

River export of pollutants: A global modelling approach

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A painting of a river landscape. In the foreground, there are green bushes and a large tree with dark brown bark and green leaves on the right side. The river flows through the middle ground, reflecting the sky and the surrounding greenery. In the background, there is a dense forest of green trees under a blue sky with light, wispy clouds. The overall style is impressionistic with visible brushstrokes.

River export of pollutants: A global modelling approach

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Chapter 1.

Introduction

1.1 Background

Water and society

Water is essential for ecosystems on earth. It plays a key role in maintaining the climate, provides for food production and drinking water and is therefore indispensable for human life. Only 2.5% of the world's water reserve is fresh water, from which only a small part (0.3%) is directly available in rivers, lakes and other surface waters (Ligtvoet, 2018).

Fresh water availability is threatened in different ways. Firstly, there are water quantity problems: large parts of the world suffer from water scarcity due to draughts or from flooding as a result of excessive rainfall, with severe consequences for ecosystems and humans. Furthermore, there is the problem of water pollution, threatening fresh water quality in many regions. These fresh water quality issues are extending to the marine environment as a result of river export. Rivers, forming the connection between land and sea, play an important role in transporting pollution, e.g., nutrients, pesticides, plastics and other substances, from land to the oceans.

Global water issues are among the most important issues of the modern world and, therefore, water is one of the main topics of the 2030 Agenda of Sustainable Development Goals (United Nations, 2018). The Sustainable Development Goals (SDGs), formulated by the United Nations in 2015, aim to achieve a better and more sustainable future for everyone (UNEP, 2016). Each SDG has a specific objective, such as poverty, inequality, environment, human rights, health and peace. There are two SDGs on water issues, i.e., SDG 6 ('Clean water and sanitation') and SDG 14 ('Life below water'), focussing on problems concerning both water quantity and quality. The different SDGs are linked to each other, which can lead to both synergies and trade-offs among them (Alcamo, 2019). Examples of such trade-off are the increase of sewerage connection as an answer to SDG 6.2, 'Adequate and equitable sanitation and hygiene for all', that could increase the emission of pollutants into rivers, the effects of growing energy crops (SDG 7, 'Affordable and clean energy') on nutrient export to coastal seas and nitrous oxide to the atmosphere and negative effects on freshwater quality as a result of the targets for SDG 2 'Zero hunger' and SDG 7 'Affordable and clean energy'.

Globally, water quality is deteriorating as a result of a lack of adequate sanitation, causing discharge of untreated wastewater into surface waters. Other important sources of water pollution include agriculture, where the use of fertilisers and pesticides form a burden on the environment, and industrial wastewater which is not adequately treated before it is discharged into surface waters (UNEP, 2016).

Water quality modelling

Worldwide, initiatives are taken to manage water quality. Monitoring programs have been set up to measure concentrations of contaminants in surface waters and ensure the quality

of drinking water (Altenburger et al., 2015; Behmel et al., 2016; UNEP, 2016). For known contaminants, these monitoring programs deliver data that can be used to understand the risks to human health and aquatic ecosystems, and to take measures to minimise those risks. Globally, monitoring activities are not equally distributed. Because of the costs of water sampling and analysis, monitoring data from developing countries are generally scarce. This problem can partially be tackled by using new monitoring techniques, like, for example, remote sensing (UNEP, 2016). Another option to obtain information about water quality if field data are lacking is modelling. For new emerging contaminants, which generally are not included in ongoing monitoring programs, modelling provides a way to predict and map the extent of water pollution. An important advantage of modelling is that it can be used to map future developments provided sufficient data about the trends in the drivers of pollution are known. In addition, modelling can play a role preventing passing on environmental problems from one SDG to another.

To manage water pollution as a result of existing issues and those yet to come, predicting future trends of contaminants in the aquatic environment has become more and more important lately. Therefore, scenarios that describe future global change have been developed, for example the IPCC scenarios (Nakicenovic and Swart, 2000), the Millennium Ecosystem Assessment (MA) scenarios (Alcamo et al., 2005) and, more recently, the Representative Concentration pathways (RCPs, (Moss et al., 2010; van Vuuren et al., 2011)) and Shared Socioeconomic Pathways (SSPs, (O'Neill et al., 2017; O'Neill et al., 2014)). The storylines of these scenarios include predictions about socio-economic development (e.g., population growth, urbanisation), climate, hydrology, land use (e.g., agricultural and industrial development) and sanitation (e.g., sewerage). Once such scenarios have been described qualitatively, they can be interpreted for quantitative assessment of future global trends. In the last decades several modelling tools have been developed to predict current and future river transport of nutrients (e.g., WaterQual (UNEP, 2016), GloBIO (Janse et al., 2015), IMAGE-GNM (Beusen et al., 2015) and GlobalNEWS (Mayorga et al., 2010)) and organic substances (e.g., SWAT (Krysanova and Arnold, 2008; Vigerstol and Aukema, 2011), GREAT-ER (Kehrein et al., 2015), PhATE (Anderson et al., 2004) and ePiE (Oldenkamp et al., 2018)). These models calculate river transport of pollutants on a global or continental scale.

GlobalNEWS

The GlobalNEWS model (Mayorga et al., 2010) has been developed to model global river transport of nutrients to coastal seas, i.e., Nitrogen, Phosphorus, Carbon and Silica. It is a global, spatially explicit model that calculates river export in terms of basin characteristics, hydrology and human activities on land. Input data for GlobalNEWS were generated using the IMAGE model (Bouwman et al., 2009) and the Water Balance Plus Model (Fekete et al., 2010) and are usually on a scale of 0.5 x 0.5 degrees longitude by latitude. GlobalNEWS is a quasi-empirical lumped model (Kroeze et al., 2012) using only a limited number of parameters to describe all sources and processes that determine the export of substances within a river basin. Dynamic characteristics of both pollutants and river basins are only

scarcely taken into account and distribution of sources and sinks in such a model is therefore often considered homogeneous for the whole river basin. GlobalNEWS includes more than 6000 river basins.

In GlobalNEWS the four future scenarios of the Millennium Ecosystem Assessment were implemented to model nutrient export by rivers in the years 2030 and 2050 (Seitzinger et al., 2010). GlobalNEWS has been used and validated in many studies over the past few years, both on a global scale (Mayorga et al., 2010; Seitzinger et al., 2010) and on a continental and regional scale (Blaas and Kroeze, 2014; Qu and Kroeze, 2010; Sattar et al., 2014; Stokal and Kroeze, 2013; Suwarno et al., 2013; Yasin and Kroeze, 2010). GlobalNEWS has been used to develop other models, like the MARINA model (Stokal et al., 2016) for modelling transport of nutrients in Chinese rivers and the model of Siegfried et al (2017), who modelled microplastics export by European rivers.

New environmental challenges

In our changing society, different social and environmental issues compete for attention, as is reflected in the Sustainable Development Goals (United Nations, 2018). Some of these issues are directly or indirectly related to water quality, for example the energy transition (described in SDG 7, i.e., 'Affordable and clean energy'). The energy transition aims at the use of a more sustainable energy mix, that globally lowers carbon dioxide (CO₂) emissions. One of the components of such a mix could be the use of biofuels, derived from energy crops (see Box 1). Large scale growing of energy crops may change fertiliser use in agriculture and increase nutrient export by rivers. Increased fertiliser use as the result of growing energy crops not only affects the direct emissions of nutrients, but also the indirect emissions of nitrous oxide (N₂O) from aquatic systems, after leaching and runoff of nitrogen from fertilised soils (Murray et al., 2015). Having a high Global Warming Potential (GWP) ((Crutzen et al., 2008)), nitrous oxide poses a major environmental threat, that has to be included in the debate about the use of sustainable energy.

Box 1

Energy crops

The term 'Energy transition' refers to the process in which traditional fossil fuels, e.g., coal, oil and natural gas, are being replaced by low-carbon energy sources, with the ultimate goal to limit climate change as a result of energy-related CO₂ emissions (Kramer and Haigh, 2009). Promising low-carbon energy sources are solar, wind, tidal and geothermal energy, biomass and hydrogen and fuel cells (Chu and Majumdar, 2012; Hoffert et al., 2002). The direction of the energy transition varies for different energy consuming processes. It will be determined by the type of fossil fuel, preferably used in the original process, and the suitability of alternative, low-carbon solutions.

For instance, biofuels may be a good alternative for fossil fuels in the transport sector. Biofuels are derived from energy crops in different ways. We distinguish (1) first generation biofuels, derived from sources like starch, sugars and vegetable oil from arable crops, (2) second generation biofuels, derived from lignocellulosic materials like grassy or woody crops, agricultural residues or waste, and (3) third generation biofuels, derived from algae (Dornburg et al., 2010; Naik et al., 2010). Biofuels may be a promising alternative for fossil fuels in terms of CO₂ emissions, but large scale cultivation of energy crops can have undesired consequences. The main concern about biofuels, especially first generation biofuels, is the competition of energy crops with food crops. Other controversial issues are the cost and availability of biofuel crops, the impact of land use change and fresh water availability. For second and third generation biofuels, the so called 'food-versus-fuel' debate does not apply, but the processes needed for the conversion of plant or algae biomass to biofuel are rather technical, often energy intensive and expensive (Hajjari et al., 2017; Jambo et al., 2016; Naik et al., 2010). Biofuels are considered carbon-neutral because energy crops absorb CO₂ for growing. However, the energy demand of production and use of biofuels may more than counterbalance this effect, and therefore the emission of greenhouse gasses is still part of the debate. Another issue related to biofuels is the release of nutrients due to changing fertiliser use (Naik et al., 2010). When transported to coastal seas, these elevated nutrient concentrations can cause algae bloom and hypoxia, altering coastal populations which may ultimately lead to a loss of biodiversity (Howarth et al., 2011).

Other water pollution issues that are rising on the political agenda are those of contaminants of emerging concern (CECs, see Box 2) and microplastics (see Box 3). These pollutants are increasingly detected in the aquatic environment and pose potential threats to ecosystem integrity and human health. CECs and microplastics are generally not included in regular monitoring campaigns and therefore their environmental fate, behaviour and effects are fairly unknown. This makes it difficult to respond adequately to the emergence of these substances (Geissen et al., 2015).

Box 2

Contaminants of emerging concern (CECs)

New emerging pollutants constantly show up in the aquatic environment. These pollutants, as well as their metabolites and transformation products, have been classified into many different categories, e.g. pesticides, hormones, industrial chemicals, nanoparticles, pharmaceuticals and plasticisers (Dulio et al, 2018; Avio et al, 2017, Sauve and Desrosiers, 2014). Sewerage is an important source of these pollutants in the aquatic environment. Wastewater treatment removes part of the contaminants, mainly during sedimentation and biological treatment (Ahmed et al., 2017; Rodriguez-Narvaez et al., 2017). However, complete removal is difficult and therefore many contaminants are discharged as part of the wastewater treatment plant outlet. The bioavailability of emerging pollutants may vary as a result of changing environmental conditions (e.g., DOC, pH and sediment type). These changing conditions make it also difficult to predict bioaccumulation and biomagnification of emerging pollutants by modelling (Noguera-Oviedo and Aga, 2016). Degradation of emerging pollutants (e.g., biodegradation, chemical oxidation and reduction, hydrolysis and photolysis) can result in the formation of metabolites, that can be more persistent and toxic than the original substance. Furthermore, water quality standards for emerging pollutants in the environment are often lacking, which makes it difficult to regulate them (Noguera-Oviedo and Aga, 2016; Petrie et al., 2015).

Box 3

Microplastics

Plastic pollution forms a major problem in the aquatic environment. The so-called 'Plastic soup' in the oceans consists of macroplastics (e.g., plastic household items, agricultural and industrial plastics) and microplastics (e.g., plastic pellets, textile fibres and microbeads used in abrasives and cosmetics) (Cole et al., 2011). Microplastics have a typical size of 1 μm to 5 mm (Eriksen et al., 2014) and they are not only found in seas and oceans, but also in freshwater and drinking water (Koelmans et al., 2019). Microplastics used directly in, for example, personal care products are referred to as 'primary microplastics', microplastics formed by degradation and fragmentation of larger plastic items as 'secondary plastics' (Andrady, 2017; Auta et al., 2017). Microplastics are emitted into the environment by both point sources (by way of sewerage) and diffuse sources. Important sources of microplastics are fishery gear, mismanaged plastic waste, car tyre abrasion, laundry fibres, abrasives and personal care products (Lambert et al., 2014; Wagner and Lamberts, 2017). Plastics in the marine environment originate for an important part from the land, transported by streams and rivers to seas and oceans (Jambeck et al., 2015; Lebreton et al., 2017). Some of the plastics that enter the rivers via sewerage can be removed in wastewater treatment plants, especially large and buoyant plastic items and also part of the microplastics can be removed, e.g., by capturing floating pieces and by settling (Carr et al., 2016; Wagner and Lamberts, 2017). Plastic debris is found in oceans, rivers, on beaches and in organisms (Li et al., 2016). The spreading of plastics in the aquatic environment causes a number of concerns. Firstly, large plastic items may harm marine animals, like seagulls, turtles and dolphins, by entanglement or, after ingestion, by blocking the intestines (Bergmann et al., 2015; Li et al., 2016). Microplastics can be ingested by a wider range of organisms, ranging from large marine animals and fish to smaller organisms such as bivalves and zooplankton, with all kinds of physical damage as a possible consequence (Wright et al., 2013). A second area of concern relates to chemical pollutants, absorbed to the microplastics' surface, and to (micro)plastic additives, e.g., plasticisers, stabilisers, pigments and flame retardants, forming a potential hazard for the aquatic environment and the organisms living in it (Andrady, 2017; Hermabessiere et al., 2017; Koelmans et al., 2017a; Lithner et al., 2011). Once in the environment, plastics are generally quite persistent. Degradation of plastics is, although depending on its specific properties, generally very slow. Photodegradation may occur, especially on beaches, but in (sea)water, the photodegradation rate decreases dramatically as a result of lower temperatures, lower light intensity and lower oxygen levels (Andrady, 2017). Larger plastic items will gradually fragment in smaller pieces and finally, as microplastics (and nanoplastics), spread in the environment (Li et al., 2016).

1.2 Problem definition

Nowadays, new environmental problems pop up regularly and succeed each other rapidly, as a result of environmental awareness, better analysis techniques and technological progress (Munn et al., 2000; Sutherland et al., 2019; UNEP, 2012; UNEP, 2016). Due to the lack of monitoring options, it is difficult to map these problems and to develop appropriate mitigation measures. Extension of monitoring is, if possible, time-consuming and expensive. Modelling makes it possible to identify hot spots, so that monitoring can be used more effectively. Furthermore, future forecasts by a model offer the opportunity to act proactively rather than just being reactive. Modelling can thus play an important role to explore the environmental impact of new challenges like those triggered by large scale biofuel production, contaminants of emerging concern and microplastics. The effect of such new issues on the environment could be predicted by adding new scenarios, new components or new substances to existing water quality models. Furthermore, proposed solutions to environmental problems can then be tested, without implementing them first.

At the time the research described in this thesis started, the GlobalNEWS model was one of the few models used to globally predict current and future river export of nutrients to coastal areas. The model uses a limited number of parameters, is globally validated and well documented. These model characteristics make the tool a suitable candidate for expansion with new, adapted scenarios and to serve as an example for modelling other pollutants.

1.3 Research objectives

The overall objective of this thesis is to explore possibilities to expand GlobalNEWS to address the environmental impact of new water pollution challenges like those triggered by large scale biofuel production, contaminants of emerging concern and microplastics. To this end, GlobalNEWS will be adapted in the following ways: (1) by developing new scenarios, i.e. for large-scale production of energy crops, (2) by including a new environmental compartment in the model, i.e. to account for N₂O emissions to the atmosphere, and (3) by including process formulations in the model for new substances, such as triclosan and microplastics (Figure 1).

These extensions of GlobalNEWS are the subject of four case studies, that are elaborated in this thesis. The case studies aim at:

- exploring possible effects of largescale biodiesel production from energy crops on coastal eutrophication in European seas through enhanced nutrient losses from agricultural land to rivers in the year 2050;
- quantifying future N₂O emissions from European river basins that are associated with the cultivation of energy crops;
- quantifying future trends in global river export of triclosan from personal care products to coastal seas;

- contributing to a better understanding of river export of microplastics from land to sea and exploring trends in global river export of microplastics for three future scenarios (year 2050) that differ in assumed levels of environmental control.

Based on the findings from the case studies, the possibilities for expanding GlobalNEWS are evaluated.

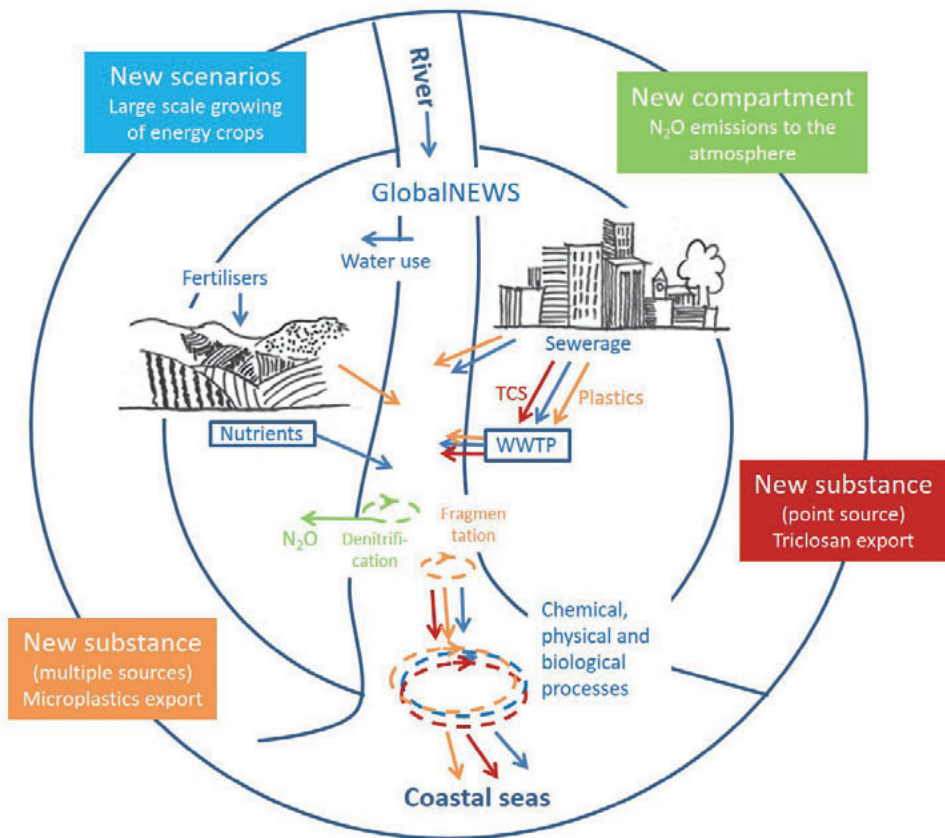


Figure 1
Overview of the scope of this thesis

1.4 Outline of the thesis

The first Chapter provides a general introduction and describes the research approach. In Chapters 2–5 the case studies are elaborated and discussed. Finally, in Chapter 6 these studies are combined to discuss the prospects for using a GlobalNEWS-like approach to model river export of different pollutants.

Brief description of the case studies

The first two case studies deal with the impacts of growing energy crops in Europe. New scenarios were developed to estimate river export of nutrients and atmospheric emissions of nitrous oxide (N₂O) as a result of large scale growing of energy crops in 2050. In these scenarios, large scale growing of energy crops and the –estimated- synthetic fertiliser use that goes with it were included. Increased fertiliser use could have consequences for coastal areas, where it can lead to eutrophication, and –indirectly- for the atmosphere. Nitrate in the aquatic environment can be converted to nitrous oxide (N₂O) by denitrification processes which is subsequently emitted into the atmosphere.

In the third case study, the GlobalNEWS model is adapted for a micro-pollutant, triclosan, developing the GlobalTCS model, that analyses global triclosan export by rivers. Used as an antibacterial agent in personal care products, triclosan is largely emitted into the aquatic environment through sewage.

In the fourth and last case study, the GREMiS model was developed. It models global river export of microplastics to coastal seas. In this model, microplastics from different sources are considered, for which the per capita input is estimated depending on economic regions as classified by the World Bank (Hoornweg and Bhada-Tata, 2012). Four different microplastics sources were considered, i.e., car tyre wear, synthetic apparel fibers, personal care products and macroplastics.

Chapter 2.

Coastal eutrophication in Europe caused by production of energy crops

Abstract

In Europe, the use of biodiesel may increase rapidly in the coming decades as a result of policies aiming to increase the use of renewable fuels. Therefore, the production of biofuels from energy crops is expected to increase as well as the use of fertilisers to grow these crops. Since fertilisers are an important cause of eutrophication, the use of biodiesel may have an effect on the water quality in rivers and coastal seas. In this study we explored the possible effects of increased biodiesel use on coastal eutrophication in European seas in the year 2050. To this end, we defined a number of illustrative scenarios in which the biodiesel production increases to about 10–30% of the current diesel use. The scenarios differ with respect to the assumptions on where the energy crops are cultivated: either on land that is currently used for agriculture, or on land used for other purposes. We analysed these scenarios with the Global NEWS (Nutrient Export from WaterSheds) model. We used an existing Millennium Ecosystem Assessment Scenario for 2050, Global Orchestration (GO2050), as a baseline. In this baseline scenario the amount of nitrogen (N) and phosphorus (P) exported by European rivers to coastal seas decreases between 2000 and 2050 as a result of environmental and agricultural policies. In our scenarios with increased biodiesel production the river export of N and P increases between 2000 and 2050, indicating that energy crop production may more than counterbalance this decrease. Largest increases in nutrient export were calculated for the Mediterranean Sea and the Black Sea. Differences in nutrient export among riverbasins are large.

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2.1. Introduction

The use of renewable energy from wind, solar and biomass is expected to increase in the future to stabilise global climate change and to enhance energy security. Energy in biomass can be converted into liquid biofuels, like bio-ethanol and biodiesel. The Global Energy Assessment (GEA, 2012) states that renewable energies are abundant, widely available and increasingly cost-effective. However, GEA also indicates that it is a major challenge to assure the sustainability of the proposed renewable technologies. Energy from biomass could play an important role in 'decarbonising' the energy supply, assuming that the carbon in this biomass is part of the 'short' carbon cycle and does not contribute to the enhancement of CO₂ levels in the atmosphere. In the GEA-study different groups of future pathways are defined. The intermediate pathways (indicated as GEA-Mix) are characterised by a mix of efficiency improvements and cleaner supply-side technologies. These GEA-Mix pathways indicate that worldwide supply of energy from biomass (biofuels and co-processing of biomass with coal or natural gas) could grow from 45 EJ in 2005 to 80-140 EJ by 2050. In the GEA-Mix pathway liquid biofuels constitute about 80% of total fuel use in the world wide transport sector in 2100.

It is easier to switch to biofuels than to (renewable) electricity for the transport sector. Biofuels do not require major adjustments of the present fossil fuel based infrastructure for energy supply to the transport sector. This makes biofuels popular alternatives to liquid fossil fuels (petrol and diesel) presently used in the transport sector. The demand for liquid biofuels can be met by either first generation (derived from sources like starch, sugar, and vegetable oil from arable crops), second generation (derived from lignocellulosic materials like grassy or woody crops, agricultural residues or waste) or third generation (derived from algae) liquid biofuels (Dornburg et al., 2010). Several studies analysed the potentials of different types of biofuels (de Wit et al., 2011; Fischer et al., 2010). Currently, mainly first generation liquid biofuels are produced in Europe. Second and third generation fuels are as yet too expensive for commercial production.

The European Union aims to increase the use of renewable energy. The European Directive 2009/28/EC of 23 April 2009 on the promotion of renewable energy (EU, 2009) aims to achieve, by 2020, a 20% share of energy from renewable sources in the EU's overall consumption of energy and a 10% share of energy from renewable sources in each member state's transport energy consumption.

Growing crops for biofuel production can have negative effects on food security: energy crops compete with food and feed crops for natural resources like arable land and water (Spiertz and Ewert, 2009). The shift in agricultural production from food or feed crops towards energy crops is likely to increase food prices and endanger food security (Baffes, 2013). In addition, the production of biofuels could give rise to negative impacts on the environment. In particular negative effects on biodiversity and carbon stocks due to direct and indirect land use change have been pointed out extensively (DiMaria and Van der Werf,

2008; Fargione et al., 2008; Lapola et al., 2010; Searchinger et al., 2008). Furthermore, Erisman et al. (2010) indicate that growing first generation biofuel crops will result in increasing N₂O emissions from fertiliser use.

To mitigate negative environmental effects of extensive biofuel production, the EU directive includes environmental sustainability criteria to ensure that growth of energy crops is sustainable and is not in conflict with overall environmental goals. The directive states that *“Where biofuels and bioliquids are made from raw material produced within the Community, they should also comply with Community environmental requirements for agriculture, including those concerning the protection of groundwater and surface water quality”*. The sustainability criteria in the directive do not specifically address adverse eutrophication effects in coastal waters due to nutrient (nitrogen (N) and phosphorus (P)) leakages induced by the cultivation of energy crops. From an environmental point of view it is important to take this cultivation into account since energy crops probably will be grown on low input agricultural land or non-agricultural land. This could lead to enhanced fertiliser use in Europe, to higher nutrient leakages to groundwater and surface waters and, as a result, higher nutrient export by rivers. Eventually this could lead to increasing eutrophication of coastal waters. Fischer et al. (2010) estimated for an energy oriented scenario, considering substantial land use conversions including the use of pasture land, that the potential for energy crops in 2030 in EU-27 (the 27 EU member states¹ until July 1st, 2013) is 45.2 million hectares, consisting of 30.5 million hectares of existing arable land and 15.2 million hectares of pasture land. Fertiliser input to these pasture lands was originally low and therefore the transformation of this area to agricultural land for energy crops with a higher fertiliser input could result in increase of EU-wide nitrogen fertiliser use by about 1.8 Tg N/y or 17.5% of the present total nitrogen-fertiliser use in EU-27 (FertilizersEurope, 2013).

The purpose of this study was to explore possible effects of large-scale biodiesel production from energy crops on coastal eutrophication in European seas, through enhanced nutrient losses from agricultural land to rivers, in the year 2050. To this end, we defined a number of illustrative scenarios in which the biodiesel production increases. We assumed only first generation energy crops in our study and used a hypothetical energy crop for our calculations, which represents a typical crop that can be grown throughout Europe. The use of a hypothetical energy crop simplified our calculations, enabling us to give a transparent and systematic analysis of nutrient export to European coastal waters using a widely accepted environmental model and scenario approach as a basis. The scenarios differ with respect to the assumptions about the area that is allocated for cultivation of energy crops: either land that is currently used for agriculture, or land that currently has a non-agricultural purpose. In our scenarios, future biodiesel production equals about 10-30% of the current fossil diesel use.

¹ The EU-27 member states until July 1st, 2013 were: Austria, Belgium, Bulgaria, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, the Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain, Sweden and the United Kingdom

2.2. Method

2.2.1 Scenario overview

We calculated nutrient export by a selection of European rivers for a number of scenarios assuming increased cultivation of first generation energy crops for the production of biodiesel. We used the Global *NEWS* models (Mayorga et al., 2010) to calculate nitrogen and phosphorus export by rivers to coastal waters. We selected river basins following Blaas and Kroeze (Blaas and Kroeze, 2014), who identified the 48 largest rivers in the 27 EU countries to study nutrient export by rivers associated in scenarios assuming large-scale cultivation of micro-algae for biodiesel on land. These EU-27 river basins were selected on the basis of their nitrogen load at the river mouth (>10 Gg/y). In our study we excluded rivers in which less than 5% of the area is used for agriculture, because these river basins are apparently less suitable for agriculture as a result of environmental conditions. As a result, 42 river basins were included in our analysis (Table 2.1).

The total area of the selected basins is 2.9 million km² while the total area of the EU-27 river basins is 4.3 million km² (data derived from the Global *NEWS* models (Mayorga et al., 2010; Seitzinger et al., 2010)). Thus the selected river basins in this study cover about two-thirds of the area of the EU-27. The total discharge of the 42 rivers included in this study is 713 km³/year. This is about 50% of the total European (EU-27) river discharge according to Global *NEWS*.

