFSA WORK ACTIVITY DETAILS 2013								FORM	<b>FORM 6 – OP0</b>					
EFF	RENG	E									_			
					OPI	ERA	тоі	R NI	UMBER					
VEI	RAGE	DAI	LY/	WE)	<b>EKL</b>	YH	ou	RS						
N						APF	ł			JUL	00	т		
EB					Į.	MA	Y			AUG	NO	v		
AR						JUN	1			SEP	DE	с		
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#### Farmer contact and recruitment

The identification of suitable farms and subsequent contact and agreement to participate in the survey was an important component for successful data collection. However, continued farmer participation from initial contact right through to the collection of the last piece of required information was crucial. Farmers' willingness to participate was dependent on a number of factors including their history of participation in previous surveys, the level of detail in existing PPP records and availability of their time to complete the lengthy survey.

During the initial visit, all **forms** except **3** and **6** were filled in and it was explained to farmers how to record data in **form 3** and **6** for the upcoming year. After the initial visit, contact was maintained by means of email or phone on a monthly basis. The ease of this contact varied among farmers, but these contacts were – in general – very useful to remind farmers to fill in the **forms 3** and **6**. Most farmers were able to confirm that the necessary data were being or would be recorded in **form 3** and **6**. One farmer informed us that he was no longer willing to participate and another requested a second visit as some problems had arisen. Farmers were asked to send intermediate results during the monthly contacts by means of email or phone to allow the quality of the records being checked. Some were able to send intermediate results whereas others would only fill in the records at the end of the year.

# 2. Details of the active substances and products applied on farms

Table A-1 Overview	of summed	hectares	treated	(ha)	per	crop	group	surveyed	in 2	2013	(Flanders;
based on Form 3).											

Crop group	AM	SD	Median	min	max
arable crops	8.4	7.1	7.1	0.2	30.5
grassland and fodder	4.9	3.1	3.8	1.3	9.6
maize	153.2	142.8	119.2	2.5	522.6
potato	267.0	364.6	101.4	0.1	1593.9
soft fruit	0.7	0.7	0.5	0.1	2.5
sugar beet	158.6	247.3	48.3	3.3	827.6
vegetables	45.0	42.3	30.0	1.6	158.5
wheat	86.6	93.1	50.4	0.6	409.9

AM = average mean; SD = standard deviation; min = minimum; max = maximum.

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	substances per crop	group hecta	re treated	surveyed	l in 2013 (Fla	nde	rs; ba	sed	on <b>F</b>	orm 3	<b>3</b> ).		
	Table A-2 Overview	of applied	amount o	of active	substances	(kg	a.s.)	for	the	top	5 to	9	active

Crop group	Active substance	n	AM	SD	Median	min	max
arable crops	iodosulfuron-methyl (iodosulfuron-	2	0.02	0.00	0.02	0.02	0.02
	methyl including salts, expressed as						
	iodosulfuron-methyl)						
arable crops	mefenpyr-diethyl <sup>a</sup>	2	0.2	0.0	0.2	0.2	0.2
arable crops	mepiquat	7	1.1	0.6	1.1	0.1	2.0
arable crops	prohexadione	6	0.2	0.1	0.2	0.0	0.3
arable crops	prothioconazole	6	0.5	0.4	0.3	0.1	1.0
arable crops	tebuconazole	5	0.4	0.4	0.2	0.1	1.0
grassland and fodder	clopyralid	2	0.2	0.1	0.2	0.1	0.3
grassland and fodder	florasulam	3	0.0	0.0	0.0	0.0	0.0
grassland and fodder	fluroxypyr	2	0.4	0.3	0.4	0.2	0.6
grassland and fodder	glyphosate	2	1.2	1.6	1.2	0.0	2.3
grassland and fodder	МСРА	4	1.7	0.8	1.5	1.0	2.8
maize	dimethenamid-P	27	5.6	5.6	3.2	0.8	22.0
maize	isoxadifen ethyl <sup>a</sup>	22	0.4	0.4	0.2	0.0	1.2
maize	mesotrione (sum of mesotrione	44	0.5	0.8	0.2	0.0	4.6
	and MNBA (4-methylsulfonyl-2-						
	nitrobenzoic acid), expressed as						
	mesotrione)						
maize	nicosulfuron	53	0.2	0.2	0.1	0.0	0.9
maize	tembotrione	22	0.8	0.8	0.4	0.0	2.5
maize	terbuthylazine	69	3.4	3.7	1.8	0.2	16.7
potato	boscalid	66	0.6	0.6	0.3	0.0	2.7
potato	cyazofamid	148	0.7	0.7	0.5	0.0	3.3
potato	cymoxanil	195	2.8	5.8	0.5	0.0	37.6
potato	mancozeb	137	8.5	10.3	5.0	0.2	57.2
potato	propamocarb	136	6.1	7.7	2.8	0.3	35.2
potato	pyraclostrobin	66	0.2	0.2	0.1	0.0	0.7