We analysed a baseline scenario and five alternative scenarios. Starting point of the scenario building was an estimate of the maximum amount of biodiesel that could be produced in 2050. To replace all current transport fuels by biofuels in the 27 EU countries 0.4 billion m³ biodiesel is needed per year (Wijffels and Bardosa, 2010). If this amount of biodiesel were to be produced from first generation energy crops like rapeseed, an area of 3 million km² would be needed to grow these crops. This is about the total basin area of the 42 rivers in our analysis, or about two thirds of the total EU27 area. Therefore, it is unrealistic to assume that biodiesel from a first generation energy crop will replace all fossil diesel. Our scenarios aim to produce a considerable amount of biofuel from first generation energy crops; our assumptions on land use change imply that biodiesel production increases to about 10-30% of the current diesel use.

In Europe rapeseed (North Western and Central Europe) and sunflower (Central and Southern Europe) are the main crops used for feedstock for biodiesel production. For our analyses, we assumed the production of a hypothetical first generation energy crop. N and P fertiliser input for first generation energy crops like rapeseed in Europe are generally in the range of 100 – 200 kg N/ha/y and 15 – 40 kg P /ha/y (Pimentel and Patzek, 2005; Ulgiati et al., 2004; van der Voort et al., 2008). We used a nitrogen input of 121 kg N /ha/y and a phosphorus input of 28 kg P /ha/y for this hypothetical crop, corresponding with the values given by de Vries et al. (2013) based on a study on biofuel cropping in Germany.

Table 2.1.

European rivers included in the study; 42 rivers from the Global NEWS model discharging into the EU-27 countries coastal waters (modified from Blaas and Kroeze, 2014) (Mayorga et al., 2010; Seitzinger et al., 2010)

River	Country where the river mouth is located	Basin area (km ²)	Perc. agricult. land (2050) ^a	Ocean/Sea
Wisla	Poland	179883	42	Baltic Sea
Odra	Germany	118731	44	Baltic Sea
Nemunas	Lithuania	95532	26	Baltic Sea
Daugava	Letland	83279	21	Baltic Sea
Narva	Estland	54374	25	Baltic Sea
Danube	Romania	785306	56	Black Sea
Po	Italy	100297	57	Mediterranean Sea
Rhone	France	98660	32	Mediterranean Sea
Ebro	Spain	81901	64	Mediterranean Sea
Loire	France	117340	77	North Atlantic Ocean
Douro	Portugal	95455	32	North Atlantic Ocean
Seine	France	72838	75	North Atlantic Ocean
Tejo	Portugal	72290	53	North Atlantic Ocean
Guadiana	Portugal	64196	56	North Atlantic Ocean
Garonne	France	57858	54	North Atlantic Ocean
Guadalquivir	Spain	53249	64	North Atlantic Ocean
Dordogne	France	25744	57	North Atlantic Ocean
Shannon	Ireland	20831	24	North Atlantic Ocean
Thames	UK	16833	9	North Atlantic Ocean
Trent	UK	16948	11	North Atlantic Ocean
Basin no. 885 ^b	UK	11876	66	North Atlantic Ocean
Adour	France	13010	17	North Atlantic Ocean
Basin no. 1090 ^b	France	10320	100	North Atlantic Ocean
Basin no. 1405 ^b	Ireland	7168	25	North Atlantic Ocean
Basin no. 1434 ^b	Ireland	6242	25	North Atlantic Ocean
Basin no. 1448 ^b	Ireland	6864	73	North Atlantic Ocean
Basin no. 1857 ^b	UK	5594	32	North Atlantic Ocean
Basin no. 1875 ^b	UK	5671	33	North Atlantic Ocean
Basin no. 1941 ^b	UK	5171	64	North Atlantic Ocean
Basin no. 1972 ^b	Ireland	4351	48	North Atlantic Ocean
Basin no. 2348 ^b	Ireland	3526	94	North Atlantic Ocean
Basin no. 4520 ^b	Ireland	1912	100	North Atlantic Ocean
Rhine	The Netherlands	163750	45	North Sea ^c
Elbe	Germany	148118	50	North Sea ^c
Gota	Sweden	44107	12	North Sea ^c
Weser	Germany	45389	30	North Sea ^c
Meuse	The Netherlands	43284	50	North Sea ^c
Humber	UK	23670	23	North Sea ^c
Scheldt	The Netherlands	20604	79	North Sea ^c
EMO	Germany	14989	25	North Sea ^c
Basin no. 1095 ^b	UK	10066	17	North Sea ^c
Basin no. 1456 ^b	UK	6264	50	North Sea ^c

^a Rounded percentages are derived from the Global NEWS models, from the GO2050 scenario (Mayorga et al., 2010)

^b In the Global NEWS models, river basins with a small basin area are referred to with a number

^c In our study river basins that flow into the North Sea form a group separated from the other rivers that flows into the North Atlantic Ocean.

We used an existing Millennium Ecosystem Assessment Scenario for 2050, Global Orchestration 2050 (GO2050), as a baseline scenario (S0) for our scenario building (Table 2.2 and Figure 2.1) ((Alcamo et al., 2005; Carpenter et al., 2005; Cork et al., 2005)). In our first four alternative scenarios (S1-S4) we assumed that energy crops will be grown on non-agricultural land to produce a reasonable amount of biodiesel without harming food and feedstock production too much. Another reason to use non-agricultural land for energy crops was the decrease, by about 10%, of total agricultural area in the GO2050 scenario (our baseline scenario) relative to the situation in the year 2000. In the baseline scenario, this land may be converted to non-agricultural land (such as urban and recreational areas) (Mayorga et al., 2010). In our first four alternative scenarios (S1-S4) we assumed that an area as large as 10% of the total area of each watershed could be used as agricultural land for growing energy crops. For the total study area this meant that 19% of the non-agricultural area will be converted to energy crops, fully compensating for the 10% of agricultural land lost between 2000 and 2050 in the MEA-GO2050 scenario. In addition ten per cent of agricultural or non-agricultural land or both is assumed to be used for energy crops in the scenarios S2-S4 (Figure 2.1). In the individual river basins land use differ strongly for all scenarios, as is showed in Figure 1 for four different European river basins (the Loire, the Gota, the Shannon and the Guadiana)

Scenario S5 assumes that 30% of the existing agricultural land of the baseline scenario will be used for growing energy crops. This estimate was based on Fisher et al. (2010) indicating that 30% of the European agricultural area could be used for energy crops without being a threat to food production (Fischer et al., 2010). The last scenario (S6) assumes that 60% of the existing agricultural land is used for growing energy crops. This scenario could provide for 30% of the current diesel demand, but is a rather extreme scenario. In many European countries using 60% of the agricultural area for energy crops means a serious threat to food production. However, in some river basins it might be possible to reallocate such a large proportion of agricultural land for cultivation of energy crops (Fischer et al., 2010).

Table 2.2.

Scenario description: assumptions about growing energy crops in the study area for six scenarios, using the GO2050 Millennium Ecosystem Assessment (MEA) scenario as a baseline.

Scenario	Description
S0	Baseline scenario, assuming no production of energy crops. This scenario is the MEA 2050 scenario Global Orchestration as implemented in Global NEWS ^a
S1	As S0, but assuming that 10% of the total area of each watershed is used for energy crops. We took this area from the non-agricultural land in S0, thus enlarging the total agricultural area.
S2	As S1, but assuming that in addition 10% of the existing agricultural land in S0 is used for energy crops.
S3	As S1, but assuming that in addition 10% of the existing non-agricultural land in S0 is used for energy crops.
S4	As S1, but assuming that in addition 10% of both the existing agricultural and 10% of the existing non-agricultural land in S0 is used for energy crops.
S5	As S0, but assuming that 30% of the existing agricultural land in S0 is used for energy crops. ^b
S6	As S0, but assuming that 60% of the existing agricultural land in S0 is used for energy crops. ^b

^a (Seitzinger et al., 2010)

^b Based on (Fischer et al., 2010)

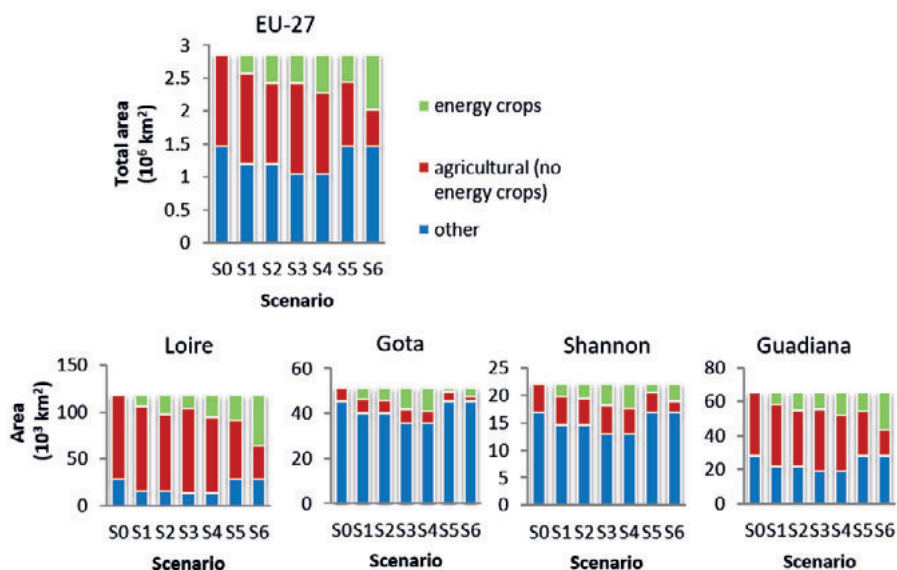


Figure 2.1.

Land use in the study region in scenarios S0-S6 (top graph; see Table 2.2 for scenario descriptions; source: Global NEWS). The bottom graphs display land use in four selected European river basins.

We ran the Global *NEWS* models to calculate river export of nutrients for our scenarios, accounting for the additional fertilisers needed to grow energy crops (i.e. we changed model input parameters for N and P from fertiliser in the Global *NEWS* models). We did not change the manure inputs to the basins, implying that animal numbers remain at their S0 levels in all scenarios. We obtained river basin area data and fertiliser application data for the conventional agricultural area from the Global *NEWS* models (GO2050 scenario). The fertiliser application for the energy crops we calculated by using the fertiliser input for our hypothetical energy crop and the area that was allocated for growing energy crops.

All our scenarios assume an increase in synthetic fertiliser (N and P) use (Figure 2.2). In Table 2.3 the model input for each scenario is summarised.

Table 2.3.

*Scenario overview: Model inputs for the selected 42 river basins (basin area and fertiliser application data for S0 are derived from the Global *NEWS* models).*

Scenario	Total area (10 ⁶ km ²)	Agricultural area excl energy crop (10 ⁶ km ²)	Additional area for energy crop (10 ⁶ km ²)	Fertiliser N use for energy crop (Tg N/y)	Total fertiliser N use (Tg N/y)	Fertiliser P use for energy crop (Tg P/y)	Total fertiliser P use (Tg P/y)
S0 ^a	2.85	1.38	0	0	9.27	0	1.50
S1 ^b	2.85	1.38	0.28	3.44	12.71	0.80	2.30
S2 ^b	2.85	1.24	0.42	5.11	13.46	1.18	2.54
S3 ^b	2.85	1.38	0.43	5.22	14.50	1.21	2.71
S4 ^b	2.85	1.24	0.57	7.00	15.24	1.62	2.95
S5 ^b	2.85	0.97	0.41	5.00	11.48	1.16	2.21
S6 ^b	2.85	0.55	0.83	10.00	13.71	2.31	2.92

^a Global *NEWS* results (Seitzinger et al., 2010)

^b This study

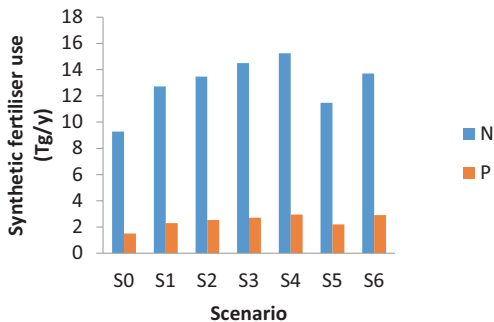


Figure 2.2.

Synthetic fertiliser use (nitrogen and phosphorus) in the study region in the scenarios S0-S6 (see Table 2.2 for scenario description, fertiliser application data for S0 are derived from the Global NEWS models).

We calculated the nitrogen and phosphorus export to European coastal waters for our six alternative scenarios. We focussed on dissolved inorganic forms of N and P since these forms are readily bioavailable and will directly contribute to eutrophication. Also organic and particulate N and P may contribute to aquatic eutrophication. However, in this study we focus on the effects of energy crops on dissolved inorganic N and P, because fertiliser is an important source of dissolved N and P in rivers.

2.2.2 Global NEWS and the Millennium Ecosystem Assessment scenarios

We analysed the scenarios using the Global NEWS (Nutrient Export from WaterSheds) models (also referred to as ‘Global NEWS’) (Mayorga et al., 2010; Seitzinger et al., 2010). These models are a set of sub models that calculate river export of nutrients as a function of human activities on the land, basin characteristics and hydrology (Bouwman et al., 2009; Fekete et al., 2010). The Global NEWS models estimate river export in more than 6000 river basins for nitrogen (N), phosphorus (P), carbon (C) and silica (Si) in different forms. Nutrient inputs to land are important drivers of N and P loads of rivers in Global NEWS (Van Drecht et al., 2009). These nutrient inputs include fertilisers and animal manure used in agriculture, but also biological N₂ fixation and atmospheric deposition. These nutrients can be transported from land to rivers as a result of leaching and runoff. In addition, point sources of nutrients in rivers, e.g. discharge from sewage systems, are included in the model. Global NEWS accounts for nutrient retention on the land and in rivers.

The Global NEWS models are spatially explicit. They use global input data at a scale of 0.5 × 0.5 degree latitude by longitude. Input databases for the Global NEWS models were generated by the IMAGE model and the Water Balance Plus model (Bouwman et al., 2009; Fekete et al., 2010; Van Drecht et al., 2009). The Global NEWS models have been used to analyse future trends in nutrient export by rivers to coastal waters worldwide. This was done by implementing the Millennium Ecosystem Assessment (MEA) scenarios (Alcamo et al.,

2005; Carpenter et al., 2005; Cork et al., 2005) in *Global NEWS*. To this end, the storylines of the MEA scenarios were interpreted to obtain input data sets for *Global NEWS* for diffuse sources of nutrients (Bouwman et al., 2009), point sources (Van Drecht et al., 2009) and hydrology (Fekete et al., 2010). The Global Orchestration (GO) scenario for the year 2050 we used in our study assumes a globalised world in terms of socio-economic aspects, and a reactive approach towards environmental problems. So it is characterised by a fast economic growth, and environmental policies solving problems only after they appear.

The *Global NEWS* models are widely accepted models for analyses at the global, continental, and regional scale including Europe (Blaas and Kroeze, 2014). The models have been validated in different earlier studies, not only at the global scale (Mayorga et al., 2010; Seitzinger et al., 2010), but also at the continental scale (Qu and Kroeze, 2010; Yasin and Kroeze, 2010) and at the regional scale (Blaas and Kroeze, 2014; Sattar et al., 2014; Strokal and Kroeze, 2013; Suwarno et al., 2013). These studies indicate that the model can be applied to analyse river export of dissolved inorganic N and P.

In the *Global NEWS* model the nutrient export at the river mouth is calculated for different nutrient forms F as follows (Mayorga et al., 2010):

$$Yld_F = (RS_{pnt,F} + RS_{dif,F}) \times FE_{riv,F} \quad (1)$$

$$RS_{dif,F} = FE_{ws,F} \times (WS_{dif_{nat},F} + WS_{dif_{ant},F}) \quad (2)$$

$$WS_{dif_{ant},N} = WS_{dif_{fe},N} + WS_{dif_{ma},N} + WS_{dif_{fix,ant},N} + WS_{dif_{dep,ant},N} - WS_{dif_{ex},N} \quad (3)$$

$$WS_{dif_{ant},P} = WS_{dif_{fe},P} + WS_{dif_{ma},P} - WS_{dif_{ex},P} \quad (4)$$

where Yld_F is de river export (in kg/km² basin area/y) and the river sources (RS) include point sources ($RS_{pnt,F}$) and diffuse sources ($RS_{dif,F}$). $FE_{riv,F}$ is the retention factor (0-1) for nutrients in the river and $FE_{ws,F}$ the retention factor (0-1) for watersheds (Mayorga et al., 2010). $RS_{dif,F}$ is calculated as a function of anthropogenic ($WS_{dif_{ant},F}$) and natural inputs of N to the land ($WS_{dif_{nat},F}$). The anthropogenic inputs of N include synthetic fertilisers ($WS_{dif_{fe},N}$), manure ($WS_{dif_{ma},N}$), natural fixation ($WS_{dif_{fix,ant},N}$), atmospheric deposition ($WS_{dif_{dep,ant},N}$) and is corrected for crop export ($WS_{dif_{ex},N}$). For P the anthropogenic inputs are similar, but do not include natural fixation nor atmospheric deposition. In this study we changed the assumed use of synthetic fertiliser following our assumptions on the production of energy crops (see Table 2.2). As a result, the fertiliser input (P and N) used as input to the model differs from the original GO scenario. We ran the *Global NEWS* model with the resulting $WS_{dif_{ant},F}$ values to calculate the nutrient export in our alternative scenarios for all rivers considered. For more details on the *Global NEWS* models we refer to Mayorga et al. (2010).

2.3 Results

2.3.1. Drivers

The alternative scenarios (S1-S6) will provide for extra biodiesel in the future. Biodiesel from the hypothetical first generation energy crop we used required about 90 g nitrogen and 20 g phosphorus per litre (Table 2.3. 'Fertiliser N use for energy crop (Tg N/y)' and 'Fertiliser P use for energy crop (Tg P/y)' and Table 2.4. 'Biodiesel (10⁶ m³)').

Table 2.4.

Scenario overview: Model output, estimated biodiesel yield and total dissolved inorganic N (DIN) and dissolved inorganic P (DIP) export of scenarios S0-S6 (see for scenario description Table 2.2)

Scenario	Biodiesel (% of current EU27 diesel use)	Biodiesel (10 ⁶ m ³)	River export of DIN (Tg N/y)	River export of DIP (Tg P/y)
S0	0	0	1.17	0.120
S1	9.3	37	1.42	0.135
S2	14	56	1.46	0.139
S3	14	57	1.56	0.143
S4	19	76	1.60	0.147
S5	14	55	1.28	0.132
S6	28	111	1.39	0.143

The export of nutrients by rivers to coastal waters was calculated to increase in the scenarios, as a result of increased use of nutrients for growing energy crops (Table 2.4). In our scenarios for 2050 (S1-S6), DIN export increases by about 20-35% compared to the baseline scenario (S0) and DIP export by about 10-20%.

River export of DIN increases in the alternative scenarios between 2000 and 2050 (Figure 2.3), by about 10-25% of the DIN export in 2000. River export of DIP hardly changed in the alternative scenarios relatively to the DIP export in 2000. However, in the baseline scenario S0, nutrient export by rivers is projected to decrease between 2000 and 2050 (DIN by about 5%, DIP by 15% of the export in 2000) as a result of agricultural and environmental policies. Thus, cultivation of energy crops in our alternative scenarios (S1-S6) counterbalances this decrease: these scenarios show an increase in nutrient export to coastal areas by increasing use of synthetic fertiliser. The relative share of fertiliser in DIN and DIP river export is higher in scenarios S1-S6 than in both the baseline scenario (S0) and the 2000 scenario (up to two or three-fold). Because the increase in total fertiliser use in the EU-27 in these scenarios is about 30-40% (Figure 2.2), the additional fertilisers are apparently used in basins with low nutrient retentions, so that the increase in nutrient export by rivers exceeds the increase in fertiliser use. This may lead to undesirable consequences if the basins drain to vulnerable coastal areas (Tysmans et al., 2012). An increase in N and P export to coastal waters is not in

line with European efforts to improve water quality by means of environmental and agricultural policies (Seitzinger et al., 2010).

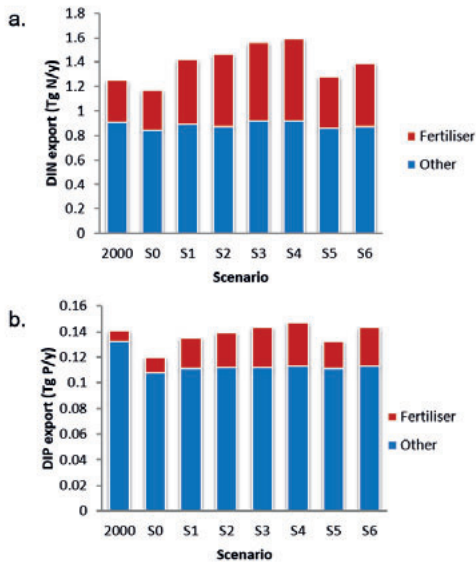


Figure 2.3.

River export of dissolved inorganic N (a) and dissolved inorganic P (b) by 42 selected EU-27 rivers to the coastal seas of Europe for six different energy crop scenarios (S0 represents the baseline scenario GO2050 and S1-S6 alternative energy crop scenarios. The first bar represents the nutrient export in 2000 (Mayorga et al., 2010)). In grey the part of the export that has its origin in synthetic fertiliser use.

The increase in agricultural area is an important driver of nutrient export by rivers. In the original MEA scenarios (e.g. S0) the total agricultural area in 2050 is about 10% smaller than in 2000. In the first alternative scenario (S1) we compensated for this loss of agricultural area by converting a considerable part of non-agricultural land to cultivate energy crops (Figure 2.1). In scenario S3 another part (10%) of the non-agricultural land from the baseline scenario (S0) is re-allocated in this way. The fertiliser use accompanying this resulted in a higher nutrient export to the coastal areas. In scenarios where agricultural area was used for first generation energy crops (scenarios S2, S4, S5 and S6) the change in nutrient use depended on the former land use and the associated fertiliser application.

The percentage of agricultural land in the GO 2050 scenario (S0) ranges from less than 30% for Scandinavian and Baltic basins to more than 60% in French basins (Figure 2.4). This percentage influences the change in nutrient use in the alternative scenarios. Converting non-agricultural land to energy crop lands (e.g. in scenario S1) will influence the nutrient use and consequently the nutrient export of basins with a small percentage of agricultural land more than basins with large percentages of agricultural land, resulting in regional differences in nutrient export.

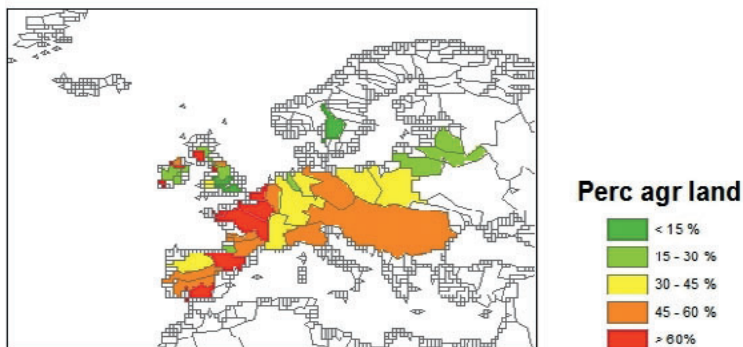


Figure 2.4.

Percentage agricultural land for the 42 river basins in the study region in the baseline scenario (GO2050) (see Table 2.1 for basin description)

Fertiliser use in the baseline scenario (S0) varies among the different river basins. In Figure 5 nutrient application (N and P) is shown in the 42 different European basins (Seitzinger et al., 2010). This figure shows that agricultural areas in Northern European regions, like Germany, Poland and the UK are more heavily fertilised than those in regions round the Mediterranean Sea.

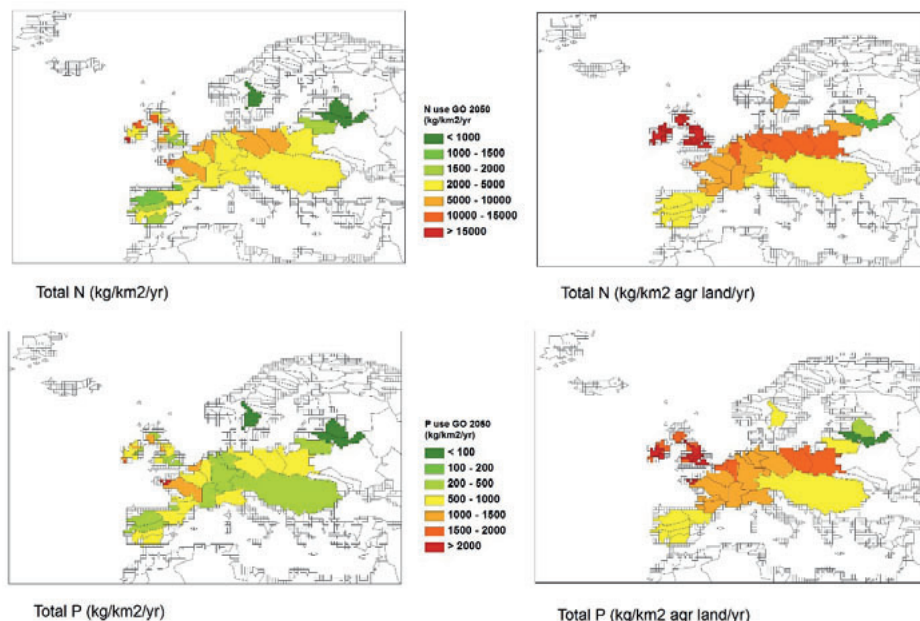


Figure 2.5.

Nutrient application (from synthetic fertiliser) in GO 2050 in the 42 selected EU-27 watersheds. In the left panel annual N and P input is displayed as kg per square kilometre ($\text{kg km}^{-2}\text{y}^{-1}$), in the right panel as kg per square kilometre agricultural land ($\text{kg km}^{-2}\text{ agr. land y}^{-1}$).

2.3.2. Nutrient export by rivers

2.3.2.1. Nitrogen export

The DIN river export to coastal waters increases in all alternative scenarios relative to S0 (Figure 2.3a). The increase is the highest if non-agricultural land is to be used for growing energy crops, as illustrated by the differences in nitrogen export between scenarios S0 and scenarios S1 (21% increase), S5 (9% increase) and S6 (19% increase). In scenarios S2, S3 and S5, which have a comparable biodiesel yield, the nitrogen export relative to S0 increased by 25% (S2), 33% (S3) and 9% (S5), indicating that nitrogen export is rather dependent on the type of land that was converted.

Nitrogen export as result of growing energy crops differed strongly among coastal regions (Figure 2.6a). The increase in nitrogen export to the Mediterranean Sea, the Baltic Sea and the Black Sea areas exceed that for other regions. For example, the increase as result of scenario S5 in these regions was up to 30 per cent in comparison with the baseline scenario, where in other coastal areas (North Atlantic and North Sea) this scenario did not result in a significant increase in nitrogen export.

2.3.2.2 Phosphorus export

River export of DIP shows a similar, but more moderate pattern as DIN export (Figure 2.6b). The total DIP-export increases for each individual scenario, as seen for nitrogen, even for scenario S5, where no additional non-agricultural land was used for energy crop (Figure 2.3b). Looking at the European coastal waters shows that increase of phosphorus export is region-dependent. Growing energy crops in the Mediterranean Sea and Black Sea watersheds affects the phosphorus export to the coastal waters the most (increases up to 15-40%) (Figure 2.6b).

2.3.2.3 Spatial patterns

Calculating the nutrient export on a regional scale showed an even more diverse picture. In Figures 2.7 and 2.8 nitrogen (DIN) and phosphorus (DIP) export is shown as percentage of nutrient export of the baseline scenario for each individual watershed. For scenarios S1-S4, where non-agricultural land is transformed into land for energy crops, we calculated an increasing N export from all the basins, but especially for those discharging in the Mediterranean Sea and the Black Sea (Figure 2.7). The increase of P export in the river basins is lower, but shows the same pattern.

To understand the spatial variability better we calculated the nitrogen *input* to selected river basins for the different scenarios (Table 2.5). We selected four river basins: the Loire, the Gota, the Shannon and the Guadiana. These basins differ strongly with regard to the percentage of agricultural land, climate and agricultural practise. We showed the differences in land use for these four basins for all the scenarios (S0-S6) in Figure 2.1.

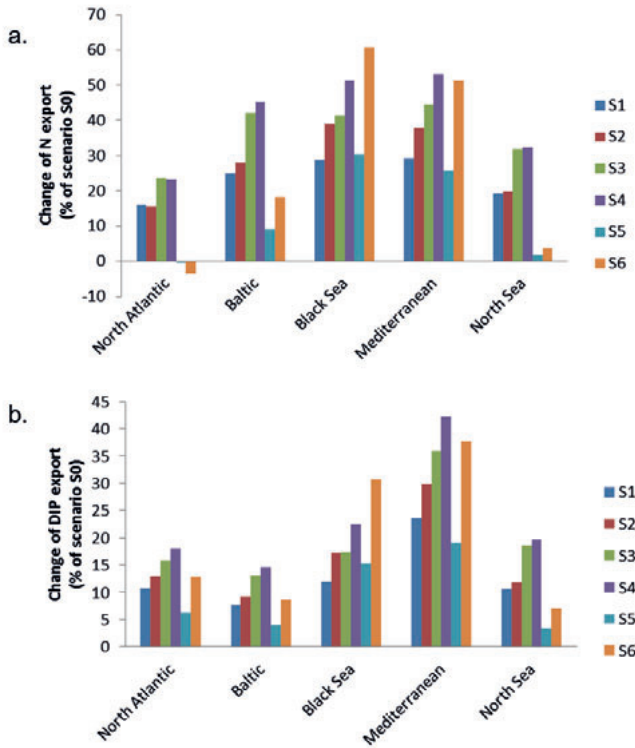


Figure 2.6.

Change of river export of dissolved inorganic N (DIN) (a) and dissolved inorganic P (DIP) (b) in scenarios S1-S6 as percentage of the baseline scenario (S0) to the different European seas (see Table 2.2 for scenario overview).

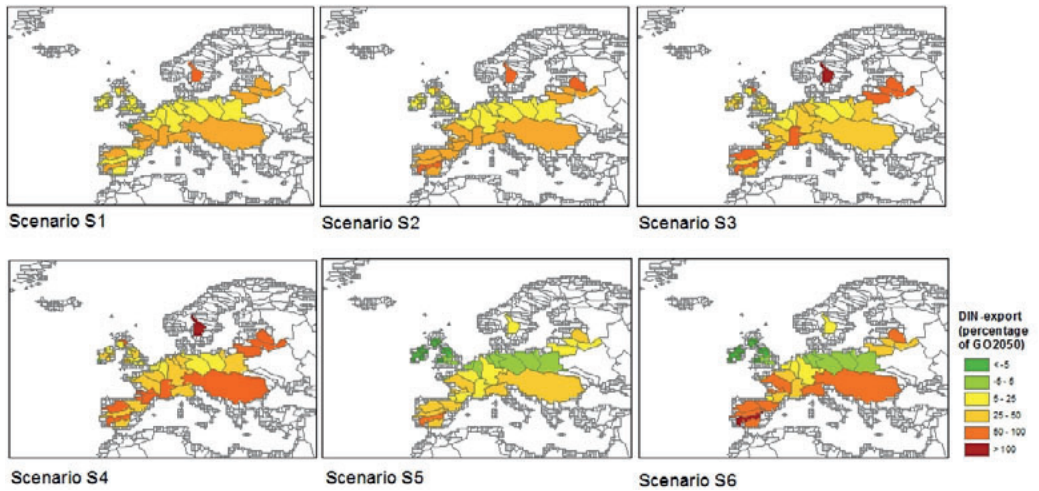


Figure 2.7.
 Difference in river export of dissolved inorganic N (DIN) between scenarios S1-S6 and the baseline scenario (as percentage of the baseline, calculated (for scenario Sx) as $\text{DIN export (Sx-S0)/S0} \times 100\%$) (see Table 2.1 for scenario overview).

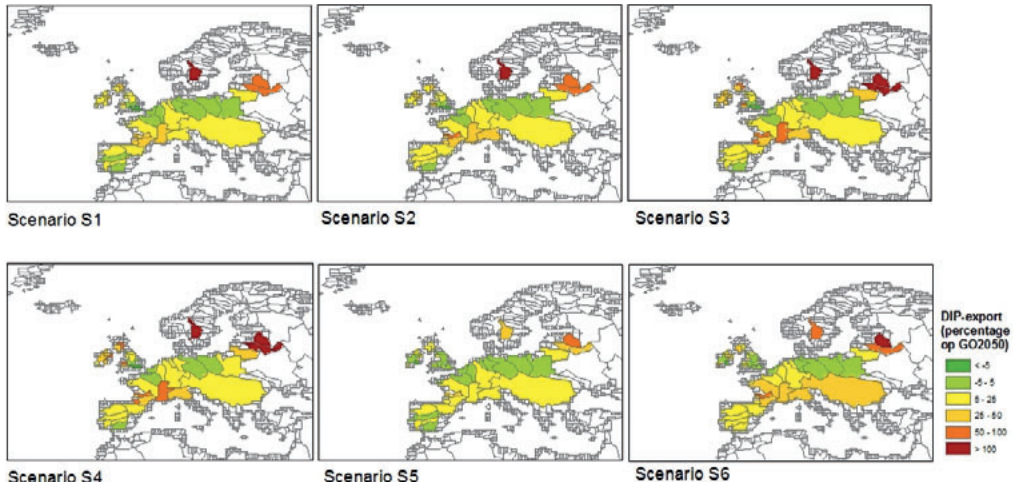


Figure 2.8.
 Difference in river export of dissolved inorganic P (DIP) between scenarios S1-S6 and the baseline scenario S0 (as percentage of the baseline, calculated (for scenario Sx) as $\text{DIP export (Sx-S0)/S0} \times 100\%$) (see Table 2.1 for scenario overview).

First we compared two river basins with different percentages of agricultural land: the French Loire basin (77%) and the Swedish Gota basin (12%). In both basins the nitrogen demand will increase if 10% of the agricultural area is used for energy crops (scenario S2). For the Loire basin this nitrogen demand exceeds that of scenario S3 where 10% of the non-agricultural land is used for energy crop, although the use of non-agricultural lands leads to a relatively larger nutrient input per square kilometre compared with scenario S0. In contrast, for the Gota basin conversion of 10% of the non-agricultural land results in a major enlargement of the agricultural land (Figure 2.1), with a subsequent need of nutrient input. This example indicates that the percentage of agricultural area within the basins strongly affects the calculated future trends in N and P in our scenarios (Figure 2.4).

Table 2.5.

Agricultural areas and synthetic N fertiliser inputs in four river basins (with different percentages of agricultural land in the baseline scenario). The agricultural area refers to the total agricultural area in the associated scenario, including area for energy crops.

Scenario	Loire (77 % agricultural land in S0)		Gota (12 % agricultural land in S0)		Shannon (24 % agricultural land in S0)		Guadiana (56 % agricultural land in S0)	
	Agricultural area (10 ³ km ²)	N-input (kg N/km ² /y)	Agricultural area (10 ³ km ²)	N-input (kg N/km ² /y)	Agricultural area (10 ³ km ²)	N-input (kg N/km ² /y)	Agricultural area (10 ³ km ²)	N-input (kg N/km ² /y)
S0	90	5333	6.2	682	5.2	4740	36.7	2062
S1	102	6543	7.5	1892	7.4	5950	43.2	3272
S2	102	6936	7.5	1969	7.4	5761	43.2	3749
S3	105	6828	12.1	2957	9.1	6874	46.0	3799
S4	105	7220	12.1	3034	9.1	6686	46.0	4276
S5	90	6511	6.2	913	5.2	4175	36.7	3493
S6	90	7688	6.2	1142	5.2	3610	36.7	4924

Also the original use of the agricultural land and the associated nutrient inputs are important. When the fertiliser use in S0 is relative low compared with that of first generation energy crops that are assumed in S1-S6, the fertiliser use will increase, and, as a result, nutrient export by rivers as well. In the UK and Ireland there is a lot of livestock breeding, therefore an important crop is grass. Although pasture land originally has a low fertiliser input (Fischer et al., 2010), grassland used for dairy and beef cattle has a relatively high nutrient demand (FertilizerManual(RB209), 2010). Replacing this crop with energy crops decreases the fertiliser inputs compared with scenario S0. For river basins like the Shannon basin a conversion of agricultural land to energy crops (like in scenarios S2, S4, S5 and S6) could result in a decreased fertiliser input. Finally, in regions like the Spanish Guadiana river basin cultivation of crops might be limited by the amount of water that might result in a moderate use of nutrients in the baseline scenario. In our alternative scenarios we assumed the same yield of the energy crops, independent of local environmental conditions. Therefore, we assumed the absence of water limitation; explicitly we assume that irrigation is applied to secure a certain average yield. Therefore, changing from the traditional crops to (irrigated) energy crops will increase the fertiliser use.

The spatial patterns of nutrient export (Figures 2.7 and 2.8) show large differences between the different regions for scenario S5 and S6, in which respectively 30% and 60% of the agricultural area is used for cultivating energy crops. For the Mediterranean and Black Sea region these scenarios imply a high nutrient export, where for the northern European regions the situation does not alter in relation to the baseline scenario. This difference may be explained by the former use of the agricultural land in these regions: in these northern European regions, the nutrient input in the baseline scenario (S0) is relatively high. This could be a result of livestock breeding in these regions (Figure 2.5).

2.3.2.4. Sensitivity analysis

We performed our calculations for a hypothetical first generation crop that we assumed to grow in all European regions. By doing this we did not analyse the best suitable energy crop for each region and, therefore, the nutrient application for this hypothetical energy crop may differ from the actual fertiliser use for the preferred crops in different basins. In Europe, rapeseed and sunflower are currently the two most commonly used biofuel crops. Both crops use synthetic fertiliser and typical input values for N and P are for both in the same wide range (de Vries et al., 2013; Pimentel and Patzek, 2005). To have some understanding of the consequence of choosing a hypothetical crop, we carried out a limited sensitivity analysis of our scenario S6 (Table 2.6). We compared the calculated river export of nutrients with those of the original S6 scenario and that of the baseline scenario (S0). The calculations show that even if the nutrient application is decreased by 25%, the DIN-export and the DIP-export to coastal waters will increase in comparison with the baseline scenario (S0) by 4% (DIN) and 10% (DIP).

Table 2.6.

River export of dissolved inorganic N and P (DIN and DIP) in scenario S6 assuming different synthetic fertilisation rates (N and P) for energy crops (ranging from 75-125% of the default fertilisation rate (121 kg N/ha and 28 kg P/ha)). The nutrient export is expressed as a percentage of that in scenario S6 and that in the baseline scenario (S0).

Applied N (kg N/ha)	Applied N (% of S6)	DIN export (% of S6)	DIN export (% of S0)
91	75	87	104
109	90	95	113
121	100	100	119
133	110	105	125
151	125	113	134

Applied P (kg P/ha)	Applied P (% of S6)	DIP export (% of S6)	DIP export (% of S0)
21	75	93	110
25	90	97	115
28	100	100	119
31	110	103	123
35	125	107	128

2.4 Discussion

We chose to keep our scenario assumptions on energy crops simple and transparent, recognising the complexity of the Global *NEWS* model and of the baseline scenario S0. However, we made a number of assumptions that may be questioned. Firstly, we used a hypothetical energy crop. We did this because the N and P fertiliser use is the main driver of water pollution, and these differ not only among crops, but also among farming systems. Our sensitivity analysis shows that fertiliser use indeed is a main driver. Even in case the fertiliser application rate is decreased by 25%, the DIN and DIP export to coastal waters increase in comparison with the baseline scenario (S0) by 4% and 10%, respectively. For the management of water quality it is therefore more important to manage fertiliser use, than crop type. This justifies our choice to model a hypothetical crop with a fixed fertiliser demand.

A second concern may be that we assumed for each river basin a similar percentage land conversion from agricultural or non-agricultural land to cultivate energy crops. Especially in basins with a large percentage of non-agricultural land it may be unrealistic to assume that ten or even twenty percent of this area will be converted to agricultural land, because this land presumably will be unsuited for agricultural practices. Moreover, we ignored that conditions for cultivating first generation energy crops differ among and within river basins. It would be interesting for future studies to further explore the possibilities within specific regions for spatial optimization of land use in order to maximise biodiesel production at lowest environmental impact. This, however, is beyond the scope of this study. We limited ourselves to generic scenario assumptions, illustrating how large scale cultivation of first generation energy crops may affect European coastal waters.

Thirdly, our scenario analysis shows that it will be difficult to supply a considerable amount of biodiesel from first generation crops in Europe, without affecting agricultural practice and the environment. Even in a rather extreme scenario (S6) in which 60% of the European agricultural area has been used for growing energy crops, the biodiesel produced could meet only 30% of the current biodiesel demand. This scenario S6 will probably have negative consequences for both food production and the environment. Such extreme scenarios may require considerable reallocation of agricultural area for growing energy crops. These scenarios will have practical implications making them not very realistic.

The MEA Global Orchestration (GO) scenario for the year 2050, that we used as a starting point of our scenario building, assumes that the nutrient use efficiency in countries with a surplus of nutrients, as in Europe, will not change in the future (Alcamo et al., 2005). The nutrient use efficiency for N (NUE) and P (PUE) reflects the ratio of fertiliser applied to fertiliser in crop yields (Bouwman et al., 2009). It is likely that the nutrient use efficiency in Europe can be improved. This could reduce nutrient losses to the environment. Our scenarios do not account for such efficiency improvements. This could also be subject of further analyses.

Elevated levels of exported nutrients (N and P) may lead to an increased eutrophication in European coastal waters. However, whether or not algal blooms and hypoxia develop is not only caused by nutrient enrichment in a particular coastal area, but largely dependent on the changes in the nutrient stoichiometry (Billen and Garnier, 2007; Howarth et al., 2011). In general, when nitrogen and phosphorus are in excess over silica the conditions are favorable for harmful algal blooms. Such conditions correspond with a positive value (>0) of the Indicator of Coastal Eutrophication Potential (ICEP). For the baseline scenario (S0) positive ICEP values for N and P are calculated, especially for the Mediterranean Sea and the Black Sea (Garnier et al., 2010). Therefore, an increase in N and P export to these, and other European seas relative to the baseline scenario is likely to cause an increased risk of negative effects of eutrophication.

The calculated increase in nutrient export by rivers in our scenarios indicates that large scale cultivation of first generation energy crops in Europe could cause an increase in eutrophication of coastal seas, in particular in Southern and Eastern Europe. The nutrient export by rivers, as calculated in our scenarios, includes export of dissolved forms of N and P (the dissolved inorganic forms, DIN and DIP and the dissolved organic forms, DON and DOP). However, fertiliser use in agriculture not only results in an increase in dissolved forms of N and P in runoff, but also in an increase in particulate P and, to a lesser extent, particulate N (Hart et al., 2004; Sobota et al., 2011). Particulate N and P are often not directly bioavailable. Nevertheless, during transportation or arrived at the coastal seas, these particulate forms could be partly transformed into more bioavailable forms and, therefore, contribute to eutrophication. In this study, particulate forms of N and P were not considered, because the Global NEWS models do not include fertiliser as a direct driver of these species in rivers. The Global NEWS particulate submodel is based on multiple regression analysis. Particulate N and P are calculated as a function of biophysical and hydrological parameters (Mayorga et al., 2010). When interpreting our results it should be noted, therefore, that the calculated increases of river export of dissolved N and P may be an underestimation of the total eutrophying potential of increased energy crop production.

Our study shows that large scale biofuel production in the EU-27 is not without dispute: our scenarios illustrate possible environmental consequences of policies aimed at increased biofuel production from energy crops produced in Europe. The scenarios differ with respect to the effects on coastal ecosystems and on the remaining agricultural area for food and feedstock production. The results indicate that it is difficult to produce considerable amounts of biodiesel without affecting nature or food and feedstock production to a large extent. The scenarios with the largest biodiesel production, S4 and S6, both have their limitations. S6 is unrealistic in that the remaining agricultural area may be too small to ensure sufficient food production in Europe, and scenario S4 results in a large increase in nutrient pollution in rivers. In the other scenarios the biodiesel yield is relatively small, meeting only 10-15% of the current diesel demand and even these modest amounts could cause considerable environmental problems. Our scenario calculations show that using non-

agricultural area for growing energy crops possibly results in increased eutrophication of rivers and coastal waters. On the other hand, reallocation of agricultural area for this purpose could harm food and feedstock production. If, as a consequence of energy crop production, food has to be imported from outside the EU-27, this could affect the overall greenhouse gas balance of biodiesel production (de Vries et al., 2010). Furthermore, nitrous oxide emissions associated with the increased fertiliser use could become considerable (Crutzen et al., 2008).

The spatial variation in the possible effects of energy crops on coastal ecosystems is large. In all our scenarios, for instance, we calculate a considerable increase in N and P export for Eastern European rivers, such as the Danube. This is because of the relatively low nutrient use in these regions in the baseline GO scenario (Figure 2.5). Even a small increase in energy crop production in such river basins could have a considerable effect on river water quality. In case energy crops will be produced in the future in such basins, it would be important to choose crops with a relatively low nutrient demand (de Wit et al., 2014).

2.5 Conclusions

Biofuels, like biodiesel, could play a major role in future energy supply. These fuels are considered as sustainable energy sources while the growth of energy crops potentially does not result in increased greenhouse emissions. However, cultivation of first generation energy crops demands the use of synthetic fertilisers causing possible leaching of nutrient to surface waters. These nutrients could be transported by rivers to coastal seas causing eutrophication problems like algal bloom and even anoxic zones (Diaz and Rosenberg, 2008; Mayorga et al., 2010). In this study we analysed six scenarios for future energy crop production, and the associated river export of nutrients to coastal waters in Europe.

Using the GO2050 Millennium Ecosystem Assessment Scenario as a baseline we found that, as a result of environmental and agricultural policies, the amount of nitrogen (N) and phosphorus (P) exported by European rivers to coastal seas will decline from 2000 to 2050 (Seitzinger et al., 2010). Our analyses show that energy crop production may more than counterbalance the decrease of nutrient export to the European coastal areas resulting from environmental and agricultural policies.

Our scenarios indicate in which basins the conversion of agricultural and non-agricultural land for cultivating first generation energy crops may lead to an increasing nutrient export to the European coastal areas. Largest effects are calculated for the Mediterranean Sea and the Black Sea. There turns out to be big differences among river basins. The scenario with the highest biodiesel yield (S6) does not show the largest nutrient export to the coastal areas. In all scenarios the increases in nutrient export more than counterbalance previously taken measures to improve water quality by 2050. Our calculations indicate that the relative share of fertiliser in these nutrient exports will be two to three times higher in our energy crop

scenarios than in the baseline scenario for 2050, indicating that additional fertiliser application especially takes place in basins with a low nutrient retention.

Large-scale cultivation of energy crops in Europe could increase the use of biodiesel in the future. We show that this cultivation could have an undesirable side-effect: eutrophication of European coastal waters. A basin-specific approach towards energy crops is needed to minimise the adverse effects and to optimise the biodiesel yield.

Chapter 3.

Future scenarios for nitrous oxide (N₂O) emissions from biodiesel production in Europe

Abstract:

Biodiesel is increasingly used as a fuel in transportation. It is generally considered an environmentally friendly alternative for diesel from fossil oil, because of lower emissions of the greenhouse gas carbon dioxide (CO₂). However, nitrous oxide (N₂O) emissions during the growth of energy crops can be considerable. N₂O is emitted as a result of fertiliser use, needed to cultivate the energy crops. Fertiliser use not only increases the direct agricultural soil emissions, but also the indirect N₂O emissions from aquatic systems, after leaching and runoff of nitrogen from fertilised soils. The aim of this study is to quantify future N₂O emissions associated with the cultivation of energy crops in European river basins. We analyse three future scenarios for biodiesel production in Europe, and the associated N₂O emissions from fertilised fields. Our focus is on biodiesel produced from first generation energy crops. The scenarios assume that by the year 2050, 15–30% of the demand for fossil diesel is replaced by biodiesel. This would change the European fertiliser needs and, as a result, N₂O emissions from fertilised soils. Our results indicate that increased biodiesel production may increase N₂O emissions in Europe by about 25–45% relative to a scenario without a growth in biodiesel production, but not equally in all regions and all scenarios. The rate of change depends on where energy crops are grown, and whether or not they replace agricultural crops, or natural vegetation.

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3.1 Introduction

Biofuels are considered sustainable energy sources because the cultivation of the energy crops potentially does not result in an increase of greenhouse gas (GHG) emissions. In particular biodiesel is considered a fuel with a low greenhouse potential (Fischer et al., 2010) and is therefore favoured in the European energy policies (EU, 2009). The amount of carbon dioxide (CO₂) that is released as a result of combustion of biofuels is compensated by the uptake of CO₂ by the crop previously and therefore the net CO₂ emissions are generally assumed to be considerable less than the CO₂ emissions of fossil fuels (Fischer et al., 2010). However, for the cultivation of the energy crops and the production of the biofuel from these crops, additional inputs are necessary, that could adversely affect the GHG balance. For growing first generation energy crops a considerable amount of synthetic fertiliser and fossil energy is needed and also the biofuel production processes require additional fossil energy. These inputs give rise to GHG emissions additional to the emissions caused by the combustion of the biofuel itself, making the use of biofuels less favourable to combat GHG emissions, as demonstrated in studies on the life cycle of biofuels from energy crops (de Wit et al., 2011; Erisman et al., 2010). A major concern is the use of synthetic nitrogen containing fertilisers (N-fertilisers), because these could be partly converted into nitrous oxide (N₂O), which has a large Global Warming Potential (GWP), about 298 times larger than CO₂ (Crutzen et al., 2008; IPCC, 2007).

Agriculture is the most important source of atmospheric N₂O (Syakila and Kroeze, 2011). In 2005 about two-thirds (4.1 Tg N₂O-N/y) of the global anthropogenic N₂O emissions were derived from agriculture, which highly exceeded the contribution of other sources like biomass burning (0.7 Tg N₂O-N/y) and industry and fossil fuel combustion (0.9 Tg N₂O-N/y) (UNEP, 2013). The increased use of synthetic nitrogen containing fertilisers leads to an increased availability of reactive N in soils and sediments, that can be converted in N₂O by microorganisms (nitrification and denitrification processes). For this conversion in N₂O there are two major pathways: direct conversion, when N in soil is microbiologically converted to N₂O and indirect conversion, when dissolved N is transported to the aquatic environment by leaching and runoff and there converted to N₂O (IPCC, 2006).

To estimate the N₂O emissions as a result of fertiliser use at the national scale, the IPCC presents Emission Factors (EFs) for calculation of direct and indirect N₂O emissions from managed soils (IPCC, 2006). Although these EFs are derived from analysis of experimental data and have been evaluated several times, they are relatively uncertain, and also questioned in studies that focus on cultivation of energy crops (Smith et al., 2012).

Emissions of N₂O are important in the discussion about biofuels. For some fuels, the N₂O emissions as a result of increased N-fertiliser use and additional CO₂ emissions during industrial fertiliser production may equal or exceed the avoided CO₂ emissions from fossil fuels (Crutzen et al., 2008; de Wit et al., 2011). Especially first generation energy crops that

demand a lot of N fertiliser, like rapeseed and corn, could therefore have a higher GWP than the fossil fuels they replace.

The aim of this study is to quantify future N₂O emissions from European river basins that are associated with the cultivation of energy crops. We consider three future scenarios in which a considerable area is used for the production of biofuels. As a starting point for our scenario analysis we used one of the Millennium Ecosystem Assessment (MA) scenarios for the year 2050, Global Orchestration (GO) (Alcamo et al., 2005). The MA scenarios were developed as part of a United Nations initiative, starting in 2000, to assess the future consequences of changing ecosystems for society. In this context, four future scenarios (including the GO scenario) were developed, which estimated the developments up to the year 2050, differing from each other on assumed socio-economic development and ecosystem management (see section 2.2 for more information). Here we take one of these scenarios as a baseline, and assume that annually 0.4 billion m³ biodiesel is needed to replace all current transport fuels in de EU-27¹ (Wijffels and Bardosa, 2010). In our scenarios we assume that a hypothetical first generation energy crop will meet 15-30% of this European (bio)diesel demand. The scenarios differ in the assumptions about the area that is converted for growing energy crops: they assume reallocation of existing agricultural land for cultivation of energy crops and/or conversion of non-agricultural land.

3.2 Method

3.2.1 Scenario overview

We calculated N₂O emissions from agricultural soils as induced by synthetic fertiliser use for a number of European river basins. We used a collection of 42 European river basins, that were selected in an earlier study (van Wijnen et al., 2015). These river basins were selected because they have a nitrogen load at the river mouth of 10 Gg/y or more (Blaas and Kroeze, 2014) and an agricultural area of at least 5%. Among them are the largest basins in the EU-27 countries¹, in terms of total area of the river basin (Figure 3.1).

To estimated amount of N fertilisers used in European river basins are taken from the Global *NEWS* models. (Mayorga et al., 2010). Global *NEWS* is a set of global models that has been used to predict future river export of nutrients in a spatially explicit way. We analysed three future scenarios assuming increased first generation energy crop cultivation. Starting point of our scenario building (S0) was a scenario that has been implemented in the Global *NEWS* models (Mayorga et al., 2010; Seitzinger et al., 2010), for the year 2050. This is one of the Millennium Ecosystem Assessment (MA) scenarios, Global Orchestration (GO). We built three scenarios using this GO2050 scenario. The scenarios differ from each other in terms of the amount of agricultural area or non-agricultural area that was attributed to the cultivation of first generation energy crops (Table 3.1, Figure 3.2).

¹ The EU-27 member states until July 1st, 2013 were: Austria, Belgium, Bulgaria, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, the Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain, Sweden and the United Kingdom.

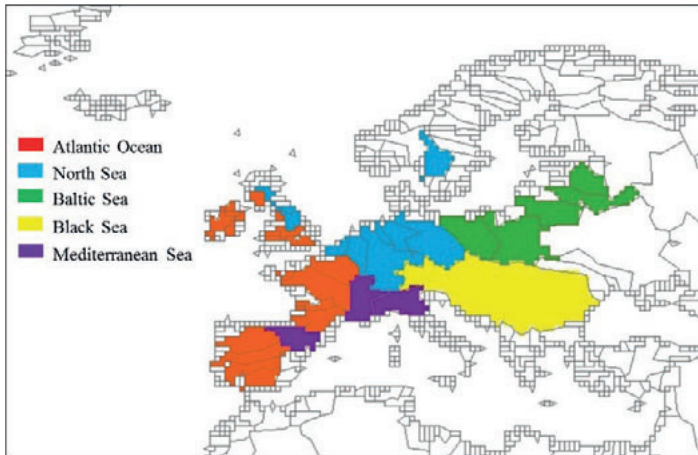


Figure 3.1.
The selected 42 river basins draining to coastal waters of the EU-27

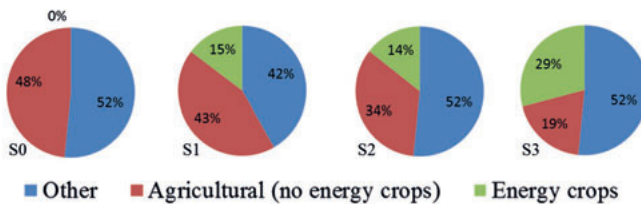


Figure 3.2.
Land use in the scenarios S0-S3 (total area: $2.9 \cdot 10^6 \text{ km}^2$, see Table 3.1 for scenario descriptions; source: Global NEWS)

We developed hypothetical scenarios. The biodiesel production in the three scenarios differed: in the first two scenarios (S1 and S2) we assumed the biodiesel production to be about 15% of the current European (bio)diesel demand (Wijffels and Bardosa, 2010) and in the third scenario (S3) about 30%. In our first alternative scenario (S1) we used non-agricultural land of the baseline scenario (S0) for growing energy crops to produce a reasonable amount of biodiesel without harming food and feedstock production too much. Another reason to use non-agricultural land for energy crops was the projected decrease, by about 10%, of total agricultural area in the GO2050 scenario (our baseline scenario) relative to the situation in the year 2000. This land is assumed to be converted to non-agricultural land (such as urban and recreational areas) (Mayorga et al., 2010). Therefore, in our first alternative scenario (S1) we assumed that in each watershed a part of the non-agricultural area as large as 10% of the total area of the watershed could be used as agricultural land for

growing energy crops. For the total study area this meant that 19% of the non-agricultural area was used to grow energy crops, fully compensating for the 10% of agricultural land that was projected to be lost between 2000 and 2050 in the MA-GO2050 scenario. In addition 10% of agricultural land was used for energy crops in this scenario (S1). In scenario S2, 30% of the agricultural area of the baseline scenario (S0) was converted for growing energy crops instead of the traditional crops for food- and feedstock. This percentage was considered the highest that can be used for cultivation of energy crops without endangering food production (Fischer et al., 2010). In scenario S3 we assumed that 60% of the agricultural area of the baseline scenario (S0) could be used for growing energy crops, providing for 30% of the current diesel demand. This scenario (S3) is on average a rather extreme scenario, because in many European countries it will severely threaten food production.