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Crop group	Active substance	n	AM	SD	Median	min	max
soft fruit	boscalid	3	0.2	0.1	0.2	0.1	0.2
soft fruit	kresoxim-methyl	2	0.1	0.0	0.1	0.1	0.1
soft fruit	lambda-cyhalothrin	2	0.0	0.0	0.0	0.0	0.1
soft fruit	metamitron	4	0.2	0.0	0.2	0.2	0.2
soft fruit	penconazole	2	0.0	0.0	0.0	0.0	0.0
soft fruit	phenmedipham	5	0.1	0.0	0.1	0.1	0.1
soft fruit	pyraclostrobin	3	0.0	0.0	0.1	0.0	0.1
soft fruit	quinoxyfen	3	0.1	0.0	0.1	0.1	0.1
soft fruit	tepraloxydim	3	0.0	0.0	0.0	0.0	0.0
sugar beet	chloridazon	51	4.5	5.8	2.7	0.2	31.5
sugar beet	desmedipham	65	0.3	0.3	0.2	0.0	1.1
sugar beet	ethofumesate	139	0.9	0.9	0.7	0.0	3.8
sugar beet	metamitron	129	3.4	2.8	2.8	0.2	13.1
sugar beet	phenmedipham	136	1.1	1.5	0.6	0.0	8.9
vegetables	clomazone	20	0.9	2.0	0.3	0.2	8.1
vegetables	dimethoate	21	1.4	0.7	1.4	0.5	3.0
vegetables	linuron	18	2.1	2.6	1.2	0.5	11.3
vegetables	pendimethalin	19	3.9	2.6	3.5	1.2	9.0
vegetables	tebuconazole	22	1.4	0.7	1.2	0.4	2.8
wheat	bixafen	21	1.0	1.1	0.7	0.0	3.8
wheat	chlormequat	30	8.0	7.4	5.3	0.1	22.3
wheat	iodosulfuron-methyl (iodosulfuron-	22	59.7	274.0	0.0	0.0	1286.3
	methyl including salts, expressed as						
	iodosulfuron-methyl)						
wheat	mefenpyr-diethyl <sup>a</sup>	22	593.6	2740.5	0.3	0.0	12862.5
wheat	prothioconazole	41	1.5	1.6	1.1	0.0	7.5
wheat	tebuconazole	30	1.2	1.0	1.1	0.0	3.8

n = number of active substances; AM = average mean; SD = standard deviation; min = minimum; max = maximum; <sup>a</sup>safener.

# 3. Additional PPP application data

Table A-3 Average application rates (kg/ha) of active substances (a.s.) per crop of the ERA fields surveyed in 2013 (Flanders; based on **Form 3**).

Crop	Active substance Avera	ge active substance application rate (kg a.s./ha)
maize	bromoxynil	0.201
maize	dicamba	0.158
maize	dimethenamid-P	0.725
maize	florasulam	0.001
maize	flufenacet	0.403
maize	fluroxypyr	0.063
maize	isoxadifen ethyl <sup>a</sup>	0.041
maize	mesotrione (sum of mesotrione and	MNBA (4-methylsulfonyl-2- 0.073
	nitrobenzoic acid), expressed as me	sotrione)
maize	nicosulfuron	0.027

Crop	Active substance	Average active substance application rate (kg a.s./ha)
maize	pethoxamid	0.570
maize	S-metolachlor	0.484
maize	sulcotrione	0.240
maize	tembotrione	0.083
maize	terbuthylazine	0.445
maize	topramezone	0.046
maize	tritosulfuron	0.034
potato	acetamiprid	0.050
potato	aclonifen	1.281
potato	ametoctradin	0.223
potato	amisulbrom	0.060
potato	azoxystrobin	0.061
potato	bentazone	0.144
potato	benthiavalicarb	0.028
potato	boscalid	0.056
potato	carfentrazone-ethyl	0.060
potato	chlorpropham	0.012
potato	clomazone	0.072
potato	cyazofamid	0.095
potato	cymoxanil	0.225
potato	dimethomorph	0.177
potato	diquat	0.489
potato	esfenvalerate	0.005
potato	famoxadone	0.140
potato	flonicamid	0.085
potato	fluazinam	0.180
potato	flufenacet	0.480
potato	fluopicolide	0.090
potato	glufosinate-ammonium	0.450
potato	lambda-cyhalothrin	0.008
potato	linuron	0.532
potato	maleic hydrazide	3.010
potato	mancozeb	1.213
potato	mandipropamid	0.150
potato	maneb	1.661
potato	metazachlor	0.650
potato	metribuzin	0.308
, potato	oxamyl	2.500
, potato	pencycuron	0.250
, potato	pendimethalin	0.634
potato	propamocarb	0.902
potato	prosulfocarb	2.646
potato	, pyraclostrobin	0.014
potato	rimsulfuron	0.010
potato	tepraloxydim	0.010
potato	thiacloprid	0.096
potato	, zeta-cypermethrin	0.010

Crop	Active substance A	verage active substance application rate (kg a.s./ha)
potato	zoxamide	0.148
sugar beet	chloridazon	0.670
sugar beet	clethodim	0.091
sugar beet	clomazone	0.027
sugar beet	clopyralid	0.059
sugar beet	deltamethrin (cis-deltamethrin)	0.006
sugar beet	desmedipham	0.050
sugar beet	difenoconazole	0.101
sugar beet	dimethenamid-P	0.356
sugar beet	epoxiconazole	0.048
sugar beet	ethofumesate	0.146
sugar beet	fenpropidin	0.375
sugar beet	glyphosate	1.800
sugar beet	lambda-cyhalothrin	0.007
sugar beet	lenacil	0.250
sugar beet	metamitron	0.617
sugar beet	methiocarb	0.200
sugar beet	metolachlor (metolachlor inclue	ling other mixtures of constituent isomers 0.830
	including S-metolachlor (sum of	isomers))
sugar beet	metsulfuron-methyl	0.200
sugar beet	phenmedipham	0.165
sugar beet	pirimicarb	0.125
sugar beet	propiconazole	0.105
sugar beet	pyraclostrobin	0.129
sugar beet	quinmerac	0.153
sugar beet	tepraloxydim	0.025
sugar beet	tetraconazole	0.088
sugar beet	tri-allate	0.152
sugar beet	triflusulfuron-methyl	0.010

<sup>a</sup>safener.

Table A-4 Average application rates (kg/ha) of formulated mixtures (defined as a mixture of one or more active substances with diluents, carriers and other materials to form the packaged product, Glass et al., 2012) per crop of the ERA fields surveyed in 2013 (Flanders; based on **Form 3**).

Crop	Active substance	Average formulated mixture application rate (kg/ha)
maize	bromoxynil	0.201
maize	dicamba	0.144
maize	dicamba/tritosulfuron	0.199
maize	dimethenamid-P/terbuthyl	azine 0.901
maize	dimethenamid-P	0.756
maize	florasulam/fluroxypyr	0.063
maize	flufenacet/terbuthylazine	1.084
maize	isoxadifen ethyl <sup>a</sup> /tembotric	one 0.124

Crop	Active substance Average formulated mixture applicatio	n rate (kg/ha)
maize	mesotrione (sum of mesotrione and MNBA (4-methylsulfonyl-2-	0.104
maize	nitrobenzoic acid), expressed as mesotrione) mesotrione (sum of mesotrione and MNBA (4-methylsulfonyl-2-	0.329
	nitrobenzoic acid), expressed as mesotrione)/terbuthylazine	
maize	nicosulfuron	0.027
maize	pethoxamid	0.570
maize	S-metolachlor/terbuthylazine	0.775
maize	sulcotrione	0.240
maize	topramezone/dimethenamid-P	0.814
potato	acetamiprid	0.050
potato	aclonifen	1.281
potato	ametoctradin/dimethomorph	0.411
potato	amisulbrom	0.060
potato	azoxystrobin	0.061
potato	bentazone	0.144
potato	carfentrazone-ethyl	0.060
potato	chlorpropham	0.012
potato	clomazone	0.072
potato	clomazone/metribuzin	0.352
potato	cyazofamid	0.095
potato	cymoxanil	0.487
potato	cymoxanil/famoxadone	0.280
potato	cymoxanil/mancozeb	1.239
potato	cymoxanil/propamocarb	1.062
potato	dimethomorph/mancozeb	1.521
potato	diquat	0.489
potato	esfenvalerate	0.005
potato	flonicamid	0.085
potato	fluazinam	0.177
potato	fluazinam/dimethomorph	0.400
potato	fluopicolide/propamocarb	0.998
potato	glufosinate-ammonium	0.450
potato	lambda-cyhalothrin	0.008
potato	linuron	0.538
potato	linuron/clomazone	0.443
potato	maleic hydrazide	3.010
potato	mancozeb	1.326
potato	mancozeb/ametoctradin	0.560
potato	mancozeb/benthiavalicarb	1.148
potato	mancozeb/zoxamide	1.136
potato	mandipropamid	0.150
potato	maneb	1.661
potato	metazachlor	0.650
potato	metribuzin	0.245
potato	metribuzin/flufenacet	0.830
potato	oxamyl	2.500

Crop	Active substance Average	formulated mixture application rate (kg/ha)
potato	pencycuron	0.250
potato	pendimethalin	0.640
potato	prosulfocarb	2.646
potato	pyraclostrobin/boscalid	0.070
potato	rimsulfuron	0.010
potato	tepraloxydim	0.010
potato	thiacloprid	0.096
potato	zeta-cypermethrin	0.010
sugar beet	chloridazon	0.588
sugar beet	clethodim	0.091
sugar beet	clomazone	0.027
sugar beet	clopyralid	0.059
sugar beet	deltamethrin	0.008
sugar beet	desmedipham/ethofumesate/phenmed	ipham 0.244
sugar beet	dimethenamid-P	0.356
sugar beet	ethofumesate	0.169
sugar beet	fenpropidin/difenoconazole	0.475
sugar beet	glyphosate	1.800
sugar beet	lambda-cyhalothrin	0.008
sugar beet	lambda-cyhalothrin/pirimicarb	0.131
sugar beet	lenacil	0.250
sugar beet	metamitron	0.617
sugar beet	methiocarb	0.200
sugar beet	metolachlor (metolachlor including othe	er mixtures of constituent isomers 0.830
	including S-metolachlor (sum of isomers	5))
sugar beet	metsulfuron-methyl	0.200
sugar beet	phenmedipham	0.265
sugar beet	phenmedipham/ethofumesate	0.312
sugar beet	propiconazole/difenoconazole	0.210
sugar beet	pyraclostrobin/epoxiconazole	0.177
sugar beet	quinmerac/chloridazon	1.068
sugar beet	tepraloxydim	0.025
sugar beet	tetraconazole	0.088
sugar beet	tri-allate	0.152
sugar beet	triflusulfuron-methyl	0.010

<sup>a</sup>safener.