In all three scenarios (S1, S2 and S3) we assumed cultivation of a hypothetical first generation energy crop. This crop was applied in all three scenarios and in all the river basins. The input of N fertiliser for a first generation energy crop like rapeseed in Europe is generally in the range of 100-200 kg N/ha/y. We used a N input of 121 kg N/ha/y for this hypothetical crop, based on a study on biofuel (rapeseed) cropping in Germany (de Vries et al., 2013).

Table 3.1.

Scenario descriptions: assumptions about growing energy crops in the study area for three scenarios, using the GO 2050 Millennium Ecosystem Assessment (MA) scenario as a baseline (S0).

Scenario	
S0	Baseline scenario, assuming no production of energy crops and a reduction in the total agricultural area in the study region between 2000 and 2050 ¹
S1	As S0, but assuming that: <ul style="list-style-type: none"> - 10% of the total area of each watershed in S0 is used for energy crops² and in addition: - 10% of the existing agricultural land of each watershed in S0 is used for energy crops.
S2	As S0, but assuming that 30% of the existing agricultural land in S0 is used for energy crops ³
S3	As S0, but assuming that 60% of the existing agricultural land in S0 is used for energy crops ⁴

¹ This scenario is the MA 2050 scenario Global Orchestration as implemented in Global NEWS (Seitzinger et al., 2010)

² This area has been taken from the non-agricultural area in S0

³ Scenarios S1 and S2 have a comparable yield of energy crops (and therefore of biodiesel)

⁴ Based on (Fischer et al., 2010)

3.2.2 Global NEWS and de MA scenarios

We used information from the Global NEWS (Nutrient Export from WaterSheds) models to calculate N₂O emissions in our future scenarios (Mayorga et al., 2010; Seitzinger et al., 2010).

Global *NEWS* is a global, spatially explicit model that calculates river export of nutrients as a function of human activities on the land, basin characteristics and hydrology. One of the important drivers of nutrient loads of rivers in these models is the N input to land, which includes fertiliser use in agriculture. Other nutrient inputs are for example animal manure used in agriculture, biological fixation of N₂ and atmospheric deposition of nitrogen containing compounds. The different future Millennium Ecosystem Assessment (MA) scenarios were implemented in the Global *NEWS* models to analyse future nutrient export by rivers to coastal areas (Alcamo et al., 2005; Carpenter et al., 2005; Cork et al., 2005). To achieve this, after interpretation of the MA storylines, input data sets for diffuse sources, point sources and hydrology were developed for Global *NEWS*. Most Global *NEWS* model input data are at a scale of 0.5 x 0.5 degree longitude by latitude. Input databases for Global *NEWS* were generated using the IMAGE model (a global ecological-environmental model framework for simulating the consequences of human activities) and the Water Balance Plus model (a hydrological model) (Bouwman et al., 2009; Fekete et al., 2010; Van Drecht et al., 2009). In this study, we used the Global Orchestration scenario for the year 2050 (GO2050). This scenario is characterised by a fast economic growth and a reactive approach towards environmental issues. In this study we used the GO2050 scenario N-fertiliser input data in Global *NEWS*, for the selected 42 European river basins, as a starting point of our scenario building. The Global *NEWS* models have been validated in different earlier studies, at the global scale (Mayorga et al., 2010; Seitzinger et al., 2010), for specific continents (Qu and Kroeze, 2010; Yasin and Kroeze, 2010) and at the scale of selected river basins (Blaas and Kroeze, 2014; Sattar et al., 2014; Strokal and Kroeze, 2013; Suwarno et al., 2013).

3.2.3. Input of synthetic N fertiliser

We calculated the total amount of synthetic N fertiliser applied to soils in the different scenarios using the input data of the GO2050 scenario in Global *NEWS* (Bouwman et al., 2009; Seitzinger et al., 2010). In the model, synthetic fertilisers, manure, natural fixation and atmospheric deposition contribute to the total anthropogenic input of N to watersheds ($WSd_{if,ant,N}$) (Mayorga et al., 2010). We changed for our alternative scenarios the input of synthetic N fertilisers ($WSd_{if,fe,N}$), following our assumptions on the production of energy crops in the different scenarios and used these adjusted values of $WSd_{if,fe,N}$ to calculate N₂O emissions for all river basins considered.

3.2.4. N₂O emissions

We calculated N₂O emissions associated with synthetic fertiliser use following the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006). Chapter 11 of these Guidelines (N₂O Emissions from Managed Soils and CO₂ Emissions from Lime and Urea Applications) provides equations to calculate direct and indirect emissions of N₂O. Both direct and indirect N₂O emissions depend on the input of N in synthetic fertiliser.

$$N_2O \text{ emission} = N_2O(\text{direct}) + N_2O(\text{indirect}) \quad (1)$$

For direct N₂O emission as a result of fertiliser use we applied the equation:

$$N_2O(\text{direct}) = WSdif_{fe,N} \times EF1 \quad (2)$$

For indirect N₂O emissions as a result of fertiliser use we took into account N₂O from leaching and runoff using the equation:

$$N_2O(\text{indirect}) = WSdif_{fe,N} \times Fra_{CLEACH} * EF5 \quad (3)$$

Where

WSdif_{fe,N} = annual amount of synthetic fertiliser N applied to soils (kg N/ y)

Fra_{CLEACH} = fraction of all N added to mineralised soils in regions where leaching/runoff occurs that is lost through leaching and runoff (kg N/kg of N applied)

EF1 = emission factor for N₂O emissions from N inputs (kg N₂O-N/kg N input)

EF5 = emission factor for N₂O emissions from N leaching and runoff (kg N₂O-N/kg leached and runoff)

In the IPCC Guidelines Emission Factors (EFs) and fractions are proposed for both direct and indirect N₂O emissions. We use EF1 = 0.01, EF5 = 0.0075 and Fra_{CLEACH} = 0.30, leading to:

$$\begin{aligned} N_2O \text{ emission} &= N_2O(\text{direct}) + N_2O(\text{indirect}) = WSdif_{fe,N} \times (0.01 + 0.30 * 0.0075) = \\ &= WSdif_{fe,N} \times 0.01225 \end{aligned} \quad (4)$$

Using WSdif_{fe,N} in kg N/km²/y, the equation to calculate the N₂O emission results in the amount of nitrogen (N) in N₂O (N₂O-N) in kg/km²/y; to obtain the amount of N₂O (kg N₂O/km²/y) we used a correction factor of 44/28. This equation for the total N₂O emissions implies that about 18% of the total biogenic N₂O emissions associated with the use of synthetic fertilisers is the result of indirect N₂O emissions and 82% of direct N₂O emissions.

3.3 Results

We calculated the amount of N₂O emitted as a result of cultivation of energy crops in our future scenarios (S1-S3) and the increase of N₂O emissions relative to the baseline scenario (S0). In all three scenarios the total N₂O emission from synthetic fertiliser use in Europe increased by 43-86 Gg/y (24-45%) compared with the 178 Gg N₂O/y in the baseline scenario (Table 3.2).

In the baseline scenario S0, the N₂O emissions increased by 20 Gg N₂O/y (13%) between 2000 and 2050 as a result of increased fertiliser use in the GO2050 scenario. This implies that in our alternative scenarios (S1 to S3) the N₂O emissions increase by 63-106 Gg/y (40-67%) relative to 2000. In the first two alternative scenarios (S1 and S2), which have a comparable biodiesel yield, we calculated an increase of the total N₂O emissions as a result of growing energy crops. For scenario S1 the increase was about twice as high as that in scenario S2. This is not surprising because in scenario S1 non-agricultural land was converted to grow

energy crops . For scenario S3, which was designed as S2, but with a two times higher biodiesel yield, also a larger increase of N₂O emissions was calculated.

Table 3.2.

Total N₂O emissions (Gg N₂O/y) from the use of synthetic fertilisers in the baseline scenario and the biofuel scenarios (S0-S3, see for description Table 3.1) and in 2000 and the increase of N₂O emissions in the biofuel scenarios relative to S0¹ and 2000²

Scenario	Total N ₂ O emission (Gg N ₂ O/y)	Increase in total N ₂ O emission (relative to S0) ¹	Increase in total N ₂ O emission (relative to 2000) ²
2000	158	-	-
S0	178	-	13%
S1	259	45%	64%
S2	221	24%	40%
S3	264	48%	67%

¹ calculated (for scenario Sx) as $(Sx-S0)/S0 \times 100\%$

² calculated (for scenario Sx) as $(Sx-2000)/2000 \times 100\%$

Analysis of our future biofuel scenarios (S1-S3) indicated a large spatial variation of N₂O emissions among the river basins (Figure 3.3). Although we calculated an overall increase of N₂O emissions in all three scenarios, this is not necessarily true for each individual river basin (Figure 3.3). Our calculations showed that cultivation of energy crops may lead to an increase of N₂O emissions for many river basins, but that there are also some river basins where a decrease of N₂O emissions could be expected. In scenario S1, in which agricultural as well as non-agricultural land was converted to grow energy crops, N₂O emissions increase in all river basins (calculated increase ranges from 10 to 200%). In the scenarios in which only existing agricultural land was used (S2 and S3), N₂O emissions are calculated to increase in some basins (by 10 – 200%), but to decrease in others (by up to 15%). Comparing scenario S1 with scenario S2 (the two scenarios with a comparable yield of energy crop) indicates that although scenario S1 showed a larger increase of total N₂O emissions, on a regional scale both scenarios showed large increases of N₂O emissions. For example for the Danube basin, the basins in France, Spain and the Baltic States we calculated increases of N₂O emissions by 50-100% (Figure 3.3).

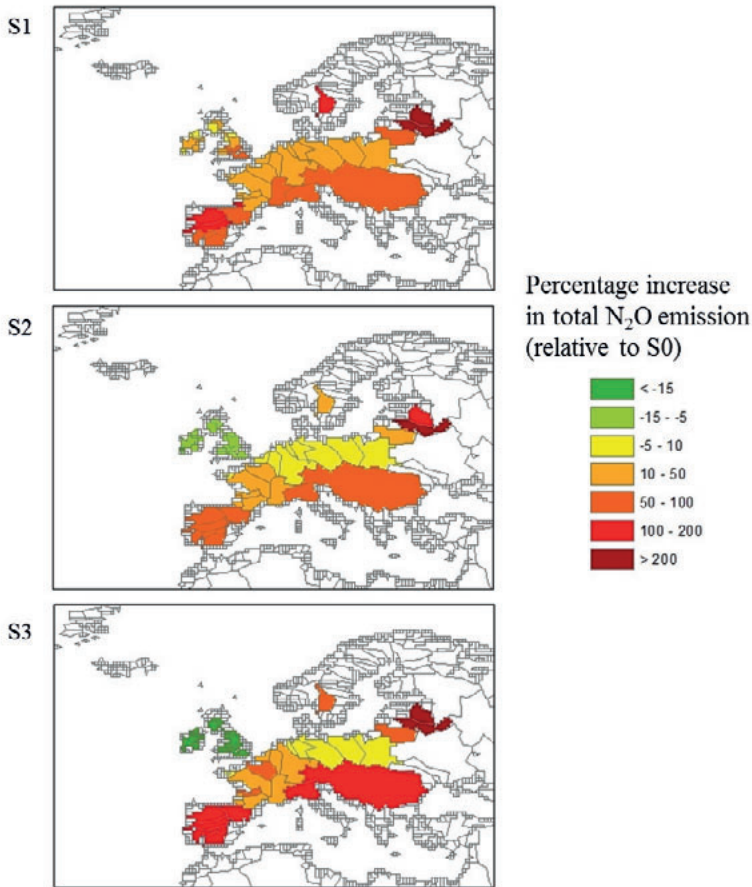


Figure 3.3.

Increase in biogenic N₂O emissions (direct and indirect) associated with the use of synthetic fertilisers in the three future scenarios S1-S3 (as percentage of the baseline scenario S₀, calculated (for scenario S_x) as $N_2O \text{ emission } ((S_x - S_0)/S_0 \times 100\%)$ (see Table 3.1 for scenario overview)

For a better understanding of the large differences in the increase of N₂O emissions as a result of cultivating energy crops for the different river basins we calculated the N₂O emissions relative to the baseline scenario in a number of large river basins (Table 3.3). We have chosen these river basins to illustrate the consequences of our different biofuel scenarios on basis of river basin dimensions and percentage of agricultural land.

Table 3.3.

The difference¹ in biogenic N₂O emissions (kg N₂O/ha/y) from selected river basins as a result of synthetic fertiliser use in energy crop cultivation in the year 2050. Differences are presented for our future scenarios (S1-S3), relative to the baseline scenario S0 (see for scenario description Table 3.1). Unit: difference with S0 in kg/ha/y (in parentheses the % change relative to S0). The last two columns show the N₂O emissions in S0 in the year 2050 in kg/ha/y.

River basin	N ₂ O emission in S1 (kg N ₂ O/ha/y)	N ₂ O emission in S1 (kg N ₂ O/ha agr area/y)	N ₂ O emission in S2 (kg N ₂ O/ha/y)	N ₂ O emission in S2 (kg N ₂ O/ha agr area/y)	N ₂ O emission in S2 (kg N ₂ O/ha agr area/y) ²	N ₂ O emission in S3 (kg N ₂ O/ha/y)	N ₂ O emission in S3 (kg N ₂ O/ha agr area/y) ²	N ₂ O emission in S3 (kg N ₂ O/ha agr area/y) ²	N ₂ O emission in S0 (kg N ₂ O/ha agr area/y)	N ₂ O emission in S0 (kg N ₂ O/ha agr area/y)
Danube	0.31 (65%)	0.48 (55%)	0.25 (50%)	0.44 (50%)	0.44 (50%)	0.49 (101%)	0.88 (101%)	0.88 (101%)	0.49	0.87
Loire	0.31 (30%)	0.36 (27%)	0.23 (22%)	0.30 (22%)	0.30 (22%)	0.45 (44%)	0.59 (44%)	0.59 (44%)	1.03	1.34
Wisla	0.24 (27%)	0.46 (22%)	0.027 (3%)	0.064 (3%)	0.064 (3%)	0.054 (6%)	0.13 (6%)	0.13 (6%)	0.89	2.12
Elbe	0.24 (23%)	0.40 (19%)	0.035 (3%)	0.070 (3%)	0.070 (3%)	0.070 (7%)	0.14 (7%)	0.14 (7%)	1.06	2.10
Rhine	0.26 (33%)	0.47 (27%)	0.076 (10%)	0.17 (10%)	0.17 (10%)	0.15 (19%)	0.34 (19%)	0.34 (19%)	0.79	1.76
Odra	0.24 (24%)	0.44 (20%)	0.013 (1%)	0.029 (1%)	0.029 (1%)	0.026 (3%)	0.058 (3%)	0.058 (3%)	0.99	2.23
Seine	0.32 (34%)	0.37 (30%)	0.25 (27%)	0.33 (27%)	0.33 (27%)	0.49 (53%)	0.66 (53%)	0.66 (53%)	0.92	1.23
Scheldt	0.27 (18%)	0.30 (16%)	0.11 (8%)	0.14 (8%)	0.14 (8%)	0.22 (15%)	0.28 (15%)	0.28 (15%)	1.47	1.86
Meuse	0.25 (26%)	0.42 (21%)	0.055 (6%)	0.11 (6%)	0.11 (6%)	0.11 (11%)	0.22 (11%)	0.22 (11%)	0.98	1.96
Guadiana	0.32 (82%)	0.49 (70%)	0.28 (69%)	0.49 (69%)	0.49 (69%)	0.55 (139%)	0.98 (139%)	0.98 (139%)	0.40	0.70
Nemanus	0.26 (91%)	0.73 (66%)	0.09 (33%)	0.37 (33%)	0.37 (33%)	0.19 (66%)	0.73 (66%)	0.73 (66%)	0.29	1.11
Shannon	0.20 (22%)	0.59 (15%)	-0.11 (-12%)	-0.46 (-12%)	-0.46 (-12%)	-0.22 (-24%)	-0.92 (-24%)	-0.92 (-24%)	0.91	3.86
Humber	0.21 (27%)	0.63 (19%)	-0.07 (-9%)	-0.32 (-9%)	-0.32 (-9%)	-0.15 (-19%)	-0.64 (-19%)	-0.64 (-19%)	0.78	3.40

¹The difference in N₂O emissions is calculated (for scenario Sx) as the emission relative to that in the baseline scenario (Sx-S0). In parentheses the difference is expressed as percentage of the N₂O emission in scenario S0 calculated for scenario Sx as (Sx-S0)/S0

² in scenarios S2 and S3 the total agricultural area of a river basin does not change comparing to the baseline scenario S0. Therefore, the percentage difference is the same expressed for the total area and the agricultural area.

In the baseline scenario S0, the N₂O emissions associated with synthetic fertiliser use range from about 1 to almost 4 kg N₂O/ha agricultural land/y by 2050 (Table 3.3). In our biofuel scenario S1 these emissions were 15-66% higher as a result of increased fertiliser use. In the other scenarios the change in N₂O emissions varied from -12% (a decrease) to 69% (S2) and from -24 to 139% (S3). We also calculated the differences between the biofuel the baseline scenario per hectare of basin. In some basins, emissions may double as a result of increased fertiliser use.

In scenario S1 the agricultural area is larger compared to that in the baseline scenario (S0), and also the ratio of agricultural to non-agricultural land has changed. Therefore, calculated in percentages, the changes in N₂O emissions per hectare of agricultural area are lower than those in N₂O emissions per hectare of the entire river basin. In this scenario all European river basins show an increase in N₂O emissions. In river basins that show relatively small N₂O emissions in the baseline scenario, scenario S1 leads to a large increase of N₂O emissions. This is illustrated with the Lithuanian Nemanus basin, that has a small agricultural area in the baseline scenario, leading to a 66% increase of N₂O emissions in scenario S1 (relative to S0). Another example is the Spanish Guadiana basin, that seems to have a very small N-fertiliser application in S0, leading to a 70% increase of N₂O emissions in scenario S1. For scenario S2, where 30% of the agricultural area of the baseline scenario is converted to energy crops, an increase in N₂O emissions has been calculated for river basins like the Danube (50%), the Loire (22%) and the Seine (27%). Other basins show a decrease of N₂O emissions (for example the Shannon and the Humber basin (12% and 9%)) or a N₂O emission that is comparable to that of the baseline scenario (for example the Odra and the Wisla basin (only an increase of 1% and 3%)). For scenario S3, where twice as much land is used for cultivation of energy crops, the same trends are calculated as in S2, with larger differences.

3.3.1 Uncertainties

We kept our scenarios simple, which enables us to give a rather quick insight in the consequences of large scale cultivation of first generation energy crops in Europe. As a consequence, we made a number of assumptions in our study. First, we used in our calculations only one hypothetical first generation energy crop, that we assumed to grow in all European regions. This seems to be rather unlikely: the two major first generation energy crops that are applied in Europe are rapeseed and sunflower, depending on the climate of a particular region. The N-fertiliser demand of both crops differ, but is for both crops often higher than the 121 kg N/ha we used for our hypothetical crop (de Vries et al., 2013; Pimentel and Patzek, 2005).

To have some understanding of the consequences of this choice, we carried out a sensitivity analysis to test the sensitivity of our calculated N₂O emissions to changes in synthetic fertiliser use. To this end, we calculated N₂O emissions assuming that the N-fertiliser demand for the energy crops is 25% smaller (90 kg N/ha) or 25% higher (150 kg N/ha) than in our default calculations (Table 3.4). Table 3.4 indicates that changing to a crop with an N-

fertiliser demand of 25% more or less may lead to a change of the total N₂O emission by about 10% (or even more).

Table 3.4.

European N₂O emissions (Gg N₂O/y) from synthetic fertilisers in our scenarios for energy crops assuming an N-fertiliser demand that is 25% lower (90 kg N/ha) or 25% higher (150 kg N/ha) than in our (default) energy scenarios (for scenario description see Table 3.1).

Scenario	Total N ₂ O emission (Gg N ₂ O/y) ¹		
	<i>N-demand energy crop -25% lower than default</i>	<i>Default N-demand</i>	<i>N-demand energy crop +25% higher than default</i>
S0	-	178	-
S1	234 (-10%)	259	283 (+9%)
S2	196 (-11%)	221	244 (+10%)
S3	215 (-19%)	264	310 (+17%)

¹ In parentheses the change of the N₂O emissions relative to the default scenarios

A second uncertainty that we should take into account is that of the emission factors we used (EF₁ and EF₅) to estimate the direct and indirect N₂O emissions from managed soils. The range of these EFs is rather large: for EF₁ the default value is 0.01 with an uncertainty range of 0.003 – 0.03 and for EF₅ the default value is 0.0075 with an uncertainty range of 0.0005 – 0.025 (IPCC, 2006). Translation of applied N-fertiliser to N₂O emissions in our scenarios with these uncertainty ranges leads to a low and a high estimate of the total N₂O emissions as a result of our energy scenarios (Table 3.5). Because the N₂O emissions are proportional to the N-fertiliser use in a scenario, the total N₂O emissions using the low EFs for each scenario were about one fourth (26%) of those using the default EFs. Using the high EFs the N₂O emissions more than tripled (306%) compared to the N₂O emissions using the default EFs.

Table 3.5.

Total N₂O emissions (Gg N₂O/y) in our scenarios for energy crops if the emission factors are low (EF₁ = 0.003 and EF₅ = 0.0005), default (EF₁ = 0.01 and EF₅ = 0.0075) or high (EF₁ = 0.03 and EF₅ = 0.025).

Scenario	Total N ₂ O emission (Gg N ₂ O/y)		
	<i>low EFs</i>	<i>default EFs</i>	<i>high EFs</i>
S0	46	178	545
S1	67	259	793
S2	57	221	737
S3	68	264	808

A third concern might be that we assumed that in all European river basins the conditions for growing energy crops are the same, which is definitely not the case. In our scenarios each river basin has the same potential to be used for cultivating energy crops and no effort has been done to specify the potentials of individual river basins, which could have resulted in a

situation in which the best suitable river basins produce more biomass for biodiesel than others.

Finally the assumption that in all river basins 10% of the non-agricultural area can be used for agriculture (energy crops) is not realistic for all river basins. For example in river basins with a small percentage of agricultural land in the baseline scenario, it could be impossible to use 10% of the non-agricultural land for energy crops. In these basins the non-agricultural area might not be suitable for agriculture practices because the climate is too cold or the landscape too hilly. It should be realised that our scenarios are hypothetical cases.

3.4 Discussion

In this study we analysed the effect of large scale biodiesel production on N₂O-emissions in Europe. We developed three future scenarios in which cultivation of first generation energy crops played an important role and we used the Global NEWS models to analyse these scenarios. Our scenarios could be able to yield an amount of biodiesel that can replace 15 to 30% of the current diesel demand for transport in Europe. All our scenarios indicate increased N₂O emissions relative to our baseline scenario, GO2050, as a result of cultivation of energy crops. In the scenarios where only already existing agricultural land was used for cultivating energy crops (S2 and S3) the calculations showed a large spatial diversity in N₂O emissions: especially for regions in Southern- or Eastern Europe we calculated relatively large increases in N₂O emissions as a result of cultivation of first generation energy crops. Unfortunately, these regions, especially in Eastern Europe, might have the best potentials to contribute to the increases European biomass based biofuel demand (de Wit et al., 2011). Because an increase of N₂O emissions might not be in line with regional policies towards GHG emission reduction, cultivating first generation energy crops in these regions could be discouraged by the government.

We focussed in our study on the N₂O emissions as a consequence of large-scale biodiesel production in Europe. Our study helps to better understand the N₂O budget. We did not intend to present a total greenhouse gas balance for energy crops or biodiesel as is typically done in life cycle assessments. We realise that the production and use of biodiesel from energy crops also results in fossil CO₂ emissions, which we did not include in our analysis. Therefore, we cannot draw conclusions about the sustainability of biofuels. Our results give us a better understanding of the amount and the spatial variation of N₂O emissions in Europe in future scenarios.

Our scenarios assume that energy crops are replacing other land use types, among which agriculture. In some basins, the implications may be that food production is replaced by energy crop production. It should be realised that this may have consequences for food production, and for import and export of food into and out of Europe. In some basins these consequences may be larger than in others, depending on the type of crops that is replaced.

A United Nations Environment Programme (UNEP, 2013) report quantifies global anthropogenic N₂O emission to be 5.3 Tg N₂O-N/y for 2010, of which 66% is from

agriculture. Europe is responsible for about 13% of the global N₂O emissions (528 Gg N₂O-N/y). In the UNEP report several future scenarios are suggested to mitigate N₂O emissions (e.g. more efficient use of N in growing crops). Analysis of these mitigation scenarios for 2050 indicates that a reduction of global N₂O emissions to 3.0 Tg N₂O-N/y might be possible. For Europe this would mean a total N₂O emission from agriculture of 390 Gg N₂O-N/y (a 26% reduction compared to 2010).

In our energy crop scenarios the total N₂O emission increase relative to the baseline scenario is 27-55 Gg N₂O-N/y (43-86 Gg N₂O/y), about 5-10% of the total European agricultural N₂O emissions (relative to 2000 this increase is 8-13%). Although the mitigation scenarios for 2050 mentioned above are likely to affect our energy crop scenarios too, our results indicate that large scale cultivation of first generation crops most probably have a substantial negative effect on (European) mitigation programs for N₂O emissions in the future.

The assumptions we made to keep our scenarios simple lead to uncertainties in the outcome of the calculation of the N₂O emissions. The uncertainty about the amount of fertiliser needed for energy crops leads to a possible deviation of the N₂O emissions of 10-19%. However, the uncertainty of the EFs determines the predictions of the N₂O-emissions as a result of the cultivation of energy crops much more. Depending on the value of the EFs, the N₂O emissions range from 26%-306% of those calculated with the default EFs (IPCC, 2006). Different authors discuss the uncertainties in these EFs (Crutzen et al., 2008; Smith et al., 2012). More than once an underestimation of EFs was reported, indicating that the N₂O emissions we calculated as a result of energy cropping might be higher than those calculated with the default EFs.

3.5 Conclusions

In our future scenarios we calculated both direct and indirect N₂O emissions as a result of fertiliser use in the cultivation of first generation energy crops. Our 2050 scenarios indicate that total N₂O emissions from the European river basins as a result of energy crop cultivation may be as large as 220 to 260 Gg N₂O/y. This is 24-45% higher than in our baseline scenario without increased energy crop production. European policies aim to reduce greenhouse gas emissions and therefore the UNEP has developed mitigation scenarios for the year 2050, with the aim to reduce N₂O emissions (UNEP, 2013). Our biofuel scenarios, however, indicate that cultivation of first generation energy crops may increase N₂O emissions relative to our baseline scenario (S0) by about 5-10% of the total European agricultural N₂O emissions in 2050, which is not in line with the 2050 mitigation scenarios of the UNEP.

Our scenarios indicate that the spatial variation in N₂O emissions is large. We calculate the highest N₂O emissions for the Southern and Eastern European region. Two of our scenarios, that have a comparable biodiesel yield (S1 and S2), show a quite different spatial pattern of N₂O emissions: in scenario S1, where both agricultural area and non-agricultural area were

converted to cultivation of energy crops, fertiliser use and therefore N₂O emissions increase all over Europe (on average by 45% relative to the baseline scenario S0). In scenario S2, in which only existing agricultural land was used the increase of fertiliser use differs for the river basins depending on the former agricultural land use. In some river basins in Northern Europe the fertiliser demand does not change or decrease as a result of cultivating energy crops, and therefore the N₂O emissions will not increase either.

European policy towards sustainable energy use is aimed at the increasing use of renewable energy sources, including biomass for biofuel production. Our study shows that although biofuels could contribute to lower greenhouse gas emissions, increased biomass production could increase N₂O emissions considerably in regions where energy crops are cultivated. This is a result of increased fertiliser use, especially in the Southern and Eastern European countries. We argue that, to minimise the N₂O emissions, energy crops with a low fertiliser demand deserve priority. Second generation energy crops, like miscanthus and willow may be more suitable to achieve this than first generation crops, because these crops demand less N-fertiliser (Rebelo de Mira and Kroeze, 2006; Smith et al., 2012). However, these crops are currently not applied on a large scale in Europe (Fischer et al., 2010).

Chapter 4.

River export of triclosan from land to sea: A global modelling approach

Abstract

Triclosan (TCS) is an antibacterial agent that is added to commonly used personal care products. Emitted to the aquatic environment in large quantities, it poses a potential threat to aquatic organisms. Triclosan enters the aquatic environment mainly through sewage effluent. We developed a global, spatially explicit model, the Global TCS model, to simulate triclosan transport by rivers to coastal areas. With this model we analysed annual, basin wide triclosan export for the year 2000 and two future scenarios for the year 2050. Our analyses for 2000 indicate that triclosan export to coastal areas in Western Europe, Southeast Asia and the East Coast of the USA is higher than in the rest of the world. For future scenarios, the Global TCS model predicts an increase in river export of triclosan in Southeast Asia and a small decrease in Europe. The number of rivers with an annual average triclosan concentration at the river mouth that exceeds a PNEC of 26.2 ng/L is projected to double between 2000 and 2050. This increase is most prominent in Southeast Asia, as a result of fast population growth, increasing urbanisation and increasing numbers of people connected to sewerage systems with poor wastewater treatment. Predicted triclosan loads correspond reasonably well with measured values. However, basin-specific predictions have considerable uncertainty due to lacking knowledge and location-specific data on the processes determining the fate of triclosan in river water, e.g. sorption, degradation and sedimentation. Additional research on the fate of triclosan in river systems is therefore recommended.

Capsule: We developed a global spatially explicit model to simulate triclosan export by rivers to coastal seas. For two future scenarios this Global TCS model projects an increase in river export of triclosan to several seas around the world.

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4.1 Introduction

Triclosan (5-chloro-2-(2,4-dichlorophenoxy)phenol; TCS) is a widely used antibacterial agent, which is added to personal care products (PCPs) like hand-soaps, toothpastes and disinfectants. It is effective against a wide range of bacteria and several moulds. Typically, sanitary products contain 0.1 – 0.3% (w/w) of triclosan. After use, triclosan can enter the environment mainly through the sewerage system (Dann and Hontela, 2011; Huang et al., 2014). In wastewater treatment plants (WWTPs), a removal efficiency of 58 – 95% can be achieved, depending on the specifications of the WWTP (Bester, 2005; von der Ohe et al., 2011). Two important processes driving the fate of triclosan in WWTPs are sorption to solids and biodegradation (Halden and Paul, 2005). Therefore, the two most important pathways for triclosan to enter the environment are WWTP effluent discharges to surface waters, and the application of the WWTP biosolids as a fertiliser on agricultural lands (Pintado-Herrera et al., 2014). As a consequence, triclosan has been found in surface waters like rivers and lakes, and in the sea (Dann and Hontela, 2011; Singer et al., 2002; Xie et al., 2008). Once in the aquatic environment, degradation of triclosan will take place by photodegradation and biodegradation, resulting in the formation of degradation products like chlorophenols and, occasionally, dioxins (such as 2,8-DCDD) (Fang et al., 2010) and methyltriclosan (MTCS) (Chen et al., 2011; EU, 2015; Tohidi and Cai, 2017).

Triclosan has been used in personal care products for over 30 years and has undergone extensive toxicological testing. It is generally considered to be safe for humans at current exposure levels (FDA, 2010), but it may trigger toxic effects in various aquatic organisms, among which algae are reported to be particularly sensitive (Orvos et al., 2002). Ecotoxicity studies on triclosan in the aquatic environment indicate that triclosan may act as an endocrine disruptor (Fang et al., 2010).

Even though the ecotoxic potential of triclosan has long been known, quantitative studies on the presence of triclosan in the environment are scarce. Worldwide, an annual per capita triclosan use ranging from 0.01 – 2 g per person has been reported (EPA-Denmark, 2016; Singer et al., 2002; von der Ohe et al., 2011). Some studies report measured triclosan concentrations in estuaries, generally in the ppt (ng/L) range (Fair et al., 2009; Pintado-Herrera et al., 2014; Zhao et al., 2010). The amount of triclosan that ends up in rivers depends on factors such as population density, sewerage connectivity (i.e., the fraction of the population that is connected to a sewage system) and the effectiveness of triclosan removal in WWTPs.

The aim of the present study is to quantify the future trends in river export of triclosan from personal care products to coastal seas of the world. To this end, we developed a 'Global TCS model': a global, spatially explicit model based on an existing river export model, i.e., the Global NEWS - Nutrient Export from WaterSheds – model (Mayorga et al., 2010; Seitzinger et al., 2010). We used the Global TCS model to calculate global triclosan export to coastal seas in the year 2000 and in two future scenarios for 2050.

4.2. Model description

4.2.1. Global NEWS and MEA scenarios

We first briefly describe the Global NEWS model because it formed the basis of our Global TCS model. Global NEWS is a global, spatially explicit model that has been used to analyse future trends in river export of Nitrogen (N), Phosphorus (P), Carbon (C) and Silica (Si). The model is described in detail elsewhere (Mayorga et al., 2010; Seitzinger et al., 2010). It calculates river export of these nutrients for more than 6000 rivers as a function of human activities on the land, basin characteristics and hydrology. The model uses global input data with a spatial resolution of 0.5 x 0.5 degree latitude by longitude, e.g., data on point sources and their socioeconomic drivers (e.g., population density and sewerage connectivity (Van Drecht et al., 2009), data on land use and diffuse sources (Bouwman et al., 2009), and data on hydrology (e.g., runoff, discharge and damming). These spatial data form an integral part of the model and were originally taken from sources such as the IMAGE model (Bouwman et al., 2006). Hydrology data were generated with the Water Balance Plus Model (Fekete et al., 2010; Vorosmarty et al., 2000).

To enable the analysis of future trends in nutrient export, the Millennium Ecosystem Assessment (MEA) scenarios (Alcamo et al., 2005; Carpenter et al., 2005; Cork et al., 2005) were implemented in Global NEWS (Seitzinger et al., 2010). Initiated by the United Nations in 2000, the MEA aimed to assess the consequences of ecosystem change for human well-being and to identify opportunities for conserving and using those systems sustainably. The MEA produced four scenarios for possible future development, each based on a different set of assumptions regarding future socio-economic developments (globalisation versus regionalisation) and approach taken towards ecosystem management (proactive versus reactive): Global Orchestration (GO), Order of Strength (OS), Adapting Mosaic (AM) and Techno Garden (TG). The GO and TG scenarios both describe a globalised world in which the GO scenario assumes a reactive approach towards environmental management and the TG scenario, focusing on technological development, a proactive approach. The OS and AM scenarios both focus on regionalisation. However, where the AM scenario assumes economic development and a positive approach towards ecosystem management, the OS scenario is protective, with a reactive approach to ecosystem management (Alcamo et al., 2005; Seitzinger et al., 2010). Concerning sewerage, the GO and TG scenarios aim at full access to improved sanitation and sewerage connection (Van Drecht et al., 2009), whereas the AM and OS scenarios assume no substantial increase of access to sanitation and sewerage connection.

Global NEWS has been validated in various studies, not only at the global scale (Mayorga et al., 2010; Seitzinger et al., 2010), but also at the continental and regional scale (Sattar et al., 2014; Stokal and Kroeze, 2013; Yasin and Kroeze, 2010). Global NEWS has also been used to simulate the dispersal of other agents, e.g. waterborne pathogens (Vermeulen et al., 2015).

Global TCS

We adapted the Global NEWS model to calculate triclosan export by rivers to coastal seas and called our model the Global TCS model. A detailed overview of the Global NEWS equations and data sets that were used in our Global TCS model is given in the Supplementary Material. Analogous to the nutrient export modelling in Global NEWS, the export of triclosan (Yld_{TCS} in g/km^2 watershed/y) is calculated with the overall equation:

$$Yld_{TCS} = RS_{pnt,TCS} \times FTCS_{riv} \quad (1)$$

where:

$RS_{pnt,TCS}$ is the triclosan input to rivers from point sources (g triclosan/ km^2 watershed/y);
 $FTCS_{riv}$ is the fraction of triclosan inputs exported by rivers and streams to coastal seas (0-1).

In this study we assumed that discharges from sewerage systems (treated or untreated) are the only significant source of triclosan in river water. Runoff from terrestrial systems is considered negligible (Aldous et al., 2012; Healy et al., 2017). We therefore consider the emission of triclosan as a point source. River export of triclosan is dependent on the population density of a particular watershed, the fraction of the population that is connected to sewerage systems and the fraction of the substance that is removed by wastewater treatment (Mayorga et al., 2010):

$$RS_{pnt,TCS} = (1-hw_{frem,TCS}) \times I \times WShw_{TCS} \quad (2)$$

where:

$hw_{frem,TCS}$ is the fraction of triclosan in sewage influent removed via wastewater treatment (0-1);

I is the fraction of population connected to sewer system (0-1);

$WShw_{TCS}$ is the input of triclosan by human waste to the watershed (g/km^2 watershed/y).

The input of triclosan ($WShw_{TCS}$) for each watershed is calculated by multiplying the triclosan use per capita ($g/cap/y$) and the population density (cap/km^2) in the watershed.

$FTCS_{riv}$ in Equation 1 represents the river retention and degradation of triclosan which is calculated as follows:

$$FTCS_{riv} = (1-L_{TCS})(1-D_{TCS})(1-FQ_{rem}) \quad (3)$$

where:

L_{TCS} is the retention fraction along the river network that counts for the loss of triclosan as a result of sorption, regular sedimentation and biodegradation processes (0-1);

D_{TCS} is the retention factor that counts for reservoir trapping and sedimentation as a result of damming (0-1);

FQ_{rem} is the consumptive water use, being the fraction of the river water that is removed for consumptive use (irrigation) (0-1).

The annual average triclosan concentration (c_{TCS}) in ng/L at the mouth of each river is calculated by:

$$c_{TCS} = Ld_{TCS} \times 10^3 / Q_{act} \quad (4)$$

where:

Ld_{TCS} is the total triclosan load per year in a basin ($Ld_{TCS} = Yld_{TCS} \times 10^{-6} \times$ the basin area A (km^2)) in ton/y;

Q_{act} is the annual river discharge under actual, anthropogenic conditions (km^3/y).

4.3 Model input

Global NEWS data and MEA scenarios

For spatially explicit input parameters such as population density, sewer connectivity, hydrology, consumptive water use, damming and WWTP treatment efficiency, we used the gridded scale inputs (with a spatial resolution of $0.5^\circ \times 0.5^\circ$) as implemented in the Global NEWS model (Table S4.2 in the Supplementary Material). For example, we used the annual river discharge (Q_{act}) for each basin from Global NEWS - which is corrected for water withdrawal schemes like large-scale irrigation (Mayorga et al., 2010) - to calculate the annual average triclosan concentration at the mouth of a river. We analysed triclosan export to coastal seas in the year 2000 and in two scenarios for the year 2050. For the future scenarios, we used the data from the GO and the AM scenarios as implemented in Global NEWS (GO2050 and AM2050).

Figure 4.1 shows the data on population density for the year 2000 and for the GO2050 and AM2050 scenarios as available in the Global NEWS model. In 2000, the population density is highest in Southeast Asia and, to a lesser extent, in Europe. In the future scenarios (GO2050 and AM2050), a population growth is predicted for Southeast Asia (Figure 4.1a). In Europe (Figure 4.1b) no large changes in population density are expected.

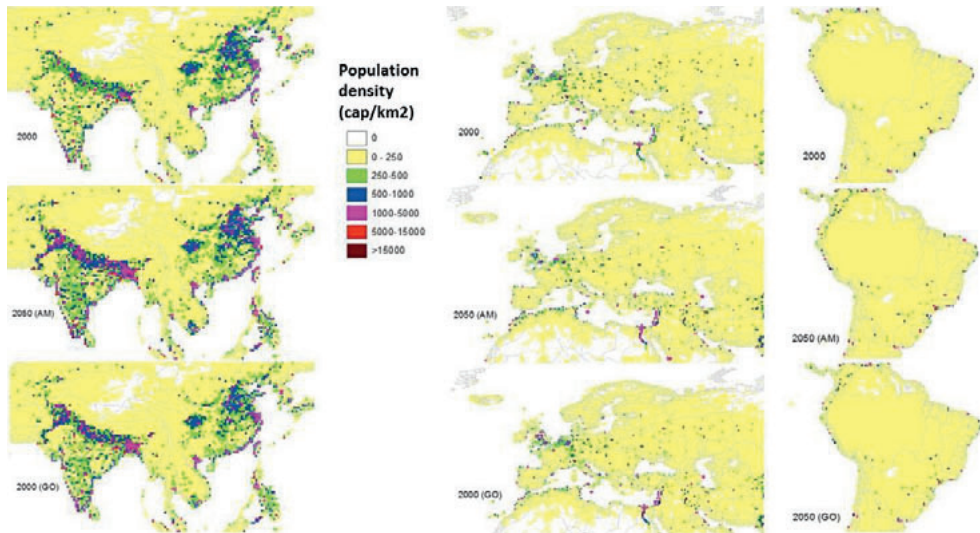


Figure 4.1.

Population density in South-East Asia (a) and Europe (b) in the 2000 scenario and the future scenarios for 2050 (GO2050 and AM 2050) (data derived from Global NEWS (Seitzinger et al., 2010)).

We estimated riverine discharge of triclosan based on the WWTP connectivity of the river basins' population, adopting the approach of Van Drecht et al. (2009) who estimated the total inputs of wastewater from sewerage systems into rivers, including river deltas. Figure 4.2 shows the data on sewerage connectivity as available in the Global NEWS model. In the year 2000, the majority of the population (60-90%) in Europe and the eastern part of the USA was connected to sewerage systems. In Southeast Asia and Africa, sewerage connectivity was low, only up to 25%, especially in rural areas. In the future scenarios for 2050, the development of sewerage connectivity is strongly associated with the scenario. For example, in the GO2050 scenario, the number of people connected to sewerage systems will increase considerably, especially in Europe, the USA and South America. In Southeast Asia, sewerage connectivity will increase too (Figure 4.2), but mainly in urbanised areas. In the AM2050 scenario, only a small increase in the number of people connected to sewerage systems is foreseen.

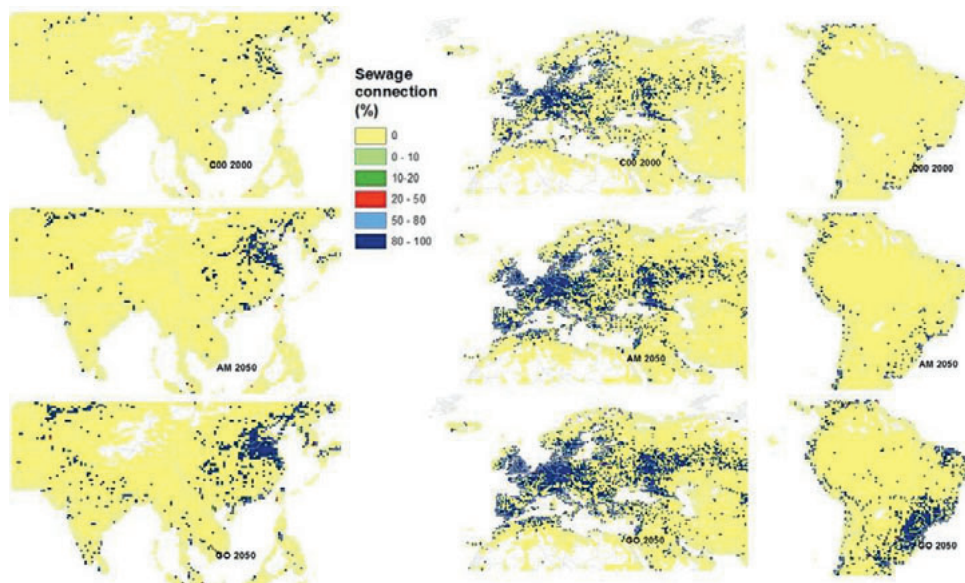


Figure 4.2. Percentage sewerage connectivity in Southeast Asia (a), Europe (b) and South America (c) in the 2000 scenario and the future scenarios for 2050 (GO2050 and AM2050) (data derived from Global NEWS (Seitzinger et al., 2010))

4.3.2. Triclosan emission

The triclosan input from personal care products in river basins ($W_{Shw_{TCS}}$ in $g/km^2/y$) was estimated using the per capita triclosan use reported in several studies. Per capita use in Europe is comparable to that in Asia and Australia (Table 4.1). Based on these data, we assumed a per capita triclosan use of 500 mg per year for all world regions.

4.3.3. Removal of triclosan at WWTPs

The fraction of triclosan removed by wastewater treatment ($hw_{rem,TCS}$) reported in literature varies. Removal rates up to 98% are reported, but also of 50-75% (Dann and Hontela, 2011). In WWTPs, triclosan can be removed from the water phase during primary treatment (wastewater settling) (Thompson et al., 2005) and secondary (biological) treatment, either by biodegradation or by sorption to sludge (Butler et al., 2012). The fraction of triclosan removed largely depends on the removal technique used, achieving higher removal rates if the residence time in the WWTP is longer (Heidler and Halden, 2007; Thompson et al., 2005). To achieve very effective removal of triclosan, up to 95% and more, more advanced removal techniques are required, such as ozonisation (Dann and Hontela, 2011; von der Ohe et al., 2011). There are also sewage collection systems discharging wastewater without or almost without treatment, especially in developing countries (WHO, 2013).

We estimated triclosan removal in river basins using the data on phosphate removal as available in the Global NEWS model as a starting point. In Global NEWS average phosphate removal data are available for more than 6000 rivers worldwide. Phosphate is removed in WWTPs through biological treatment (Van Drecht et al., 2009). We assumed that in basins with poor average phosphate removal (<20%), the dominant way to discharge sewage into the river is without any treatment and therefore, we assumed that triclosan is not being removed at all (0%). In basins with an average phosphate removal of 20-80% we assumed an average triclosan removal of 60%, mainly due to settling during primary and secondary treatment. If the phosphate removal in a basin is more than 80%, the majority of the WWTPs will perform extensive biological treatment and therefore we assumed a triclosan removal of 90% in these basins (Table S3 in the Supplementary Material).

Table 4.1.

Production of triclosan (ton/y) and estimated triclosan use (g/cap/y).

Continent/Land	TCS production (ton/y)	year	TCS use (g/cap/y)	reference
World	1500	2002	0.21	Singer et al. (2002)
World	6581	2011	0.88	EPA-Denmark(2016)
World	4760	2015	0.64	EPA-Denmark (2016)
Europe	350	2002	0.42	Singer et al. (2002)
Europe	450	2006	0.61	SCCS (2010)
Europe	1136	2011	1.7	EPA-Denmark (2016)
Europe	850	2015	1.1	EPA-Denmark (2016)
Sweden	2.3 ^a	2002	0.25	Dann and Hontela (2011)
Germany	-	2011	0.01 - 2	von der Ohe et al. (2011)
UK	-	2003	1.3	Sabaliunas et al. (2003)
US	300	2003	1.03	Halden and Paull (2005)
Australia	21	2005	1.1	NICNAS (2009)
Australia	12.5 ^a	2005	0.66	NICNAS (2009)
China	-	2013	0.50 ^b	Zhao et al. (2013)
China	-	2012	1.3 ^c	Gouin et al. (2012)
China	2787	2011	1.6	EPA-Denmark (2016)
China	1988	2015	1.2	EPA-Denmark (2016)
India	1651	2011	1.0	EPA-Denmark (2016)
India	1241	2011	0.7	EPA-Denmark (2016)

^a Triclosan production for PCPs only

^b Combined Triclosan/Triclocarban use from PCPs

^c Calculated from emission estimate

4.3.4. River retention and degradation of triclosan

We estimated a river retention and degradation factor ($FTCS_{riv}$) for each river basin by assessing the factors L_{TCS} , D_{TCS} and FQ_{rem} (Equation 3). For consumptive water use (FQ_{rem}), we used the basin-specific values available in the Global NEWS model for the year 2000 and

the two 2050 scenarios (GO and AM). The retention fraction L_{TCS} is counting for the loss of triclosan as a result of various sorption, sedimentation and degradation processes along the river. We followed the approach of Price and co-workers (2010) and used a non-process-specific overall loss rate to estimate the retention fraction (L_{TCS}), assuming that overall-loss follows first-order kinetics and estimating triclosan residence times in each basin:

$$L_{TCS} = 1 - e^{-k \times t_{res,TCS}} \quad (5)$$

where:

k is the overall loss rate coefficient for net-sedimentation and degradation;

$t_{res,TCS}$ is the average residence time of triclosan.

In our model, we used an overall loss rate of 0.06 h^{-1} ($2 \times 10^{-5} \text{ s}^{-1}$) as reported by Morrall et al. (2004) in a field study on loss rates of triclosan in a small UK river. We estimated the residence time for triclosan based on the mean residence time for channel water for the world's 50 largest rivers, reported to be in the order of 60 days (Vorosmarty and Sahagian, 2000). This value was corrected based on the size of the watershed (i.e., smaller basins have smaller residence times), and to account for the fact that the major part of the triclosan discharges take place in downstream areas because these are typically more densely populated. To correct for the size of the watershed, we took a proportionate share of the mean residence time for the world's largest rivers:

$$t_{res,w} = \text{Area}_{land} / \text{Area}_{avg} \times 60 \text{ days; with a maximum of 60 days} \quad (6)$$

where:

$t_{res,w}$ is the estimated mean residence time for water;

Area_{avg} is the mean land area of the 50 largest rivers from Global NEWS;

Area_{land} is the land area of a river basin.

We subsequently estimated the residence time of triclosan ($t_{res,TCS}$) based on the assumption that 60% of the of the point sources of triclosan, depending on the population density, are within 100 km from the coast (Hinrichsen, 1998) (see also Figure 4.1). This corresponds to a surface of approximately 5000 km^2 . The residence time of triclosan is then estimated as follows:

$$t_{res,TCS} = (0.4 + 0.6 \times 5000 / \text{Area}_{land}) \times t_{res,w} \text{ days} \quad (7)$$

where:

Area_{land} is the land area of a river basin.

For basins with $\text{Area}_{land} < 5000$ we took: $t_{res,TCS} = t_{res,w}$.

The resulting L_{TCS} value ranged from 0 to 0.9 (dimensionless).

The reservoir retention factor (D_{TCS}) represents the retention of triclosan as a result of damming. Behind dams the residence time of the water increases and therefore processes that favour triclosan sedimentation might take place. In surface waters, triclosan transport mainly takes place in the dissolved phase, but a small part is transported adsorbed to suspended sediments (average about 15%; (Gautam et al., 2014)). Behind dams, a reduced water flow can cause settling of particulates, with the adsorbed triclosan. We assumed the reservoir retention of triclosan (D_{TCS}) to be proportional to the reservoir retention factor used for total suspended sediments (D_{TSS}) for each river basin in Global NEWS (Mayorga et al., 2010) and estimated D_{TCS} by taking 15% of D_{TSS} for each basin.

4.4 Model results

4.4.1. Triclosan export to coastal areas

Figure 4.3 shows the calculated triclosan export to coastal areas for the year 2000 and for the two future scenarios (GO2050 and AM2050) in the year 2050; all based on the assumption of a worldwide triclosan use of 500 mg/cap/y. In 2000, a relatively large export was calculated for Europe, corresponding to a relatively high population density and a high sewerage connectivity. Significant triclosan export was also calculated for several Southeast Asian watersheds. In these river basins, like that of the Ganges (India) and several Chinese rivers, the percentage of the population that is connected to a sewerage system is small, especially in rural areas (according to the data from Global NEWS (Mayorga et al., 2010)). In these cases, triclosan export is mainly driven by the vast urban population, that is, at least partly, connected to the sewerage system (see also Figures 4.1 and 4.2).

In the future scenarios (AM2050 and GO2050), our Global TCS model predicts an increase in triclosan export for watersheds in Southeast Asia. In these regions, population will grow and there will be a larger fraction of the population connected to sewerage systems, especially in urban areas. Also in other urbanised areas, for example in the USA and South America, triclosan export is calculated to increase in 2050.

To better understand the changes in triclosan export, Table 4.2 summarises the results of four selected river basins, i.e. two European (the Danube and the Elbe) and two Asian (the Zhujiang and the Ganges); each with different basin characteristics. The annual average triclosan concentration in the coastal seas of these four river basins is expected to range from 1.0 to 13.6 ng/L (i.e. ppt range) on the basis of our Global TCS calculations.

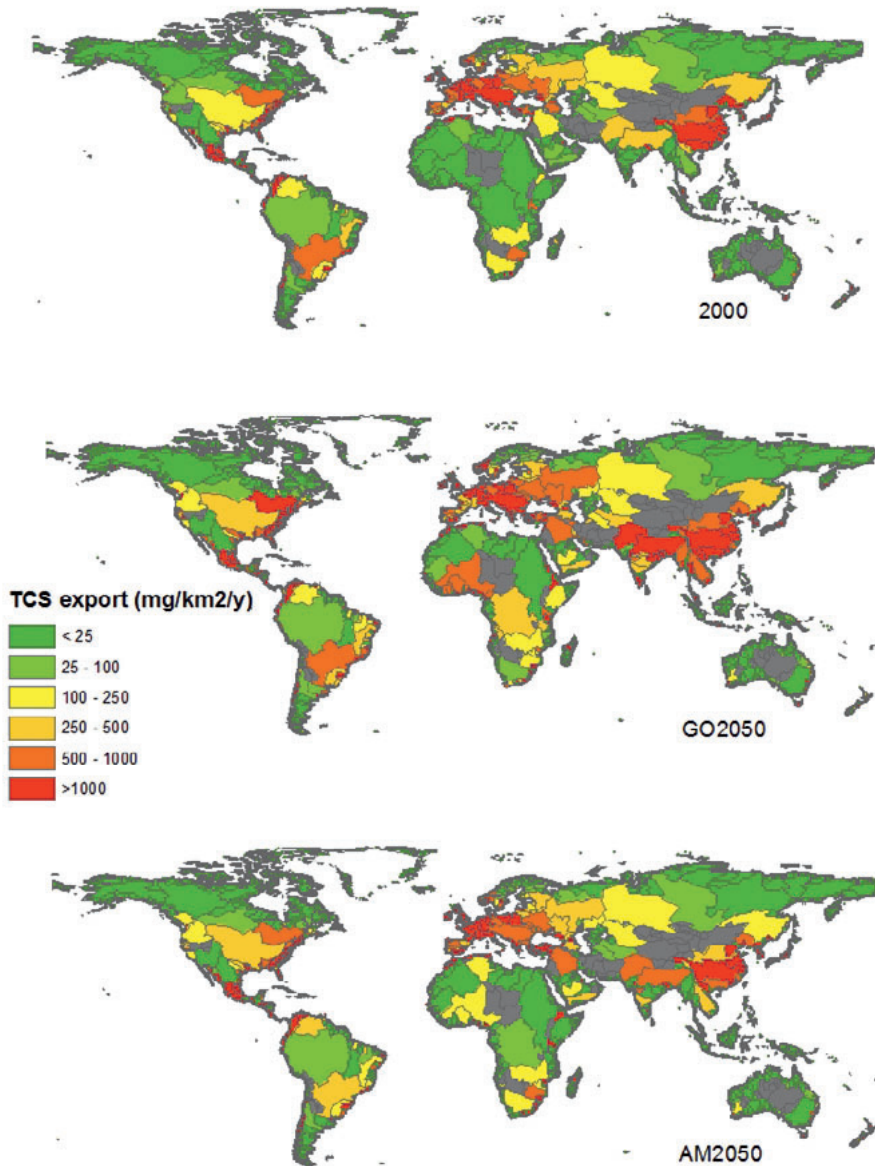


Figure 4.3.