Crop	Off-field margin	In-field margin	Width (m)	Number of fields
maize	buildings	no margin	0	1
maize	ditch	no margin	0	6
maize	dry ditch	no margin	0	1
maize	hedgerow	no margin	0	1
maize	orchard	no margin	0	1
maize	other field	herbaceous margin	12	1
maize	other field	no margin	0	21
maize	pasture (grassland)	no margin	0	9
maize	river	no margin	0	1
maize	roads and other artificial structures	no margin	0	19
maize	stream	herbaceous margin	12	1
maize	stream	no margin	0	4
maize	woodland, spinneys, copses, forests, etc.	no margin	0	9
potato	buildings	no margin	0	4
potato	ditch	no margin	0	10
potato	footpath	no margin	0	1
potato	other field	no margin	0	18
potato	pasture (grassland)	no margin	0	10
potato	roads and other artificial structures	no margin	0	16
potato	stream	no margin	0	5
potato	woodland, spinneys, copses, forests, etc.	no margin	0	6
sugar beet	arable field	no margin	0	1
sugar beet	buildings	no margin	0	4
sugar beet	ditch	no margin	0	6
sugar beet	fallow field	no margin	0	2
sugar beet	grass field	no margin	0	1
sugar beet	grass strip	no margin	0	1
sugar beet	maize field	no margin	0	1
sugar beet	other field	no margin	0	19
sugar beet	pasture (grassland)	no margin	0	5
sugar beet	pond	no margin	0	1
sugar beet	roads and other artificial structures	no margin	0	15
sugar beet	sown grass	no margin	0	1
sugar beet	stream	no margin	0	5
sugar beet	woodland, spinneys, copses, forests, etc.	no margin	0	4

Table A-5 Details of the off- and in-field margins per crop of the ERA fields surveyed in 2013 (Flanders; based on **Form 3**).

# 4. Detailed sprayer characteristics

Table A-6 Spraver	characteristics	surveyed in 2013	(Flanders: ba	ased on Form 5).
Table A 0 Sprayer	characteristics	Surveyeu in 2015	(110110013, 50	13cu on <b>10mm 3</b> .

	,					
Sprayer characteristics	n	AM	SD	Median	min	max
average speed of use (km/h)	36	6.9	1.7	7	5	12
boom height (m)	37	0.6	0.3	0.5	0.2	2
hand wash tank capacity (I)	37	18.2	60.1	0	0	350
nozzle pressure (bar)	61	3.1	1.1	3	2	6
percentage of all filling using a closed transfer system (%)	37	2.7	16.4	0	0	100
percentage of all filling using a suction lance (%)	37	0	0	0	0	0
percentage of all filling using direct pour (%)	37	52.6	49.2	90	0	100
percentage of all filling using induction bowl (%)	37	44.7	48.9	0	0	100
percentage of all spraying on the sampled farm using a certain sprayer (%)	36	96.6	15.9	100	5	100

n = number of sprayers/nozzles; AM = average mean; SD = standard deviation; min = minimum; max = maximum.

Table A-7 Sprayer characteristics surveyed in 2013 (Flanders): number of farms on which the sprayer was used (based on **Form 5**).

Number of farms	Frequency
1	30
2	5
3	1
99 (missing)	1

Table A-8 Sprayer characteristics surveyed in 2013 (Flanders): number of nozzle sets on the sprayer and nozzle type (based on **Form 5**).

Number of nozzle sets	Frequency	Nozzle type	Frequency
1	37	Air Inclusion	9
2	17	Flat Fan	45
3	7	Hollow cone	3
4	1	Off centre spray tips-herbicide applications	1
		Tee Jet	1
		Twin Jet	2
		unknown	1

#### 5. Detailed operator characteristics

Table A-9 Overview of summed exposure duration (h) per holding per active substance surveyed in 2013 (Flanders; based on **Form 3**).

n	AM	SD	min	max
101	290.1	154.8	198	792
	1	60		

n = number of active substances; AM = average mean; SD = standard deviation; min = minimum; max = maximum.

Table A-10 Overview of summed amount of active substance applied (kg) per holding per active substance surveyed in 2013 (Flanders; based on **Form 3**).

n	AM	SD	min	max
120	7.1	11.4	0.1	51.2

n = number of active substances; AM = average mean; SD = standard deviation; min = minimum; max = maximum.