Triclosan export to coastal areas in the year 2000 and the future scenarios for 2050 (GO2050 and AM2050), as a result of a worldwide triclosan use of 500 mg/cap/y. Endorheic basins (except those of the Caspian Sea and Lake Aral) are excluded (grey).

Table 4.2.

Calculated triclosan export in two European river basins (the Danube and the Elbe) and two Asian river basins (the Zhujiang and the Ganges), assuming a triclosan use of 500 mg/cap/y (population density, sewerage connection and basin discharge from Global NEWS (Seitzinger et al., 2010))

	Danube			Elbe			Zhujiang			Ganges		
	2000	GO2050	AM2050	2000	GO2050	AM2050	2000	GO2050	AM2050	2000	GO2050	AM2050
Population density (cap/km ²)	100	94	77	167	175	136	207	209	239	314	438	561
Sewerage connectivity (%)	58	73	68	88	96	96	18	32	23	4	24	8
TCS removal by WWTPs (fraction)	0.6	0.6	0.6	0.9	0.9	0.9	0	0.6	0.6	0	0.6	0.6
TCS input from point sources (g/km ² /y)	11.7	13.6	10.4	7.3	8.4	6.5	18.5	13.6	10.8	6.0	21.2	9.1
Basin discharge (km ³ /y)	203	157	162	17	9	10	142	174	167	703	594	593
TCS export (g/km ² /y)	1.1	1.2	0.95	0.71	0.82	0.63	1.4	1.0	0.8	0.41	1.5	0.63
TCS export (ton/y)	0.84	1.0	0.75	0.11	0.12	0.09	0.56	0.41	0.33	0.67	2.4	1.0
TCS concentration (ng/L)	4.2	6.1	4.6	6.4	13.6	9.7	3.9	2.4	1.9	1.0	4.1	1.7

In the Danube basin, there is an increase of sewerage connectivity in the future scenarios (26% for GO2050 and 17% for AM2050 compared to 2000). However, the population density is expected to decrease (6% for GO2050 and 23% for AM2050 compared to 2000). The net result is a more or less unaltered triclosan export in both the 2050 scenarios. The triclosan concentration at the rivers' mouth will increase in the future scenarios due to a decreasing basin discharge (about 80% of that in the year 2000), as a result of increased consumptive water use (irrigation) and, more in general, climate change (Mayorga et al., 2010). In the smallest basin, the Elbe, sewerage connectivity was already high in the year 2000 and the average removal of triclosan at WWTPs is very efficient. Therefore, future triclosan loads exported by the Elbe largely depend on population growth. However, the basin discharge of the Elbe shows a remarkable decrease in the future scenarios (53% for GO2050 and 59% for AM2050 compared to 2000), resulting in an increase in triclosan concentration at the mouth of the river in these scenarios. The Zhujiang basin shows a significantly increased sewerage connectivity in the future scenarios (78% for GO2050 and 28% for AM2050 compared to 2000), but at the same time an increased average triclosan removal at the WWTPs, leading to an overall decrease of triclosan export. The discharge will increase in the future scenarios (to about 120% of that in the year 2000), causing an average decrease of triclosan concentrations at the rivers' mouth. Finally, in the Ganges basin, population growth and increased sewerage connectivity result in an increase of triclosan export, although the average triclosan removal at WWTPs is expected to increase in the future scenarios. The future basin discharge is about 80% of that in the year 2000, causing an increase in TCS concentrations in the coastal seas.

4.5. Discussion

4.5.1. Comparing the calculated data with field data

We used the international database of the Umweltbundesamt (Aus der Beek et al., 2016) to extract triclosan measurement data for comparison with our model predictions. Since our model predicts triclosan concentrations at river mouths, we only included measurements close to the sea and in estuaries. Measurements close to a WWTP outlets were excluded because they may not be representative due to mixing zone effects. The resulting field data are listed in Table 4.3.

Table 4.3 indicates that our calculations are in the same order of magnitude as the field data, typically even within a factor of 2. The data for the Indian rivers differ because in our model the sewerage connectivity in these basins is set to zero. The Hudson river field data are very low. Wilson et al. (2009) already pointed out that this might be a dilution effect due to the numerous WWTPs in the downstream Hudson basin area.

Table 4.3.

Comparison of calculated average annual triclosan concentrations at the rivers' mouth (Global TCS model, 2000) with data reported in different field studies.

River basin	Concentration calculated at the end of the river (Global TCS)*	Concentration measured in surface water at the river mouth	
	(ng/L)	(ng/L)	reference
Charleston Harbour (USA)	17	5 – 14	Fair et al. (2009)
Taff and Ely (UK)	21	5 – 18	Kasprzyk-Hordern et al. (2008)
Guadalete (Spain)	44	27 – 310**	Pintado-Herrera et al. (2014)
Hudson river (USA)	13	3	Wilson et al. (2009)
Aire and Calder (UK), (part of Humber basin)	65	16-37	Sabaliunas et al. (2003)
Kaveri (India)	0	16	Ramaswamy et al. (2011)
Vellar (India)	0	5 – 10	
Tamiraparani (India)	0	5 - 16	
Zhujiang (China)	4	7 – 31	Zhao et al. (2010)
Greenwich Bay (USA)***	6 -38	1 – 7	Katz et al. (2013)

*In the Global TCS model average annual concentrations were calculated

**Range measured over a day in the estuary (tide)

***Greenwich Bay, Rhode Island is an estuary, in which different basins discharge.

Although the comparison of the measured and modelled triclosan concentrations in Table 4.3 generally shows good agreement, the Global TCS model has several uncertainties which are the result of the assumptions we made about the amount of triclosan entering the river basins (triclosan use, sewerage connectivity and sources of triclosan) and about the river retention. We will discuss these assumptions and uncertainties in the following sections.

4.5.2. Global triclosan use

In our Global TCS model we used an annual input of 500 mg triclosan per inhabitant based on the global triclosan use reported in different studies (Table 5.1). Triclosan is applied in personal care products used in developed regions, but also in hygiene products that are promoted in developing countries. Triclosan is also applied on the work floor, as a preservative for fibre, leather, rubber and polymerised materials (EPA-Denmark, 2016), for example in the textile industry in India (Ramaswamy et al., 2011). Therefore, triclosan use cannot easily be linked to socioeconomic factors such as annual income or GDP, and the triclosan consumption per inhabitant was assumed to be equal in different regions. For some regions (e.g. Africa, South America) triclosan consumption data are lacking completely. Using an average global triclosan consumption might be an overestimation in these regions, especially for rural areas, whereas in other parts of the world the actual triclosan use could be underestimated. We also did not account for a change in the per capita triclosan use in the future. During the last decade, the debate about the risk of triclosan for the aquatic environment has resulted in restrictions on the use of this substance in Europe and, recently, in the USA (FDA, 2016). As a result, triclosan production and use is being reduced in these

regions (EPA-Denmark, 2016). As long as these restrictions are not implemented globally, high triclosan concentrations could exist in the aquatic environment, especially in urban coastal regions.

4.5.3. Sewerage connectivity

Triclosan emissions from households not connected to sewerage systems were neglected. The WHO reports (WHO, 2013) that approximately 15% of the population has no sanitation facilities, especially in Southeast Asia (India, Pakistan and Afghanistan) and on the African continent. Part of the triclosan discharges in these areas will be to the land (e.g. irrigation water or septic tank sediments applied as fertiliser) and part directly into surface waters. The majority of the people not connected to sewerage systems live in rural areas; the urban population is better connected (WHO, 2013). Ignoring emissions from households not connected to sewerage systems may thus have resulted in an underestimation of the triclosan river input and the triclosan export to coastal areas, especially in rural areas.

4.5.4. Triclosan from sludge and soils

We ignored triclosan emissions to river water as a result of runoff and leaching from terrestrial systems, since triclosan is expected to be rather immobile in soils based upon an estimated K_{oc} of 9200 (Aldous et al., 2012; Healy et al., 2017). This could be an issue if triclosan contaminated sludge from private sewage tanks or WWTPs is being applied to agricultural land as a fertiliser. However, in their study on the fate of triclosan from sludge applied to soils, Butler et al. (2012) found a combined triclosan/methyltriclosan recovery in the top 10 cm layer of 49 – 84%, depending of the kind of soil they used. They argued that triclosan will be subject to biodegradation in the soil, with methyltriclosan as the main degradation product. Both triclosan and methyltriclosan are rather immobile in soils and therefore the loss of triclosan and methyltriclosan due to surface runoff and leachate can be considered small (Chen et al., 2011; Halden and Paul, 2005; Xu et al., 2009). This is in line with the findings of Gottschall et al. (2012), who did a study on the effects of application of dry municipal biosolids on agricultural soils. We therefore think that the simplification we made by only considering point sources is a legitimate one.

4.5.5. Overall loss of triclosan from the water column

The overall river retention and degradation factor that we estimated for each river basin ($FTCS_{riv}$) is an important source of uncertainty in our predictions. Triclosan will end up in the bottom sediment mainly by a two-step process: first sorption of triclosan to suspended particulates and then settling of the particulates to the bottom. Both processes are dynamic, i.e. resolution and resuspension occur (Morrall et al., 2004; Sabaliunas et al., 2003). Besides these sorption and sedimentation processes, triclosan will disappear by (bio)degradation. All these processes strongly depend on river characteristics such as depth, flow rate, turbidity, turbulence and river water pH (Sabaliunas et al., 2003; Ying et al., 2007). Unfortunately, insufficient knowledge and location-specific data are currently available to model these processes separately. For example, triclosan is an ionising substance with a pK_a of 7.9. The neutral form, which shows higher sorption and bioaccumulation, is dominant at pH values

below 7.9, while the ionised form, which is more easily degraded, is dominant at pH values larger than 7.9. Accurate quantitative description of such processes is currently unfeasible, especially on a global scale. We therefore used an overall loss rate coefficient to estimate a combined sedimentation and degradation factor, L_{TCS} . In literature, we found two studies (Morrall et al. (2004) and Sabaliunas et al. (2003)) that report an overall loss rate of triclosan in rivers, differing by a factor of 3 to 5. In both cases, the rivers were relatively small and the conditions may not be representative for the large rivers included in our global study. Following Price et al. (2010), we used the most conservative estimate for overall loss rate in our GlobalTCS model (overall loss rate of 0.06 h^{-1} (Morrall et al., 2004)). The actual loss rate in a large river may thus be considerably higher than the value used in our study, but it is difficult to state this with certainty. Additional research is therefore recommended to more accurately determine the overall loss rate of triclosan in large rivers and to describe each of the processes governing the fate of triclosan separately.

A further uncertainty is the triclosan residence time which we estimated for each river basin. Because we did not know the exact locations of the triclosan discharge points for each river basin we estimated the triclosan residence time as a function of the water residence time, the size of each basin and generic trends in population density along rivers. All these assumptions could result in an underestimation as well as an overestimation of the amount of triclosan at the river mouth.

4.5.6. Environmental risk

Taking all considerations into account, i.e., the comparison of predicted with measured data (Table 4.3) and the uncertainties discussed above, we feel fairly confident that our Global TCS model generally predicts triclosan concentrations are in the correct order of magnitude, especially for regions for which triclosan consumption data were available (e.g. Asia, Europe and North America). To account for some of the uncertainty, a factor of 2 will be taken into account below when the predicted concentrations are interpreted in terms of environmental risks.

Several studies have been performed to quantify the environmental risk of triclosan (Lyndall et al., 2010; von der Ohe et al., 2011), indicating that organisms most sensitive to triclosan are algae species (e.g., *Scenedesmus vacuolatus* and *Selenastrum Capricornutum* (von der Ohe et al., 2011)). For aquatic organisms, Zhao et al. (2013) derived a predicted no effect concentration (PNEC) for triclosan of 50 ng/L. Using the same methodology, but an enhanced procedure, Zhang et al. (2015) derived a PNEC of 26.2 ng/L in their study on ecological risks of personal care products in South China. Figure 4.5 presents a frequency diagram of the calculated triclosan concentrations at the river mouth of basins that cover at least 5 grid cells (i.e., 1163 basins covering 90% of the total land area). In the year 2000, the concentration at the rivers' mouth exceeded the PNEC of 26.2 ng/L in 6% (4 – 9%) of the basins. This number will increase to 9% (6 - 12%) in the AM2015 scenario and to 12% (7 - 15%) in the GO2050 scenario. This implies that in almost one out of ten river mouths of large rivers, the annual average triclosan levels exceed the PNEC of 26.2 ng/L in 2050.

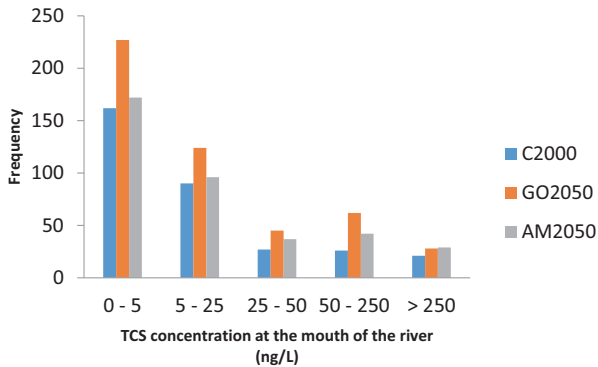


Figure 4.5.

Frequency diagram of triclosan exporting rivers to coastal areas for the scenarios C2000, GO2050 and AM2050 (the horizontal axis presents concentration areas, 0-5 means 'from 0 to 5 ng/L, 0 not included'), assuming a worldwide triclosan use of 500 mg TCS/cap/y (only the 1163 large river basins (5 gridcells or more), were taken into account).

Because our knowledge on the fate and effects of triclosan in the environment is limited, it is difficult to give an exact prognoses of the risks of using it in personal care products.

However, our simulations do show that there is reason for concern. It would be wise for authorities looking for a substitute for triclosan in personal care products. In some categories of products, triclosan can simply be left out: the antibacterial effect of triclosan does not add to the mechanical effect of washing with water and plain soap (FDA, 2010). In other products, alcohol is a good replacement. Before using other chemical agents to substitute triclosan, research has to be done on the ecotoxicological profile of those agents. Another option to reduce triclosan emissions is to equip WWTPs with advanced secondary treatment (with long WWTP residence times) and tertiary treatment to remove triclosan as good as possible (to 95-98%), for example photo degradation at a high pH (Thompson et al., 2005; von der Ohe et al., 2011) or treatment with ozone (Dodd et al., 2009; Suarez et al., 2007).

4.6. Conclusion

The Global TCS model we developed is, to our knowledge, the first global, spatially explicit model to calculate historical and future scenarios for triclosan export to coastal areas by rivers. We used it to analyse triclosan export in the year 2000 and two future scenarios for the year 2050, assuming an average global triclosan use of 500 mg/cap/y and basin-specific projections for future population growth and sewage management. Our calculations show an increased triclosan export in future scenarios (GO2050 and AM2050), especially to Southeast Asian coastal areas. This increase is caused by population growth and an increase of sewerage connectivity in that region, especially in the urban areas. We calculated that without restricting triclosan use or increasing removal efficiencies in waste water treatment, triclosan levels in rivers may increase considerably. By 2050 the number of large river basins

with a concentration triclosan at the mouth of the river that is larger than 26.2 ng/L (PNEC), could double compared to 2000. Triclosan may become a significant risk for the aquatic ecosystem in these regions. The main source of uncertainty in these prediction is the retention of triclosan in river water as determined by processes such as sorption, sedimentation and degradation. Our study can serve as a basis for more detailed future studies aimed at quantifying triclosan trends, and identifying options to reduce triclosan emissions to the aquatic environment.

Chapter 5.

Modelling global river export of microplastics to the marine environment: Sources and future trends.

Abstract

Microplastics, transported by rivers to oceans, are triggering environmental concern. This study aims to better understand river export of microplastics from land to sea. We developed the Global Riverine Export of Microplastics into Seas (GREMiS) model, a global, spatially explicit model for analysing the annual microplastics export to coastal seas. Our results indicate that riverine microplastics export varies among world regions, with several hotspots, e.g., South East Asia, and, depending on the 2050 scenario, may be doubled ('Business as usual') or halved due to improved waste management ('Environment profits'). Globally, our model simulations indicated fragmentation of macroplastics as the main source of microplastics, but this result heavily depends on the assumed fragmentation rate. Sewerage discharges contributed only 20%, ranging from 1% (Africa) to 60% (OECD countries) and decreasing by 2050 as a result of improved sanitation. We conclude that, combating microplastics in the aquatic environment requires more region-specific analyses.

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5.1. Introduction

Microplastics form a growing concern in the environment, especially in the aquatic environment. This concern is triggered by their wide distribution in both the marine environment (Bergmann et al., 2015) and surface waters (Wagner and Lamberts, 2017), and their potential adverse effects on organisms (Koelmans et al., 2016; Ryan et al., 2009; Thevenon et al., 2014; Wright et al., 2013). An important part of marine plastic pollution originates from the land and is transported by streams and rivers to seas and oceans (Jambeck et al., 2015; Lebreton et al., 2017; Schmidt et al., 2017). Lebreton and co-workers (2017) developed a regression model to calculate the global transport of land-based plastics to the sea. They estimated the global amount of plastic originating from mismanaged waste that is transported by rivers to the seas at about 1 to 2.5 million tons per year, with hotspots in Asia. These plastics are fragmented and degraded on the land, during river transport and finally in the seas and oceans, resulting in microplastics (Andrady, 2017). Microplastics are plastic particles with a typical size between 1 μm and 5 mm (Barnes et al., 2009; Eriksen et al., 2014). Important sources of microplastics in the aquatic environment are badly managed plastic waste (e.g., municipal and agricultural plastic waste), car tyres, synthetic laundry fibres, abrasives and personal care products (PCPs) (Thevenon et al., 2014). Microplastics are subdivided as 'primary microplastics', used directly in, for example, personal care products and abrasives, and 'secondary microplastics', formed from larger plastic items by fragmentation and degradation (Auta et al., 2017; Boucher and Friot, 2017; Gewert et al., 2015). Microplastics are distributed in the environment in various ways and in that context we distinguish microplastics from point and diffuse sources. In the aquatic environment, discharges of sewerage systems form an important point source (Lambert et al., 2014). Examples of microplastics originating from households and discharged into sewerage systems include microbeads from PCPs and laundry fibres. Part of the microplastics from car tyre wear will also be discharged through the sewerage system (Boucher and Friot, 2017; Kole et al., 2017). Part of the microplastics in sewerage systems will be removed by wastewater treatment plants (WWTPs), depending on the actual presence of WWTPs and the used removal techniques (Carr et al., 2016). In developing countries and in countries with a long coastline (e.g., Norway), road drainage is often directly discharged into surface waters or seas (Essel et al., 2015; Sundt et al., 2014). Plastic waste that is not properly managed, for example from households, forms a diffuse source of microplastics because it can be converted into microplastics as a result of processes like weathering, embrittlement, fragmentation and fouling (Critchell and Lambrechts, 2016; Eriksen et al., 2014; Klein et al., 2017; Koelmans et al., 2017b; Kooi et al., 2016). In WWTPs most of the macroplastics can be removed, e.g. by capturing floating plastic items and settling (Carr et al., 2016; Magnusson and Noren, 2014).

Microplastics in the aquatic environment pose a potential risk for ecosystems and organisms (Lassen et al., 2015). Research has shown that microplastics are accumulating in the world's oceans and fresh water systems, and can enter food chains (Wagner and Lamberts, 2017).

Reducing the amount of microplastics can be achieved in different ways. Besides technological solutions such as changing to (bio)degradable plastics (Boucher and Friot, 2017; Kubowich and Booth, 2017), the emissions of (micro)plastics should be reduced. This requires insight in the different sources of (micro)plastics, their spatial distribution and the presence of hotspots. Modelling can be helpful, gaining this insight. So far, on a global scale, Lebreton et al. (2017) and Schmidt et al. (2017) developed models to calculate the load of microplastics transported by rivers into the seas and oceans, both using a similar regression approach. For Europe, Siegfried et al. (2017) developed a process-oriented model that estimates the riverine export of microplastics from sewage to coastal seas, for a contemporary and some future scenarios. Siegfried et al (2017), however, did not account for other sources of microplastics, or other world regions.

The aim of this study is to contribute to a better understanding of river export of microplastics from land to sea. In addition, we aim to explore trends in global river export of microplastics for three future scenarios (year 2050) that differ in assumed levels of environmental control. We build on the approach of Siegfried et al. (2017). Special attention is paid to the relative shares of different sources of microplastics. To this end, we developed a spatially explicit model, the Global Riverine Export of Microplastics into Seas model (GREMiS), that estimates global microplastics export by rivers to coastal seas using a steady state approach.

5.2. Model description

The GREMiS model builds upon two existing spatially explicit modelling approaches: a microplastics model for point sources in European rivers (Siegfried et al., 2017) and a global model for nutrient export by rivers (Mayorga et al., 2010). Novel elements of GREMiS are (1) upscaling of the microplastics model for Europe to the globe, (2) the introduction of macroplastics as a source of microplastics, and (3) an improved representation of microplastics sources and transport.

The export of microplastics (Yld_{MP} in g/km^2 watershed/y) in GREMiS is calculated with the equation:

$$Yld_{MP} = (\sum RS_{pnt_i} + \sum RS_{diff_j}) \times F_{riv,MP} \quad (1)$$

where:

RS_{pnt_i} is the microplastics input to rivers from a point source i (g/km^2 watershed/y);

RS_{diff_j} is the microplastics input to rivers from a diffuse source j (g/km^2 watershed/y);

$F_{riv,MP}$ is the fraction of microplastics input that is exported by rivers and streams to coastal seas (0-1).

The input of microplastics into a river from point sources depends on the population density in its catchment, the population fraction connected to sewerage systems, and the fraction of the microplastics removed by wastewater treatment:

$$RS_{pt_i} = WS_{in_i} \times P_{den} \times P_{con} \times (1 - h_{w_{frem}}) \quad (2)$$

where:

WS_{in_i} is the per capita input of microplastics from a point source i into the watershed (g/cap/y);

P_{den} is the population density within the watershed (cap/km² watershed);

P_{con} is the fraction of population connected to sewerage systems (0-1);

$h_{w_{frem}}$ is the fraction of microplastics in sewage influent removed via wastewater treatment (0-1).

The input of microplastics from point source i is calculated estimating the per capita use of source i (in g/cap/y) and multiplying this with the catchment's population density (cap/km²). In this study we distinguished the sewerage discharges of three different sources as point sources, i.e., car tyre wear, laundry fibres and PCPs.

River input of microplastics from diffuse sources also depends on the population density in a watershed. An important diffuse source of microplastics is the fragmentation of macroplastics (Auta et al., 2017; Critchell and Lambrechts, 2016; Sundt et al., 2014). Fragmentation of macroplastics into microplastics is a complex process that includes different time-dependent processes, taking place both on land and in the aquatic environment. In this study we only include microplastics formed in rivers. Microplastics formed on land were disregarded, assuming that the majority of those microplastics will end up in the soil (Sundt et al., 2014).

To estimate the input of microplastics originating from the fragmentation of riverine macroplastics, we distinguished two fractions, i.e. a fast and a slow fraction. It is assumed that the fast macroplastics fraction is transported directly to the coastal sea after emission, resulting in a relatively short residence time. The slow fraction has a longer residence time as a result of getting stuck in or along the river. Since our model is non-dynamic we assume an instantaneous release of microplastics from both fractions into the river. The relative release rate of microplastics from both fractions is assumed the same:

$$RS_{diff_{MP}} = F_{macro} \times (WS_f \times t_{r,f} + WS_s \times t_{r,s}) \quad (3)$$

where:

$RS_{diff_{MP}}$ is the microplastics input to rivers as a result of the fragmentation of macroplastics (g microplastics/km² watershed/y);

F_{macro} is the relative release rate of microplastics from macroplastics (/y);

WS_f is the yearly input of macroplastics into the fast fraction (g/km² watershed/y);

WS_s is the yearly input of macroplastics into the slow fraction (g/km² watershed/y);

$t_{r,f}$ is the average residence time of macroplastics in the fast fraction (y);

$t_{r,s}$ is the average residence time of macroplastics in the slow fraction (y).

The river retention and degradation of microplastics ($F_{riv,MP}$ in Equation 1) is calculated following Siegfried et al. (2017):

$$F_{riv,MP} = (1-L_{MP})(1-FQ_{rem}) \quad (4)$$

where:

L_{MP} is the combined retention factor for microplastics as a result of sedimentation and degradation (0-1);

FQ_{rem} is the consumptive water use, being the fraction of the river water that is removed for consumptive use and irrigation (0-1).

The total average annual microplastic load is calculated by multiplying the microplastic export of each basin (Yld_{MP} in equation 1) by the basin's area and summing up all these individual loads:

$$Ld_{MP} = \sum Yld_{MP_i} \times A \times 10^{-6} \quad (5)$$

where:

Ld_{MP} is the total average microplastics load (tonnes/y);

Yld_{MP_i} is the export of microplastics from source i (g/km² watershed/y);

A is the basin area (km²).

5.3. Model input

5.3.1. Global NEWS and the MEA scenarios

The Global Nutrient Export from WaterSheds model (GlobalNEWS) (Mayorga et al., 2010) is a spatially explicit model that has been used to analyse river export of nutrients (i.e., Nitrogen (N), Phosphorus (P), Carbon (C) and Silica (Si)) to coastal zones. It describes nutrient export in terms of basin characteristics, hydrology and human activity in over 6000 catchments on a global scale. Global NEWS uses input data with a spatial resolution of 0.5 x 0.5 degrees longitude by latitude, that comprise data on land use and diffuse sources (Bouwman et al., 2009), derived from the IMAGE model (Bouwman et al., 2006), data on socio-economic drivers of point sources (Van Drecht et al., 2009) and data on hydrology, generated with the Water Balance Model Plus (Fekete et al., 2010; Vorosmarty et al., 2000). To analyse future nutrient export, the millennium ecosystem assessment (MEA) scenarios (Alcamo et al., 2005; Carpenter et al., 2005; Cork et al., 2005) were implemented in GlobalNEWS (Seitzinger et al., 2010). The four MEA scenarios, Global Orchestration (GO), Order of Strength (OS), Adapting Mosaic (AM) and Techno Garden (TG), each describe a possible global development up to the year 2050 in terms of approach towards ecosystem management (proactive or reactive) and socio-economic development (globalisation or regionalisation). Each of the MEA scenarios is characterised by two of these features: proactive, focused on globalization (TG) or regionalisation (AM) and reactive, focused on globalisation (GO) or regionalisation (OS) (Alcamo et al., 2005). The Global NEWS model has been used and validated in different studies, on a global and continental scale (Seitzinger et al., 2010; Yasin and Kroeze, 2010)

and on a regional scale (Sattar et al., 2014; Stokal et al., 2013). It has been adapted for pathogens (Vermeulen et al., 2015) and for triclosan (van Wijnen et al., 2018).

5.3.2. Basin classification

We used the economic regions as used by the World Bank (Hoornweg and Bhada-Tata, 2012) for a classification of river basins (Figure 5.1), which allowed us to allocate per capita data of microplastics discharged by sewerage systems and macroplastics from municipal solid waste to river basins in GREMiS.