Table A-11 Operator characteristics surveyed in 2013 (Flanders): percentage (%) of all spraying conducted by operator on the sampled farm (based on **Form 4**).

n	AM	SD	Median	min	max
36	92.4	19.9	100	10	100

n = number of operators; AM = average mean; SD = standard deviation; min = minimum; max = maximum.

Table A-12 Operator characteristics surveyed in 2013 (Flanders): type of certification (based on **Form 4**).

Certification type	Code	Frequency
	(null)	2
both	BO	4
practical training with use of equipment	PR	1
theory (desk based)	TH	27
unknown	UN	2

Table A-13 Operator characteristics surveyed in 2013 (Flanders): year of most recent training (based on **Form 4**).

Year of most recent training	Frequency
	3
99 (missing)	1
1968	1
1998	1
2006	1
2007	1
2011	1
2012	14
2013	13

Type of activity	PPE type	Frequency
application	coat-padded	1
application	disposable filtering half mask	2
application	full face mask	1
application	full length trousers	1
application	gloves-fabric/leather	1
application	gloves-latex	1
application	gloves-neoprene	1
application	gloves-nitrile	1
application	gloves-vinyl (PVA)	3
application	half mask, reusable with filters	3
application	leather/fabric boots	21
application	long sleeved shirt	1
application	rubber boots	15
application	valved filtering half mask	1
application	work wear: breathable (cotton/polyester)	31
application	work wear: rainwear 1 piece (vinyl, Goretex, etc.)	2
application	work wear: rainwear 2 piece (vinyl, Goretex, etc.)	1
cleaning	disposable filtering half mask	2
cleaning	full face mask	1
cleaning	full length trousers	1
cleaning	gloves-butyl rubber	1
cleaning	gloves-fabric/leather	1
cleaning	gloves-latex	2
cleaning	gloves-neoprene	6
cleaning	gloves-nitrile	4
cleaning	gloves-non-specified rubber	1
cleaning	gloves-vinyl (PVA)	6
cleaning	half mask, reusable with filters	1
cleaning	leather/fabric boots	21
cleaning	long sleeved shirt	1
cleaning	rubber boots	14
cleaning	waterproof leggings	1
cleaning	work wear: breathable (cotton/polyester)	28
cleaning	work wear: rainwear 1 piece (vinyl, Goretex, etc.)	5
cleaning	work wear: rainwear 2 piece (vinyl, Goretex, etc.)	2
mixing and loading	bib and brace	2
mixing and loading	disposable filtering half mask	6
mixing and loading	face shield	2
mixing and loading	full face mask	6
mixing and loading	full length trousers	2
mixing and loading	gloves-butyl rubber	2
mixing and loading	gloves-fabric/leather	2
mixing and loading	gloves-latex	4

Table A-14 Operator characteristics surveyed in 2013 (Flanders): type of personal protective equipment (PPE) worn during operator activities (based on **Form 4**).

Type of activity	PPE type	Frequency
mixing and loading	gloves-neoprene	14
mixing and loading	gloves-nitrile	12
mixing and loading	gloves-nitrile latex	2
mixing and loading	gloves-non-specified rubber	4
mixing and loading	gloves-vinyl (PVA)	20
mixing and loading	half mask, reusable with filters	18
mixing and loading	leather/fabric boots	42
mixing and loading	long sleeved shirt	2
mixing and loading	rubber boots	29
mixing and loading	valved filtering half mask	4
mixing and loading	work wear: breathable (cotton/polyester)	60
mixing and loading	work wear: rainwear 1 piece (vinyl, Goretex, etc.)	6
mixing and loading	work wear: rainwear 2 piece (vinyl, Goretex, etc.)	2
work related activity	disposable filtering half mask	1
work related activity	full face mask	1
work related activity	full length trousers	8
work related activity	gloves-neoprene	2
work related activity	gloves-nitrile	2
work related activity	gloves-vinyl (PVA)	1
work related activity	half mask, reusable with filters	2
work related activity	leather boots	20
work related activity	long sleeved shirt	8
work related activity	rubber boots	9
work related activity	shorts	2
work related activity	t-shirt	5
work related activity	valved filtering half mask	1
work related activity	work wear: breathable (cotton/polyester)	28

Table A-15 Operator characteristics surveyed in 2013 (Flanders): average daily hours (h) worked during each month (based on **form 6**).

Month	n	AM	SD	Median	min	max
January	19	2.16	3.25	0	0	9
February	20	1.73	2.76	0	0	8
March	19	2.41	3.03	0.3	0	8
April	16	5.99	3.26	6.5	0.5	12
May	16	5.46	3.63	6	0.05	12
June	15	5.48	4.30	7	0.03	14
July	14	5.43	4.16	5.5	0.1	10
August	15	6.05	4.24	8	0.4	14
September	15	5.80	4.51	6.4	0	14
October	17	5.33	4.46	4.3	0	12
November	18	3.74	4.13	1.6	0	11
December	19	2.49	3.41	0.4	0	10

n = number of operators; AM = average mean; SD = standard deviation; min = minimum; max = maximum.