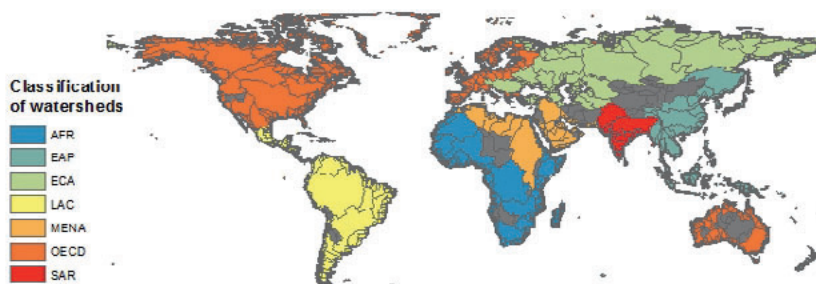


Figure 5.1.

The economic regions used in this study as distinguished by the World Bank (Hoornweg and Bhada-Tata, 2012): Africa (AFR), East Asia and the Pacific (EAP), Eastern and Central Asia (ECA), Latin America and the Caribbean countries (LAC), Middle East and North Africa (MENA), OECD countries (Organization for Economic Cooperation and Development) and South Asia (SAR).

5.3.3. Microplastics input in rivers

In this study, we distinguished microplastics from four different sources, i.e. (1) personal care products (PCPs), (2) laundry fibres, (3) car tyre wear, and (4) fragmentation of macroplastics. Microplastics from PCPs and laundry fibres can enter the environment via the sewerage system (with or without treatment) or in a diffuse way, e.g. after washing clothes in rivers. As data on diffuse sources are largely lacking, we assumed these sources were covered by the category ‘untreated sewerage discharges’ (see below). Car tyre wear can also enter the environment in different ways. We only accounted for car tyre wear discharged via the sewerage system (about one third of the total; (Boucher and Friot, 2017; Siegfried et al., 2017), assuming that the rest is absorbed by the soil where it is rather immobile (Kole et al., 2017). To estimate the microplastics input by degradation of macroplastics, we limited ourselves to macroplastics originating from municipal waste, using data from the World Bank (Hoornweg and Bhada-Tata, 2012). Other sources of macroplastics, such as mulching film used in agriculture, were left out because of the limited availability of data on these sources. We assumed that half of the inadequately managed plastic waste is transported to

ivers (Sundt et al., 2014). We used a relative release rate of microplastics from macroplastics (F_{macro}) of 3% (/y) also following Sundt et al. (2014). Once in the river, part of the plastics will reach the mouth of the river unhindered (the fast fraction), while another part will be delayed in and along the river (the slow fraction). For the fast macroplastics fraction, we estimated a basin-dependent annual average residence time as described by van Wijnen et al. (2018), whereas a residence time of 5 years was assumed for the slow fraction (see Supplementary materials for more detailed information).

Table 5.1 gives an overview of the data sets used to parameterise the model. For both years, 2000 and 2050, data are available for each river basin. In Table 5.2 the input data used for our baseline scenario (recent past) are presented. We used inputs from different data sources over the period 2000-2007 to estimate the per capita input of microplastics from car tyre wear, laundry fibres and PCPs. The estimated per capita input of microplastics in rivers from these sources for this scenario are summarised in Table 5.3, as is the per capita input of macroplastics. More detailed information about the estimated inputs is available in the Supplementary materials.

Table 5.1.

Overview of data used as model parameters in GREMIS

Model parameters	Source	Years
Population density	Mayorga et al. (2010)	2000 and 2050
Sewerage Connectivity	Mayorga et al. (2010)	2000 and 2050
Consumptive water use	Mayorga et al. (2010)	2000 and 2050

Table 5.2.

Overview of data used to estimate the per capita input of microplastics from macroplastics, tyre wear, synthetic laundry fibres and PCPs

Model input	Source	year
Municipal waste per capita	Hoornweg and Bhada-Tata (2012)	2000-2007
Percentage of plastic in waste	Hoornweg and Bhada-Tata (2012)	2000-2007
Collection rate of waste	Hoornweg and Bhada-Tata (2012)	2000-2007
Ways of waste disposal	Hoornweg and Bhada-Tata (2012)	2000-2007
Number of motor vehicles per capita	Hoornweg and Bhada-Tata (2012)	2000-2007
Laundry fiber emission	Shui and Plastina (2013)	2005
Sale of cosmetic products	Łopaciuk and Łoboda (2013)	2007

The input of microplastics originating from macroplastics depended on the per capita amount of waste, the percentage of plastic in this waste and waste collection and processing in each region (this is described in more detail in the Supplementary materials). As a result, the per capita input of microplastics originating from macroplastics in Africa, with a poor waste collection (only 45%) and a high percentage of inadequately managed collected waste

Table 5.3.

Per capita emission of macroplastics and microplastics into rivers in different economic regions as included in the baseline scenario of the current study (Figure 1) (Africa (AFR), East Asia and the Pacific (EAP), Eastern and Central Asia (ECA), Latin America and the Caribbean countries (LAC), Middle East and North Africa (MENA), the OECD countries (OECD) and South Asia (SAR)).

Plastics sources *	OECD	AFR	MENA	EAP	ECA	LAC	SAR	References
	Estimated per capita input(kg/cap/y)							
Macroplastics	8.8	27	16	23	15	23	8.5	Hoornweg and Bhada-Tata (2012), Siegfried et al. (2017), Sundt et al. (2014)
Microplastics								
Tyre wear	0.18	0.0072	0.068	0.018	0.077	0.041	0.0072	Hoornweg and Bhada-Tata (2012), Siegfried et al. (2017)
Laundry fibres	0.11	0.007	0.047	0.041	0.047	0.028	0.036	Shui and Plastina (2013), Essel et al. (2015), Siegfried et al. (2017)
PCPs	0.0055	0.0007	0.0007	0.0049	0.0049	0.0025	0.0007	Łopaciuk and Łoboda (2013), Siegfried et al. (2017)

*Only sources discharged by sewerage (car tyre wear, laundry fibres and PCPs) and macroplastics were considered.

(about 70%), exceeded that in the OECD countries, where the per capita amount of waste is larger, but the average waste collection is much higher (90%) and the leakage of waste into the environment is much smaller.

5.3.4. Removal of microplastics at waste water treatment plants

The river input of microplastics from point sources is determined by the presence and efficiency of the waste water treatment plants (WWTPs) in each basin. Plastics are removed at WWTPs that are equipped with at least primary treatment (i.e., during settling).

Macroplastics that enter a WWTP will be removed almost entirely, and also for microplastics removal efficiencies over 95% were reported (Brandsma et al., 2013; Magnusson and Noren, 2014; Talvitie and Heininen, 2014). In WWTPs with only mechanical treatment (and no extensive settling), a lower removal efficiency (50%) was reported (Magnusson and Noren, 2014).

Carr and co-workers (2016) found that microplastics particles were removed at WWTPs mainly by solids skimming and settling processes during primary treatment. The importance of effluent filters in the removal of microplastics was found to be minimal. Since detailed data on microplastics removal by WWTPs are lacking, we worked with an average microplastics removal capacity per basin. We distinguished four basin classes and estimated the average removal efficiency of each class using the average phosphorus removal for each river basin from Global NEWS (Van Drecht et al., 2009). This resulted in removal percentages of 95%, 75%, 50% and 0% for class I to IV, respectively (see Supplementary materials). Class IV (untreated sewerage discharges) refers to basins where WWTPs are generally lacking or the WWTP treatment is insufficient to remove microplastics.

5.3.5. Retention of microplastics in rivers

To model retention of microplastics particles in rivers as a result of sedimentation processes, one should ideally consider particle characteristics (particle size, particle density and biofilm formation and aggregation of particles) and basin characteristics (residence time, depth, flow rate, number of grid cells)(Besseling et al., 2016). However, detailed data on particle and basin characteristics are lacking on a global scale. The same applies for degradation data. We therefore followed Siegfried et al. (2017), who calculated microplastics export by European rivers assuming a combined retention factor for degradation and sedimentation (L_{MP}) of 0.75 for smaller basins (<4 grid cells) and of 0.9 for larger basins.

5.3.6. Future scenarios

To analyse trends on river export of microplastics for the year 2050, we created a number of future scenarios. As a starting point we took the Global Orchestration-scenario (GO scenario) from GlobalNEWS for the year 2050 (Seitzinger et al., 2010). The GO scenario is one of the four Millennium Ecosystem Assessment (MEA) scenarios that describes a future development in terms of a globalised world and a reactive approach towards environmental management, in which full access to improved sanitation and sewerage connection is pursued. In this study, we constructed three future scenarios based on the datasets of the

GO scenario for the year 2050, i.e. a ‘business as usual’ scenario (BAU), an ‘equal world’ scenario (EQW), and an ‘environment profits’ scenario (ENV) (Table 5.4).

Table 5.4.

Scenarios for microplastics export by rivers in the year 2050, based on the GO scenario i.e. business as usual (BAU), equal world (EQW) and environment profits (ENV).

Scenario	Description
BAU	In this scenario, the GO2050 datasets from GlobalNEWS ¹ for population density, sewerage connection and hydrology are used. The input of microplastics of all sources is equal to that used in the recent past ² .
EQW	As in BAU, but with a per capita microplastics input for all regions that matches that of the OECD countries in the recent past ² .
ENV	As in BAU, but with a municipal solid waste collection rate of 90% and adequate management of the collected waste, and a WWTP removal rate of 95%.

¹ Mayorga et al. (2010), Seitzinger et al. (2010)

² Contemporary calculations are referred to as ‘recent past’

5.4. Results

5.4.1. Global river export of microplastics to coastal seas

We ran GREMiS to analyse the microplastics export by rivers in the recent past and in three future scenarios for 2050, i.e., BAU, EQW and ENV (Figure 5.2). For the recent past, a total global microplastics export of 47 thousand tons was calculated, of which approximately 80% originated from macroplastics. The remaining 20% resulted from sewerage discharges, i.e., car tyre wear, laundry fibres and PCPs, with an almost negligible contribution of PCPs (i.e., less than 1%). In two future scenarios, BAU and EQW, the total microplastics export increased compared to the recent past; in the BAU scenario to 71 thousand ton/y and in the EQW scenario to 57 thousand ton/y. In the ENV scenario, the total export of microplastics more than halved to 17 thousand tons.

Figure 5.3 shows the microplastics export per region. In the BAU scenario, the microplastics export increases in almost all regions when compared to the recent past, especially in Africa (AFR), the East Asian countries (EAP) and South Asia (SAR). The EQW scenario shows a more differentiated picture with substantial increases in microplastics export in Africa (AFR), Middle East and North Africa (MENA) and South Asia (SAR), and decreases in the countries of the OECD and East Asia (EAP). These trends are mainly driven by changes in the average per capita plastic input per region, which is expected to rise in regions such as AFR, MENA and SAR. In the ENV scenario, the microplastics export decreases in all regions. Figure 5.4 shows the microplastics export for different basins within the economic regions, ranging from almost zero for catchments with small populations to over 500 ton/y in hotspot areas in (South) East Asia, the America’s and Africa.

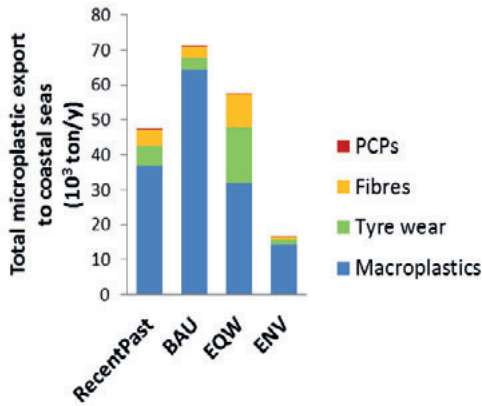


Figure 5.2.

Total global microplastics export by rivers to coastal seas as predicted for the recent past and in three future scenarios for 2050, i.e. BAU, EQW and ENV. Predictions only include riverine emissions from macroplastics and sewerage discharges containing car tyre wear, laundry fibres and PCPs.

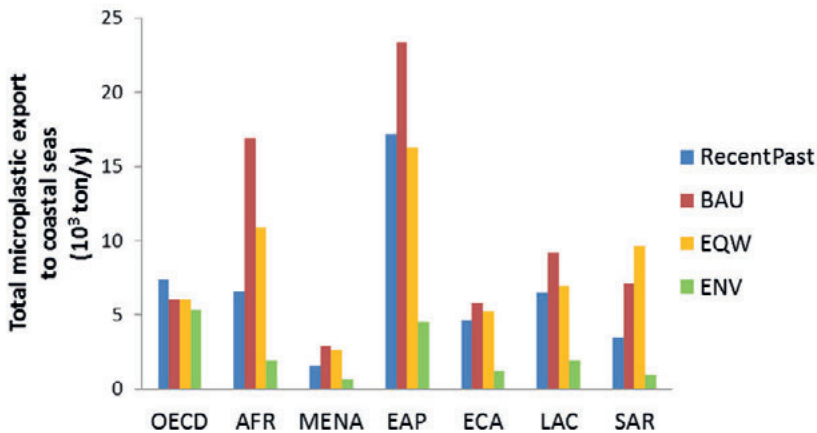


Figure 5.3.

The total microplastics export (in 10³ ton/y) in 7 global regions (i.e., Africa (AFR), East Asia and the Pacific (EAP), Eastern and Central Asia (ECA), Latin America and the Caribbean countries (LAC), Middle East and North Africa (MENA), countries of the Organization for Economic Cooperation and development (OECD) and South Asia (SAR)) for the recent past and three different GO2050 scenarios (i.e., BAU, EQW and ENV).

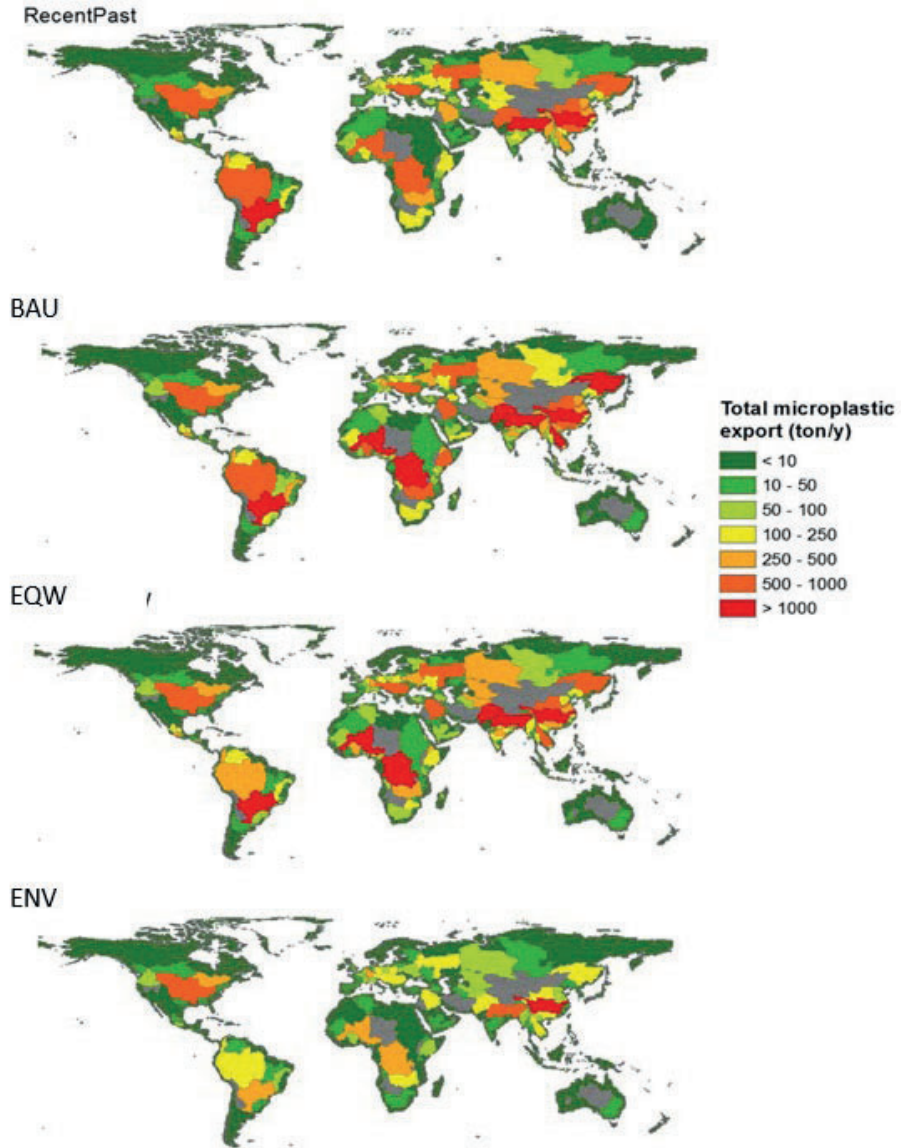


Figure 5.4. Total river export of microplastics to coastal areas for the recent past and the three future scenarios. Endorheic basins (except those of the Caspian Sea and Lake Aral) are excluded (grey).

5.4.2. Sources of microplastics in rivers

Figure 5.5 illustrates the contribution of different emission sources to the export of microplastics. In the recent past scenario, microplastics from sewerage discharges (i.e., car tyre wear, laundry fibres and PCPs) turned out to be a negligible source in Africa (about 1%), whereas in other regions the share of these sources is up to 60% (OECD), 35% (MENA and ECA) and 25% (LAC). The BAU scenario provides for increased sewerage connection and an increased removal of microplastics at WWTPs, resulting in a global decrease of microplastics export from sewerage discharges. In the EQW scenario the share of sewerage sources increased again in all regions which is mainly driven by a reduced contribution of macroplastics due to a higher waste collection efficiency, especially in regions that differed strongly from the OECD countries in the recent past i.e., AFR, LAC, EAP and SAR. Finally, the ENV scenario further reduces the emission of microplastics from WWTPs by setting their removal efficiency to 95%, resulting in a larger relative contribution from macroplastics.

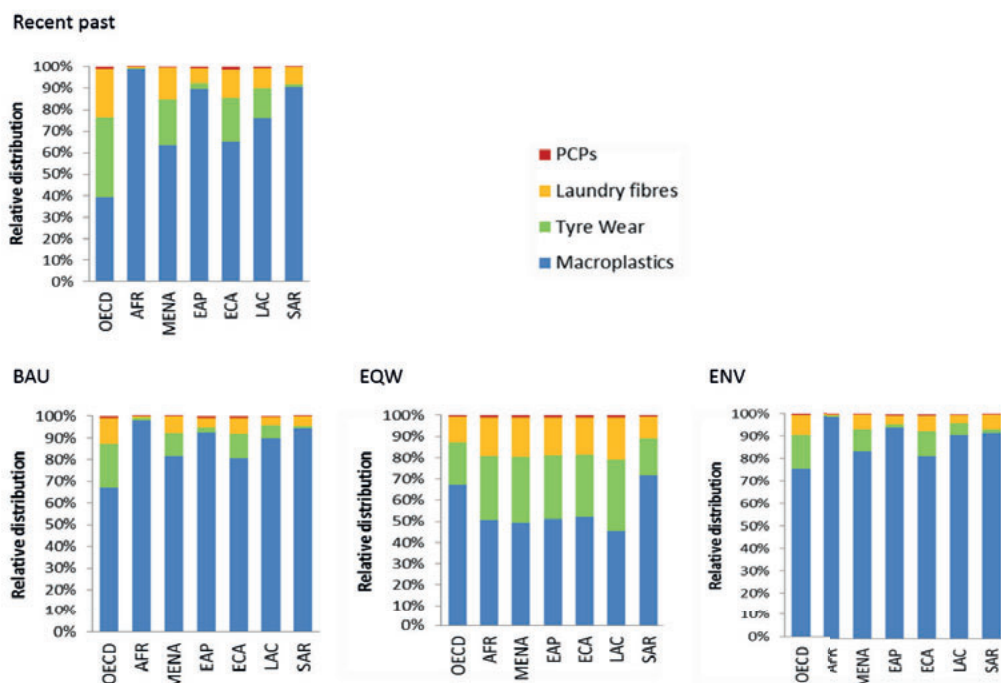


Figure 5.5.

Source distribution of exported microplastics to the coastal areas in 7 global regions (i.e., Africa (AFR), East Asia and the Pacific (EAP), Eastern and Central Asia (ECA), Latin America and the Caribbean countries (LAC), Middle East and North Africa (MENA), countries of the Organization for Economic Cooperation and development (OECD) and South Asia (SAR)) for the recent past and three different GO2050 scenarios (i.e., BAU, EQW and ENV).

5.5. Discussion

5.5.1. GREMiS predictions compared to field data and other model predictions

We compared the microplastics export that we estimated with GREMiS with field measurements and data reported in other studies (Table 5.5). The number of field data for microplastics at the mouth of rivers is limited and reported concentrations show considerable variation. This is the result of varying sampling methods (both temporally and spatially) that influence the measured amounts of microplastics. Nonetheless, the export predicted by GREMiS is within the range of values reported for major river basins, showing a large variation between different measurements. An illustration of this is a study on the Yangtze by Zhao and co-workers (2014). The amounts of microplastics measured in the Eastern Chinese Sea were much lower at 0.03-0.455 n/m³ (or 0.0009 – 0.014 g/m³) than in the Yangtze estuary at 500-10,200 n/m³ (or 1.5 – 30.6 g/m³). It is not clear to what extent these enormous differences reflect a real-world phenomenon or whether these are caused by artefacts such as differences in mesh size, the influence of wind speed, local concentration variations, differences in sampling depth, or erroneous assumptions about the average particle weight when translating particle numbers to weight.

Table 5.5.

Comparison of GREMiS data with field measurements (data adapted from Lebreton et al. (2017))

	Model predictions	Field data	
River basin	Estimated average concentration of microplastics (g/m ³)	Measured/calculated* concentration of microplastics (g/m ³)	References
Danube	0.0041	0.0020 0.0098	Lechner et al. (2014)
Seine	0.018	0.0011*	Dris et al. (2015) Lebreton et al, 2017
Rhine	0.0043	0.015* 0.0056*	Van der Wal et al. (2015)
Po	0.0035	0.044* 0.0030* 0.037*	Van der Wal et al. (2015) Lebreton et al, 2017
Yangtze	0.012	0.0009-12**/**	Zhao et al, 2014
Patapsco	0.00031	0.00051 0.0016 0.000071 0.00054	Yonkos et al. (2014)

*Actual measured microplastics concentrations are scarce. In a number of studies, numerical concentrations were reported, that were converted to mass concentrations using an average particle mass of 0.003 g (Lebreton et al, 2017).

**Measured in the Yangtze estuary and in the coastal area in the Eastern Chinese sea.

Schmidt and co-workers (2017) developed two models to calculate global microplastics export. Using a regression approach, they correlated the amount of mismanaged municipal waste with riverine (micro)plastics concentrations reported in various studies, including that of Zhao et al. (2014) on the Yangtze river. The total global microplastics export, calculated by these two models, differed considerably (with 25-75% intervals of $0.21 - 1.12 \times 10^6$ tons/y for one of the models and $1.72 - 4.38 \times 10^6$ tons/y for the other). In the Schmidt model, the share of the Yangtze basin in the total load is rather large (50-60%) compared to that in GREMiS (9%). Disregarding the contribution of the Yangtze to the total load, our total microplastics load fits in the lower range of the Schmidt calculations. For the Yangtze, our estimate is considerably lower.

5.5.2. Main challenges of modelling microplastics export by rivers

In this study, we explored the possibilities to model the global microplastics export by rivers, using GREMiS. The model includes parameters to describe processes that we considered relevant for modelling river export of plastics. These processes describe the behaviour of microplastics in rivers and on land, including macroplastics as an important source of microplastics. Regarding microplastics, the retention and degradation in rivers and the removal of microplastics in WWTPs were the main processes for which we had to make assumptions and estimate parameter values using limited data. With regard to macroplastics, little was known about a number of processes, of which the most important were the fate of macroplastics on land, the fragmentation and degradation of macroplastics both on land and in rivers, and the river retention of macroplastics. In this first attempt to model global river export of microplastics using a process-oriented approach, we made assumptions to account for these processes and estimated unknown parameters rather roughly. We discuss the implications below.

Because of insufficient knowledge and lack of spatially explicit data, we assumed an overall river retention of microplastics following Siegfried et al. (2017), knowing that this factor should actually be determined taking into account all time-dependent degradation and sedimentation processes of the microplastics and river characteristics such as flow rate, depth, turbulence and damming. The different pathways in which microplastics, originating from macroplastics, could end up in rivers are also not yet described in detail. Arisen on land, plastic waste that is not properly collected and processed will be partially discharged into rivers, leaving the remainder on the land, where it will also be subjected to different fragmentation and degradation processes. Although the mechanisms of these processes were subject of several studies (Andrady, 2017), little is known about their rates, which depend on the location and the type and composition of the plastic items (Lassen et al., 2015). In this study, we only included microplastics from macroplastics if these microplastics are formed in the rivers. Unfortunately, very little is known about the processes that describe the transport of macroplastics from the land into rivers and the conversion of macroplastics into microplastics in rivers. Following estimates on these processes reported by Sundt et al. (2014), we assumed that half of the mismanaged plastic waste is transported to rivers and used the estimated conversion rate of macroplastics to microplastics (3%/year)

reported by Sundt et al. (2014) for oceans. To apply this conversion rate to rivers, we assumed two riverine fractions of macroplastics mass, i.e., the 'fast fraction' and the 'slow fraction' (as described in the section on model inputs). All these assumptions have an important impact on our results since mismanaged macroplastics are the most important source of microplastics export by rivers in almost all global regions according to our simulations, and each of these assumptions directly influences the amount of microplastics released from macroplastics. Only in the OECD countries, with high waste collection and processing rates, sewerage discharges were more important sources of microplastics, a finding that is supported by Boucher and Friot (2017), who calculated equivalent emissions of primary and secondary plastics in OECD countries. For the marine environment, several authors reported that the majority of the microplastics resulted from secondary sources (Essel et al., 2015; Hidalgo-Ruz et al., 2012; Moret-Ferguson et al., 2010; Yamashita and Tanimura, 2007). However, empirical evidence that this also holds for microplastics in rivers is currently lacking. We conclude that, in order to improve the reliability of our predictions, it is essential that more empirical research is performed on: removal of microplastics in WWTPs, degradation and retention of microplastics in rivers, the amount of mismanaged plastic waste reaching rivers, the conversion rate of macroplastics to microplastics in rivers and factors influencing this conversion rate, the residence time of macroplastics in rivers, and the amount of microplastics in rivers originating from macroplastics degraded on land.

5.5.3. Microplastics from car tyre wear, laundry fibres and PCPs

We considered three different sources of microplastics discharged by sewerage, i.e., synthetic laundry fibres, car tyre wear and personal care products. Especially in regions with poor sewerage connectivity, we probably underestimated the input of laundry fibres and PCPs, because the population that is not connected to sewerage is likely to somehow also discharge (part of) their laundry water and personal care products into surface waters. This reasoning does not seem to apply to car tyre wear. Globally, about one third of the car tyre wear ends up in the sewerage systems (Boucher and Friot, 2017), leaving the remainder in the soil nearby the roads and in the air (Kole et al., 2017). For some basins, this assumption will not apply because road drainage will be discharged directly into surface waters or seas, causing an underestimation of the amount of microplastics transported to the seas in these regions. Taking into account the laundry fibres and PCPs that are not discharged by sewerage in our analyses, globally the percentages of microplastics from these sources increase (for fibres from 10% up to 29% and for PCPs from 0.7% up to 2%, depending on the percentage of these microplastics that actually find their way to surface waters), especially in regions where the sewerage connectivity is small, e.g., in South Asia (SAR) and East Asia (EAP). Microplastics that were removed by waste water treatment end up in the sludge that may be partly used on (agricultural) land as fertilizer (Horton et al., 2017). Thus, part of the removed microplastics will return to the environment. Although these microplastics probably become part of the soil and therefore rather immobile, it would be better to remove them permanently, e.g. by processing the sewage sludge before further use.