# Appendix A

Month	n	AM	SD	Median	min	max
January	19	14.53	22.00	0	0	63
February	20	11.57	18.35	0	0	56
March	19	15.98	20.49	2	0	56
April	16	39.63	20.37	42	4	72
May	16	36.05	23.75	39	0.35	72
June	15	35.85	27.74	42	0.2	84
July	14	36.07	27.99	38.5	1	70
August	15	39.70	27.02	56	3	84
September	15	37.95	28.94	45	0	84
October	17	34.99	29.31	30	0	77
November	18	24.94	27.83	11	0	77
December	19	16.47	23.01	2	0	70

Table A-16 Operator characteristics surveyed in 2013 (Flanders): average weekly hours (h) worked during each month (based on **Form 6**).

n = number of operators; AM = average mean; SD = standard deviation; min = minimum; max = maximum.

# Appendix B

# 1. Spray records of various crops

Date	PPP group	Product	Active substance
22/05	herbicide	Bonalan	benfluralin
	herbicide	Centium 36 CS	clomazone
	herbicide	Dual Gold	S-metolachlor
14/06	herbicide	Corum	bentazone
			imazamox
23/07	fungicide	Rovral SC	iprodione
	fungicide	Topsin M 500 SC	thiophanate-methyl
07/08	fungicide	Rovral SC	iprodione
	fungicide	Topsin M 500 SC	thiophanate-methyl
18/09	herbicide	Roundup	glyphosate

Table B-1 Spray records of brown bean.

Table B-2 Spray records of chicory.

Date	PPP group	Product	Active substance
11/04	herbicide	Bonalan	benfluralin
13/04	herbicide	Kerb 400 SC	propyzamide
24/04	herbicide	Safari	triflusulfuron-methyl
	fungicide	Tifex	epoxiconazole
	surfactant	Trend 90	isodecyl alcohol ethoxylate
	herbicide	Kerb 400 SC	propyzamide
	herbicide	C.I.P.C. Protex	chlorpropham
29/05	insecticide	Karate Zeon	lambda-cyhalothrin
30/05	herbicide	Safari	triflusulfuron-methyl
	fungicide	Tifex	epoxiconazole
	surfactant	Trend 90	isodecyl alcohol ethoxylate
16/06	herbicide	Biathlon	tritosulfuron
	fungicide	Tifex	epoxiconazole
30/06	herbicide	Dual Gold	S-metolachlor

Table B-3 Spray records of spring barley.

Date	PPP group	Product	Active substance	
28/4	herbicide	Primstar	florasulam	
			fluroxypyr	
29/4	herbicide	U46 M750	MCPA	
22/5	growth regulator	Moddus	trinexapac-ethyl	

Date	PPP group	Product	Active substance
25/03	herbicide	Tornado	metamitron
12/04	herbicide	Betanal Carrera	desmedipham
			phenmedipham
	herbicide	Tornado	metamitron
28/04	herbicide	Betanal Carrera	desmedipham
			phenmedipham
	herbicide	Tornado	metamitron
	herbicide	Frontier Elite	dimethenamid-P
	herbicide	Fusilade Max	fluazifop-P-butyl
14/05	herbicide	Betanal Carrera	desmedipham
			phenmedipham
	herbicide	Tornado	metamitron
09/08	fungicide	Retengo Plus	epoxiconazole
			pyraclostrobin

Table B-4 Spray records of sugar beet.

# Table B-5 Spray records for both plots of triticale.

Date	PPP group	Product	Active substance	
20/11	herbicide	Bacara	diflufenican	
			flurtamone	
09/04	fungicide	Cherokee	chlorothalonil	
			cyproconazole	
			propiconazole	
	growth regulator	Cycocel 75	chlormequat	
27/4	fungicide	Pallazo	epoxiconazole	
			fenpropimorph	
			metrafenone	
22/5	fungicide	Evora XPro	bixafen	
			prothioconazole	
			tebuconazole	

Date	PPP group	Product	Active substance
2/4	herbicide	Axial	cloquintocet-mexyl <sup>a</sup>
			pinoxaden
	herbicide	Biathlon	tritosulfuron
	herbicide	Spitfire	florasulam
			fluroxypyr
	growth regulator	Moddus	trinexapac-ethyl
	fungicide	Evora xPro	bixafen
			prothioconazole
			tebuconazole
24/04	growth regulator	Terpal	ethephon
			mepiquat chloride
	fungicide	Evora xPro	bixafen
			prothioconazole
			tebuconazole
02/07	herbicide	Roundup	glyphosate

Table B-6 Spray records for the plot of winter barley (BBCH 85-8
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BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale); <sup>a</sup>safener.

Table B-7 Spray records of winter wheat.

Date	PPP group	Product	Active substance
14/3	herbicide	Atlantis WG	iodosulfuron-methyl-sodium
			metenpyr-diethyl <sup>*</sup>
			mesosulfuron-methyl
	additive	Fieldor max	ethoxylated triglyceride 10 EO
	herbicide	Capri duo	cloquintocet-mexyl <sup>a</sup>
			florasulam
			pyroxsulam
29/3	growth regulator	Cycocel 75	chlormequat
21/4	growth regulator	Cycocel 75	chlormequat
	fungicide	Allegro	epoxiconazole
			kresoxim-methyl
24/5	fungicide	Evora XPro	bixafen
			prothioconazole
			tebuconazole
a .			

<sup>a</sup>safener.

Crop	Number of applications	Product	PPP group	Active substance
maize	1	Laudis	herbicide	isoxadifen ethyl <sup>a</sup>
			herbicide	tembotrione
	1	Aspect T	herbicide	flufenacet
			herbicide	terbuthylazine
	1	Samson Extra 60 OD	herbicide	nicosulfuron
onion see	ed 1	Agro-Mancozeb 80 WP	fungicide	mancozeb
	1	Fandango	fungicide	fluoxastrobin
			fungicide	prothioconazole
rye	2	Cycocel 75	growth regulator	chlormequat
	1	Ceando	fungicide	epoxiconazole
			fungicide	metrafenone
spelt	1	Bacara	herbicide	diflufenican
			herbicide	flurtamone
	1	Cycocel 75	growth regulator	chlormequat
	1	Evora xPro	fungicide	bixafen
			fungicide	prothioconazole
			fungicide	tebuconazole
	1	Palazzo	fungicide	epoxiconazole
			fungicide	fenpropimorph
			fungicide	metrafenone

Table B-8 Spray records of maize, onion seed, rye and spelt.

<sup>a</sup>safener.

## 2. Overview of examined active substances on LCMS-MS

Table B-9 Overview of quantified active substances on LCMS-MS with corresponding device parameters.

Active substance	Parent ion (m/z)	1 <sup>st</sup> Transition (m/z)	2 <sup>nd</sup> Transition (m/z)
azoxystrobin	404.0	372.0	329.0
boscalid	342.9	307.0	139.9
carbofuran	222.1	165.1	123.0
cyprodinil	226.0	93.0	108.0
fludioxonil	249.1	158.1	229.1
hexythiazox	353.0	168.1	228.1
imazalil	297.0	159.0	69.0
metalaxyl	280.1	220.1	192.1
methiocarb	226.0	121.0	169.0
methiocarb sulfone	258.1	122.1	107.1
methiocarb sulfoxide	242.0	185.0	122.0
piperonyl butoxide	356.3	176.9	119.0
pirimicarb	239.1	72.0	182.1
prothioconazole	344.0	326.0	189.0
pyraclostrobin	388.1	163.0	193.9
trifloxystrobin	409.0	186.0	145.0

## 3. Results of monitoring

Table B-10 Residue levels (µg/kg plant material) of fludioxonil found in winter wheat (BBCH 85-89).

Sampling	Residue level (µg a.s./kg plant material)
sample 1	1.44
sample 2	14.9

a.s. = active substance; BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale).

Table B-11 Residue levels (µg/kg plant material) of fludioxonil found in winter wheat (BBCH 14-16).

Sampling	Residue level (µg a.s./kg plant material)
sample 1	31.7
sample 2	28.5
sample 3	18.5
sample 4	1.14

a.s. = active substance; BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale).

Table B-12 Residue levels ( $\mu$ g/kg plant material) of methiocarb, methiocarb sulfone and methiocarb sulfoxide found in the roots of maize (BBCH 85-89).

Active substance	Sampling	Residue level (µg a.s./kg plant material)
methiocarb	sample 1	2.02
	sample 2	-
	sample 3	-
	sample 4	-
methiocarb sulfone	sample 1	-
	sample 2	-
	sample 3	-
	sample 4	-
methiocarb sulfoxide	sample 1	0.736
	sample 2	5.75
	sample 3	-
	sample 4	5.24

a.s. = active substance; BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale).

Active substance	Sampling	Residue level (µg a.s./kg plant material)
methiocarb	sample 1	-
	sample 2	-
	sample 3	-
methiocarb sulfone	sample 1	31.5
	sample 2	37.8
	sample 3	29.3
	sample 4	-
methiocarb sulfoxide	sample 1	0.818
	sample 2	1.18
	sample 3	-

Table B-13 Residue levels ( $\mu$ g/kg plant material) of methiocarb, methiocarb sulfone and methiocarb sulfoxide found in the lower leaves of maize (BBCH 85-89).

a.s. = active substance; BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale).

Table B-14 Residue levels ( $\mu$ g/kg plant material) of methiocarb, methiocarb sulfone and methiocarb sulfoxide found in the top leaves of maize (BBCH 85-89).

Active substance	Sampling	Residue level (µg a.s./kg plant material)
methiocarb	sample 1	-
	sample 2	-
	sample 3	-
methiocarb sulfone	sample 1	40.9
	sample 2	42.0
	sample 3	61.5
	sample 4	41.5
methiocarb sulfoxide	sample 1	-
	sample 2	-
	sample 3	-

a.s. = active substance; BBCH = Biologische Bundesanstalt, Bundessortenamt und Chemische Industrie (BBCH-scale).

# Appendix C

# 1. Survey questionnaire

# <u>Q1. Gender</u>

- o Male
- o Female

# <u>Q2. Age</u>

- o < 21 years
- o 21-30 years
- o 31-40 years
- o 41-60 years
- o > 61 years

# Q3. Postal code

# Q4. Profession

- o Student
- o Labourer
- o Clerk
- o Executive
- o Self-employed person
- o Liberal profession
- o Retired person
- o Job-seeker
- o Other

# Q5. Is your profession related to agriculture?

- o Yes
- o No

# Q6. Highest education

- o Primary school
- o Secondary school
- o High school
- o University

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## Q7. Do you have children?

- o Yes
- o No

<u>Q8. Plant protection products (PPPs) are products used to control diseases, pests or weeds</u> both indoors and outdoors. Products targeting humans and animals (e.g. antifungal medicines or animal drugs against fleas) are not included in this survey. Do you use PPPs?

- o Yes
- o No

If yes, go to question 10. Otherwise, answer the following question.

#### Q9. You are not using PPPs. Why not? (several answers possible)

- o PPPs are too expensive
- o Many risks are associated with the use of PPPs
- o Products treated with PPPs are not healthy
- o The use of PPPs is bad for the environment
- o The use of PPPs is unnecessary
- o Other

#### Q10. How many different products do you use to control INSECTS?

- o 1-5
- o 5-10
- o More than 10
- o None

#### Q11. How many different products do you use to control MOSSES?

- o 1-5
- o 5-10
- o More than 10
- o None

#### Q12. How many different products do you use to control WEEDS?

- o 1-5
- o 5-10
- o More than 10
- o None

## Q13. How many different products do you use to control FUNGI?

- o 1-5
- o 5-10
- o More than 10
- o None

## Q14. If you know the used products by name, which products do you use?

- o Metaldehyde (e.g. Metason)
- o Fluroxypyr (e.g. Silvanet, Weedol, Luoxyl Extra)
- o Glyphosate (e.g. RoundUp)
- o Pyrethrins (e.g. Pyrethro-Pur)
- o Other
- o I do not know

#### Q15. Where do you use PPPs? (several answers possible)

- o Lawn
- o Greenhouse
- o Hedge
- o Flowerbed
- o Vegetable garden
- o Terrace
- o Driveway
- o Orchard
- o Indoor plants/Indoors

#### Q16. How many times a year do you apply PPPs?

- o 1-5 times
- o 5-10 times
- o More than 10 times

#### Q17. Do you use organic, non-chemical PPPs?

- o Nematodes (against leatherjackets, grubs or vine weevils)
- o Parasitoid wasps (against whiteflies)
- o Gall midges (against aphids)
- o Beetles (against mealybugs)
- o Slag traps
- o Other
- o I do not use organic PPPs

## Appendix C

# Q18. Where do you buy PPPs? (several answers possible)

- o Garden centre
- o Supermarket
- o Do-it-yourself shop
- o Internet
- o Other

#### Q19. Which factors play a role in the purchase of PPPs? (several answers possible)

- o Advice from a professional user or garden centre
- o Recommended by friends or family
- o Information on internet
- o The product seems safe
- o The product seems easy-to-use
- o I used the product once before
- o The product seems effective
- o Other

#### Q20. Is the label on the PPP clear enough?

- o Yes
- o No
- o More or less

# <u>Q21.Which type(s) of protective clothing do you wear during application, mixing and loading</u> of PPPs? (several answers possible)

- o Gloves
- o Hat/Cap
- o Safety mask
- o Boots
- o Trousers
- o Goggles
- o Long-sleeves
- o Mouth mask
- o Rubber/Latex gloves
- o Coverall
- o None
- o Other

#### Q22. Which amount of PPP do you use during an application?

- o The amount indicated on the label
- o More than indicated on the label
- o Less than indicated on the label
- o I determine the amount intuitional

#### Q23. How do you apply PPPs? (several answers possible)

- o Ready-to-use sprayer
- o Trigger sprayer
- o Watering can
- o Pressure sprayer
- o Pressurised knapsack sprayer
- o Granules (e.g. slug pellets)
- o Other

#### Q24. How do you get rid of the leftover spray liquid?

- o Throw it away in the sink
- o Spray it on the treated object
- o Save it for later use
- o Bring it to a collection point (e.g. recycling centre)
- o Other

#### Q25. What do you do with rinse water after cleaning the spraying equipment?

- o Throw it away in the sink
- o Spray it additionally onto the treated object
- o Bring it to a collection point
- o Other

#### Q26. Which of the following statements is applicable to you? (several answers possible)

- o I keep in mind the presence of remains (residues) of PPPs on home-grown foods
- o I keep in mind the presence of remains (residues) of PPPs on purchased foods
- o I keep PPPs in a locked place out of reach of children and/or pets
- o I have knowledge of the meaning of the different hazard symbols on the label of a PPP

# Appendix C

# Q27. How do you remove any remains (residues) of PPPs on home-grown fruits and vegetables? (several answers possible)

- o Not
- o Peeling
- o Cooking
- o Baking
- o Frying
- o Washing
- o Squeezing

#### Q28. Do you think, after completing this questionnaire, your PPP use will change?

- o Yes, reducing
- o Yes, increasing
- o Not changing

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