5.5.4. Scenarios for future microplastics export by rivers

Starting point of the future scenarios was the Global Orchestration scenario for 2050. This scenario assumes a future with a high sewerage connectivity and improved sanitation that is accessible for everyone (Van Drecht et al., 2009). As a result of increased population density and sewerage connectivity, the export of microplastics will increase, although the Global Orchestration scenario also provides for an improved removal of microplastics at WWTPs. Thus, in almost all regions the net result of implementing the GO 2050 scenario is a higher microplastics export in the BAU scenario compared with that of the recent past, except in the region of the OECD. In the OECD countries, the sewerage connectivity and collection of waste is already optimised in the recent past scenario. By improving the removal of microplastics at WWTPs, the microplastics export from WWTP discharges in this scenario decreased, as expected. In the EQW scenario, the per capita input of microplastics is equal for all catchments, since in this scenario, every person in the world is assumed to contribute equally to the emission of microplastics. The total microplastics export in this scenario is smaller than in the BAU scenario (Figure 5.3). Notwithstanding the equal per capita contributions, the EQW scenario shows considerable regional differences in river export of microplastics. This is determined by the differences in the amount, treatment, and plastic content of municipal waste and population density in each region. Finally, in the ENV scenario input from both diffuse sources and point sources is diminished by assuming better waste reduction and increased removal by WWTPs, resulting in a global reduction of microplastics export of more than 50%. All regions benefited in this scenario except the OECD region, where waste reduction and increased removal at WWTPs just changed enough to outweigh the population growth. Estimating the future amount of microplastics export, we assumed a constant plastics consumption. The World Bank (Hoorweg and Bhada-Tata, 2012) presented a prognosis of the per capita waste generation for 2025, predicting an increase for all regions except the OECD countries, and a more or less constant share of plastic in this waste. Therefore, our estimation of future microplastics export might be a cautious one.

5.5.5. Options to reduce the microplastics export

Our study projects that, globally, the major source of microplastics export by rivers is the result of fragmentation of poorly managed plastic waste. Therefore, reducing the amount of plastic waste seems a logical step to reduce microplastics export to the seas (Boucher and Friot, 2017). This could be accomplished by improving collection, processing and recycling of plastic waste and by optimising wastewater treatment, as in our 'environment profits' scenario (ENV). However, even in this scenario there is export of microplastics to the sea, which could be further diminished by policy measures that lead to a decreased use of plastics. An example of a measure that prevents plastic litter entering the environment is a ban on plastic shopping bags, as implemented in the last decades by governments in different countries (Clapp and Swanston, 2009; Steensgaard et al., 2017; Xanthos and Walker, 2017). For the OECD countries, where waste collection and processing is already well organised, sewerage discharges have a relatively large share in the total microplastics

export of rivers, although wastewater treatment removes already a large part of the microplastics. In these countries, policies aimed at prevention could actually be the only viable option to lower river export of microplastics to coastal areas.

5.6. Conclusion

River export is considered the most important source of microplastics in the marine environment. We developed a global, spatially explicit model, GREMiS, to identify possible hotspots and future trends of microplastics export by rivers. We considered four sources of microplastics, i.e., degradation and fragmentation of macroplastics, car tyre wear, laundry fibres and PCs. Our analyses indicate that river export of microplastics varies considerably among world regions. We identified a number of hotspots with high microplastics river exports, for example in South East Asia and South America. Not only the amount but also the source distribution of the exported microplastics varies between regions. According to our results, the majority of the microplastics transported by rivers originates from fragmented and degraded macroplastics items that enter rivers in a diffuse way. Globally, about 20% of the microplastics originates from car tyre wear and laundry fibres, discharged in the sewerage system, and the share of personal care products is negligible. However, in Africa, the share of microplastics from sewerage discharges is only about 1%, where in the OECD countries the share is about 60%. To mitigate microplastics export by rivers, the focus should primarily be on managing macroplastics waste and reducing plastic consumption. Decreasing the amount of microplastics from sewerage discharges, for instance by improving the removal of microplastics at WWTPs will also be beneficial, especially in regions where, at the moment, removal of microplastics at WWTPs is not effective.

Analysis of our future scenarios confirms the need to reduce the per capita plastic consumption. Our 'business as usual' scenario (BAU) showed that microplastics export of rivers may double, even if the per capita consumption of (micro)plastics does not increase. In the 'environment profits' scenario (ENV), some mitigating measures were implemented (i.e., better waste collection and optimal WWTP removal). This scenario resulted in a 50% reduction of global microplastics export by rivers. Further reduction of microplastics export might require a change in the consumption of plastic.

We should stress that the numerical results presented above strongly depend on some important assumptions about the behaviour of (micro)plastics in rivers and on land, and on parameter values estimated based on limited data. As such, our numerical results should be interpreted with care. To improve the reliability of our predictions, more empirical research has to be performed. Insight into processes, important for formation and distribution of microplastics, could help policy makers to prioritise mitigation measures for combating microplastics in the aquatic environment. The GREMiS model, that does not yet provide for detailed calculations of microplastics concentrations in individual catchments, can serve as a source of inspiration for future, more region-specific analyses.

Chapter 6.

Synthesis

6.1. Introduction

Water pollution is an important concern of the present world. Water quality is threatened by an excess of pathogens, salinity, nutrients and micro-pollutants as a result of human activities, e.g., agriculture, industry and domestic activities (United Nations, 2018).

Wastewater from these activities is discharged into surface waters and transported to coastal areas. To reduce water pollution, it is necessary for stakeholders like governments, industry, agricultural organisations and health institutes, to communicate about the issue, e.g. the causes, the effects and potential measures. The debate on water quality is supported by data generated by special monitoring programs. Modelling plays a valuable and necessary complementary role to a data driven approach, since, in general, monitoring programs are expensive and therefore not equally covering the globe. Furthermore, modelling can be used proactively by predicting environmental problems before they actually arise. It can thus provide insight in the dispersal of new emerging pollutants and in the consequences of changing practices and processes.

The aim of this thesis was to explore possibilities to expand GlobalNEWS as a modelling tool to address the environmental impact of new water pollution challenges like those triggered by large scale biofuel production, contaminants of emerging concern and microplastics. In Chapter 2-5, four case studies were elaborated, focusing on these challenges. In the following paragraphs, the findings from these case studies will be summarised, followed by a reflection on the use of GlobalNEWS in these case studies. Then, a number of models, including GlobalNEWS, are compared to identify options to improve the extension of GlobalNEWS for micro-pollutants. Finally, suggestions are presented for future research.

6.2. Findings from the case studies

In the first case study, six scenarios for the year 2050 were developed to study the effect of large-scale cultivation of first generation energy crops. In these scenarios, biodiesel production increases by around 10-30% of the diesel consumption in the year 2000. An existing Millennium Ecosystem Assessment scenario for 2050, Global Orchestration, was used as a baseline. The newly developed scenarios use either agricultural land or land that was used for other purposes for growing energy crops. Analysis of these scenarios by GlobalNEWS showed an increase (up to 30%) of nutrient export by European rivers in 2050, especially to the Mediterranean sea and the Black sea, but differing considerably among river basins. The scenarios with the largest biodiesel production (20-30% of the diesel demand of the year 2000) revealed considerable problems, e.g., the remaining area for agriculture could become too small to ensure sufficient food production and the nutrient pollution in rivers could become unacceptable. Therefore, to minimise adverse effects of biofuel production, a basin-specific approach towards energy crops could help, taking into account the specific characteristics of each basin, for example regarding the type of energy

crop that is best suitable, the specific location of farmlands and the current land use in those areas.

The second case study, expanding the first one by estimating the amount of nitrous oxide released into the atmosphere as a result of large scale growing of first generation energy crops, shows that, by 2050, the nitrous oxide emission may increase by 25- 45% depending on the scenario used. The nitrous oxide emissions vary greatly among river basins. The largest emissions were calculated for Southern and Eastern European countries. This study shows that although biofuels could contribute to lower greenhouse gas emissions, the increased emission of nitrous oxide forms an undesired side effect of growing energy crops. To minimise these nitrous oxide emissions, energy crops with a low fertiliser need, like second generation energy crops (e.g., miscanthus and willow), are preferred.

In the third case study, the GlobalTCS model was developed to estimate river export of triclosan, an organic antibacterial agent. The model was used to analyse river export of triclosan to seas in the year 2000 and in two future scenarios for the year 2050. Assuming an average triclosan use and basin-specific projections for future population growth and sewage management, this study showed an increased triclosan export to coastal seas by the year 2050, with hot spots in Southeast Asia. This increase was caused by population growth and an increase in the number of people connected to the sewerage system, especially in urban regions. In these regions, triclosan may become a significant risk for the aquatic ecosystems, with triclosan concentrations in coastal areas that exceed the predicted no effect concentration (PNEC) for aquatic organisms. In 2050, the number of large rivers with a concentration of triclosan at the mouth larger than the PNEC, may have doubled compared to 2000. To mitigate the triclosan load on rivers and coastal waters, policy makers should focus on restricting triclosan use and improving the efficiency of triclosan removal during waste water treatment.

Finally, the GREMiS model was developed for analysing global river export of microplastics to coastal seas. Various sources of microplastics were considered, i.e. tyre wear, laundry fibres, personal care products and degradation and fragmentation of macroplastics. This case study showed that both the amount of microplastics exported to coastal areas and the source distribution varies widely between regions. For example, globally, the majority of the microplastics originates from fragmented and degraded macroplastics that enter the river in a diffuse way. Car tyre wear and laundry fibres discharged in the sewerage system only contribute for about 20% and the share of PCPs is negligible. However, this global trend does not apply to all regions. In Africa, for instance, the share of microplastics from sewerage discharges is only about 1% while in the OECD countries it is about 60%. By 2050 the microplastics export may double, even without increasing the per capita plastic consumption. Therefore, in an effective strategy to mitigate microplastics export by rivers the focus would be on (1) reducing the per capita plastic consumption and (2) other measures to minimise microplastics export that are more region-specific, such as the improvement of waste collection and waste processing.

6.3 GlobalNEWS as a starting point for water quality modelling

In Chapters 2-5, GlobalNEWS was used as a starting point for the modelling of new scenarios and new pollutants. GlobalNEWS is a comprehensive, transparent and spatially explicit model that is relatively easy to use (Mayorga et al., 2010). GlobalNEWS calculates annual average loads for each basin, suitable for analysing trends on a global scale, especially when detailed input information, needed for detailed modelling of river export, is unavailable. Therefore, it provides a useful basis for extending it with new scenarios and pollutants (see Figure 1). Moreover, to model future river export, future developments in population and prosperity growth, improved sewerage connection, improved waste management and land use change as a result of food and energy demand have to be considered. Thanks to the implementation of future scenarios, i.e., the Millennium Ecosystem Assessment scenarios, GlobalNEWS is suitable for future predictions on a global scale (Seitzinger et al., 2010).

GlobalNEWS was developed to calculate river transport of nutrients, and to that purpose a lot of basin characteristics, like slope, land use, precipitation are accounted for in the model. However, to extend the model for other pollutants, it will be necessary to add other basin characteristics that influence the behaviour of these pollutants in the river basins. For example, for ionisable substances, solubility and speciation will be influenced by pH, and thus it will be important to take into account the pH profile of each river. An example of such an ionisable substance is triclosan, used in the third case study. Expansion of GlobalTCS with data on the pH profile of each basin might therefore contribute to a more adequate calculation of triclosan exports to the coastal areas. Furthermore, for some pollutants various time-dependent processes are relevant. For example, in GlobalTCS, an overall degradation and sedimentation term was added, to account for various time-dependent degradation and sedimentation processes that influence the final transport of triclosan to the mouth of a river. For microplastics an important time-dependent process is the fragmentation of macroplastics, both in rivers and on land, that was estimated in GREMiS. To include these time-dependent processes, more data on basin characteristics are needed, for example about the residence time of the water in each basin.

In the scientific literature, various water quality modelling tools for different pollutants are reported, with different goals, on different scales and with different characteristics (see for more information Section 6.4). In general, the choice of a certain tool is mainly determined by the purpose of the intended research. In addition, the type of contamination and the availability of the tool also determine this choice. For the case studies described in this thesis, a GlobalNEWS-like approach fits rather well because they focused on predicting global trends and identify hot spots. The choice for this approach was also determined by the type of contaminants, i.e. nutrients, that were focused on at the start of the study. In the next section, a number of large-scale water quality models, including GlobalNEWS, will be compared, aiming to understand alternative modelling approaches and to determine whether these approaches contain useful options for global modelling of pollutants in a GlobalNEWS-like approach.

6.4 Other water quality modelling tools

In the last decades, many different tools to model river export of pollutants have been developed. To compare different modelling approaches, eleven models, important for river export of nutrients, chemicals and (micro)plastics were selected (Table X1), all operating on a large spatial scale. Each model has its own strengths and limitations (Table X2).

Comparison of the selected models raises the question what characteristics a modelling tool that simulates riverine export of different pollutants simultaneously should have. Features of such a tool that emerge from both Tables X1 and X2 (and the underlying literature) are:

- The spatial and temporal scale of the input and output data is sufficiently detailed for the purpose of the intended investigation;
- To take into account the total amount of a pollutant released into the environment, both point and diffuse sources must be included;
- The underlying hydrological model accounts for those river characteristics that are relevant for predicting the concentrations or loads of pollutants, such as the flow-rate, total discharge and pH profile.
- Population density and sewerage connectivity in each river basin are taken into account;
- The characteristics of waste water treatment (in WWTPs) along the rivers are taken into account;
- Degradation and sorption processes in the rivers, behind dams and on the land, are included;
- The model provides output for different substances and includes source attribution of them;
- The model can be used to explore future trends through scenarios analysis;
- The model is relatively simple to use and computational demand is reasonable;
- The model is validated and trustworthy.

Several of these characteristics apply to each model in Table 6.1. None of the models from Table 6.1 is already used for as well nutrients, organic pollutants and microplastics. In general, the tools are used for different contaminants within a larger group, e.g., various nutrients or different pharmaceuticals and pesticides. Interactions between different pollutants in rivers is not yet included in the models. There are models in which the rivers are divided into segments, making it possible to use more detailed input data. These models generally show a rather high complexity and a correspondingly high computational demand. The complexity of the models increases with the number of processes used for modelling. For chemicals, e.g., pharmaceuticals and pesticides, including time-dependent processes, such as (bio)degradation and sedimentation, in the modelling approach seems essential, as was shown in the case study on triclosan. However, including more processes often means that more input data are required. If these data are not available, they must be estimated, increasing the uncertainty of the model predictions. The spatial and temporal scale of the model also contributes to the complexity of a model. To model different pollutants with the

same approach on a global scale, a balance must be found between all these characteristics (e.g., spatial and temporal scale, computational demand, complexity of the model, availability of input data). This balance largely depends on the purpose of the intended research. For a quick determination of global hot spots of aquatic pollution or the indication of global trends in future scenarios, a lumped model, that calculates annual averages, like GlobalNEWS, could turn out to be an appropriate choice. To calculate pollutant concentrations at specific locations in a river basin, a finer scale model, that takes into account the precise location of cities, agricultural and industrial areas and the location of existing waste water treatment plants, will probably be a better choice.

Water pollution is a complex problem. Many pollutants, that end up in surface water every day as a result of all kinds of (human) activities, are transported by the rivers independently, but will often interact. To model the river export of these pollutants together, especially at higher spatial scales, using a multi-pollutant model is preferred to different single-pollutant models. Multi-pollutant modelling is advantageous for a number of reasons (Kroeze et al., 2012; Kroeze et al., 2016): (1) Different pollutants often have similar sources and sinks, (2) Different pollutants undergo similar chemical, physical and biological degradation and transformation processes, (3) River characteristics apply for all pollutants, (4) Interactions between pollutants can be easier accounted for, (5) A multi-pollutant approach can be managed and communicated more easily to different stakeholders and (6) Developing new modelling tools each time a new issue arises, costs considerable resources. The development of a multi-pollutant model, that models various pollutants, taking into account interactions between them, is complicated. Formulating various scientific challenges to overcome, including the lack of input data, the difference in modelling approaches used so far and the complexity of the behaviour of various contaminants in rivers, Strokal et al. (2019) proposed a promising multi-pollutant modelling strategy.

Table 6.1.
Water quality models for nutrients, chemicals and (micro)plastics

Model	Environmental compartment	Pollutants	Spatial Scale	Temporal Scale	Comments	References
GlobalNEWS	River basins, Coastal waters	Nutrients (N, P, C and Si)	Global (>6000 basins)	annual	Calculates annual river export of nutrients to coastal areas (steady state approach), from both diffuse sources and point sources, in the past, present and for future scenarios (2030 and 2050, MEA scenarios).	(Mayorga et al., 2010; Seitzinger et al., 2010)
IMAGE-GNM	Surface waters	Nutrients (N, P)	Global 0.5° x 0.5°	annual	Nitrogen and Phosphorus export to surface waters (mass balance approach), from both diffuse sources and point sources, in the past, present and for future scenarios (SSP scenarios).	(Beusen et al., 2015)
GloBio-aquatic	Rivers, lakes, wetlands	Nutrients	Global (30° x 30')	annual	Models empirical relationships between environmental drivers (e.g., land-use change, hydrological disturbance and climate change) and their impact on biodiversity	(Janse et al., 2015)
WorldQual	Rivers, lakes	Biochemical oxygen demand, Bacteria, Total phosphorus and Total dissolved solids	Continental (5° x 5')	monthly	Models water pollution loadings to surface waters. WorldQual is part of the WaterGAP modelling and is used in addition of the GEMStat data (REFs). In WorldQual, future scenarios are not yet implemented.	(UNEP, 2016)
GREAT-ER	Rivers	Pharmaceuticals	Europe	monthly	Distribution of APIs in rivers, (taking into account API consumption, removal at WWTPs, human metabolism and dilution/dissipation) estimating PECs throughout rivers.	(Kehrein et al., 2015)
Phate	Rivers	Pharmaceuticals	US	annual	Steady state mass balance approach. Single media tool (only rivers)	(Anderson et al., 2004)
ePIE	River basins	Pharmaceuticals	Europe (30 arcsec)	monthly	Mass balance approach to estimate PECs of APIs in US rivers. APIs that result from human medicine use. Input into rivers is based on average annual human consumption.	(Oldenkamp et al., 2018)
WASP	Various water bodies	Nutrients (N and P), Biochemical oxygen demand	Can be used globally	daily	A high-resolution spatial model to calculate concentrations of active pharmaceutical ingredients, that answers locally specific questions.	(Gao and Li, 2014; Wool et al., 2003)
SWAT	River basins and sub-basins	Nutrients, pesticides	Global (subbasin)	daily	A hydrological model that calculates the volume and quality of water on a daily basis, taking into account various water resources and diffuse pollution.	(Vigerstol and Aukema, 2011)
Plastic model (Siegfried)	River basins	Point-sources of microplastics	Europe (basins)	annual	Calculates annual river export of microplastics to European coastal areas (steady state approach) from point sources, in past, present and a future scenario (2050, MEA scenarios)	(Siegfried et al., 2017)
Plastic models (Lebreton and Schmidt)	River basins	(micro)plastics from mismanaged plastic waste	Global (30 arcsec (population data))	monthly	Regression models, that calculate plastic inputs from rivers into oceans from mismanaged plastic waste. Seasonality is taken into account. Schmidt et al (2017) refines the Lebreton model by taking a larger data set and treating macroplastics and microplastics separately. Population data used with a resolution of 30 arcsec, plastic waste data and waste disposal data on a country scale.	(Lebreton et al., 2017; Schmidt et al., 2017)

Table 6.2. *Strengths and limitations of water quality models for nutrients, chemicals and (micro)plastics (as listed in Table 6.1)*

Model	Strengths	Limitations
GlobalNEWS	Relatively simple model Performs future scenario simulations and analysis Diffuse sources and point sources are included Provides output for different nutrients and includes source attribution of them.	Calculates annual average river export at the rivers' mouth, using basin averages for waste water treatment and river retention. Uses future scenarios (MEA scenarios) that are rather outdated
IMAGE-GNM	Calculates output at grid cell scale Implements SSP scenarios to perform future scenarios simulations and analysis (in development) Provides output for N and P and includes source attribution of them. Implements different water bodies, e.g., rivers, lakes and wetlands.	Rather complex model, that has a high computational demand Treats wastewater discharge like some diffuse source Does not include consumptive water use Goal is to model impact of environmental drivers, among which nutrient load, and biodiversity.
GloBio-aquatic	Part of the WaterGAP modelling Provides output for BOD, Total phosphorus, total dissolved solids and faecal coliform bacteria.	Future scenarios are not implemented. Point-sources are not included. Does not provide extensive output for nutrients
GREAT-ER	Uses WWTP-outlet as source The model considers volatilisation and degradation. Degradation processes were included ((pseudo) first order).	Only calculates pharmaceuticals in European riverbasins Point sources are accounted for (WWTP outlet), non-point sources are considered as evenly distributed along the total length of the river.
Phate	Uses WWTP-outlet as source Includes WWTP removal efficiency for each treatment type Degradation processes were included ((pseudo) first order)	Only calculates pharmaceuticals in 11 US riverbasins Point sources are accounted for (WWTP outlet), non-point sources are considered as evenly distributed along the total length of the river.
ePIE	Characterisation of river flow (annual mean, monthly highest and lowest mean) with FLOIK dataset (based on an ensemble of artificial neural network regressions) at a spatial resolution of 30 arc sec. Emissions from WWTPs and from agglomerations with incomplete connection. Removal during treatment accounted for via SimpleTreat4 Degradation processes were included ((pseudo) first order). WASP has a high flexibility permitting simulations of different kinds of surface water. It provides lots of possibilities to link the model to other models.	Point sources are accounted for (WWTP outlet) and agglomerations with incomplete connection to WWTPs. Diffuse sources are not specified further. Input data needed for APIs are country-specific yearly consumption data Not yet implemented globally
WASP	WASP has a high flexibility permitting simulations of different kinds of surface water. It provides lots of possibilities to link the model to other models.	Rather complex model, that has a high computational demand, requires an extensive amount of data for calibration and verification.
SWAT	Distributed, physically based model Dynamic modelling	Rather complex model, that has a high computational demand Focus on agricultural areas
Plastic model (Siegfried)	Relatively simple model Performs future scenario simulations and analysis Includes different point sources Includes different plastic sources, e.g. car tyre wear, personal care products and fibres	Only calculates microplastics export in European riverbasins Calculates annual average river export at the rivers' mouth, using basin averages for waste water treatment and river retention. Uses future scenarios (MEA scenarios) that are rather outdated Does not include diffuse sources of microplastics
Plastic models (Lebreton and Schmidt)	A regression model, based on empirical data Treating macroplastics and microplastics separately.	The underlying data set was compiled from different data, that where sampled at different locations and with different sampling methods. No future scenario simulations and analysis included

6.5 Building trust in the models

In this thesis three models were used to predict river export of different pollutants. The use of models, in which by definition the reality is greatly simplified, entails uncertainties. Four types of uncertainty can be distinguished, i.e., uncertainties regarding (1) the framing of the environmental problem, (2) the model, (3) the future scenarios and (4) the input data (Halffman and Ragas, 2016; Huijbregts et al., 2003). These types of uncertainty also apply to the case studies, albeit not to the same extent for every case study.

Framing uncertainties are related to the definition of the environmental problem. They are the result of differences in the perception of problems and prevailing norms and values in society. Uncertainties that are the result of choices about what should be included in a study and what not, belong to this category (Halffman and Ragas, 2016). In this discussion, framing uncertainties will be limited to these delineation uncertainties, present in all case studies. In the biofuel case studies, only first generation biofuel crops were taken into account and the impact on nutrient transport was only calculated as the result of changing land use, disregarding the possible consequences for, for example, livestock farming. In GlobalTCS and GREMIS, delineation mainly concerned the sources that were included in the study. In GlobalTCS, only triclosan discharge by way of sewerage was included, disregarding the triclosan that was directly discharged on surface waters or that was reintroduced in the environment by using the sludge from wastewater treatment plants in agriculture. Furthermore, the degradation products of triclosan in the aquatic environment have been disregarded. In GREMIS, several sources were not taken into consideration, such as, for example, the plastic covering film used in agriculture. For the sources that were included in this study, assumptions have been made to quantify them for each world region, introducing large uncertainties.

Model uncertainties arise from assumptions and simplifications made in the model, reflected in the limited selection of model parameters and process descriptions. Since GlobalNEWS was used as the basis of the modelling, its uncertainties, described by Mayorga et al. (2010) also applied to the case studies. Extending the model resulted in more uncertainties, because processes were added to the model, of which the parameters, at least partly, had to be estimated. For example, assumptions were made to calculate N₂O emissions due to aquatic nitrate concentrations (see Chapter 3), and to account for river retention of triclosan and microplastics (see Chapters 4 and 5).

Scenario uncertainties are associated with the choice for certain scenarios and the input needed for these scenarios. Together, the various future scenarios provide a picture of the possible future developments. Thus, the 'biofuel scenarios' in the first two case studies outline the possible nutrients export as a result of large-scale growing of energy crops. In these studies, the scenario uncertainty is rather large, since assumptions were made about the kind of energy crops used in these scenarios and the associated fertiliser use. Furthermore, assumptions about future population growth and development of sewerage systems, as implemented in GlobalNEWS, were followed. In the triclosan and microplastics

case studies, the scenario uncertainty is also large, because there are only few estimates about the future use of triclosan or (micro) plastics on different continents.

Finally, there are also *input uncertainties*, associated with other input parameters. The input parameters, used in the different studies, were derived from different sources, including GlobalNEWS (Seitzinger et al., 2010). In the microplastics case study, different data from the Worldbank (Hoornweg and Bhada-Tata, 2012) were used. These data are often supplied by governments and are of varying quality. Nevertheless, these data are frequently used in various studies (Jambeck et al., 2015; Lebreton et al., 2017).

Due to these uncertainties, the output of the models developed in this thesis, i.e., GlobalTCS and GREMiS, is not necessarily representative. To gain confidence in predicting river export of different pollutants with these models, different methods are available. The method mostly used in model development is validation, i.e., comparing modelling outputs with measured field data (Augusiak et al., 2014). However, comparing model predictions with measured data is not always possible, for example when it concerns future predictions or when the amount of available field data is limited, as in the case studies on triclosan and microplastics. Stokal (2016) described several other options to build trust in a model, such as comparing the model results with those of other modelling studies, sensitivity-analysis, assessing the input data used and by using expert knowledge. The latter can be used, for example, to identify and to work out important model parameters, for which literature is lacking. An example of such model parameters are those that describe the conversion of macroplastics into microplastics in the microplastics case study.

GlobalNEWS, used in the biofuel case studies, was extensively validated for nutrients by Mayorga et al. (2010) and Seitzinger et al. (2010) and in different other studies, on different scales (Amin et al., 2107; Pedde et al., 2017; Qu and Kroeze, 2010; Sattar et al., 2014; Stokal and Kroeze, 2013; Suwarno et al., 2013; van der Struijk and Kroeze, 2010; Yasin and Kroeze, 2010). This makes GlobalNEWS a reliable model to predict global riverine export of nutrients. Data on triclosan concentrations in coastal areas are scarce. Therefore, validation of GlobalTCS by comparing model outputs and measured field data is limited and perhaps not convincing. However, in GlobalTCS a lot of model parameters are taken from or based on those of GlobalNEWS, contributing to the reliability of the model. Finally, there is also relatively little field data available for microplastics in coastal seas, making validation of GREMiS by comparing field data with model outputs only possible to a limited extent. However, river transport of microplastics to coastal seas has also been modelled by Schmidt et al (2017), which allows a comparison of the results of both models for validation. Given the rather different approaches and varying uncertainties of both modelling methodologies, they match surprisingly well (as discussed in Chapter 5).

The lack of adequate data sets is a general problem of modelling on a global scale. Both the model parameters (e.g., hydrology) and the scenario data (e.g., future population data) are scarce for several economic regions. For example, very little data are available for Africa,

both about the input of pollutants into river basins and about measured concentrations of those in the environment. The problem of data scarcity is enhanced by producers hesitating to make the available data on their production and activities public. The UN Aarhus convention, that entered into force in 2001, established a number of rights to the public, among which the right of public participation in environmental decision-making and of public access to environmental information (Hartley and Wood, 2005), making it possible for the public to access available governmental data. However, for non-government organisations the Aarhus convention does not apply. Therefore, gaining access to production, consumption and emission data is difficult and, if possible, it can be rather expensive. To solve this, relevant stakeholders, e.g., industry, should be encouraged, or even forced, to make more data public, requiring international collaboration.

6.6 Future research

This thesis shows that the approach of GlobalNEWS can be applied to other compartments and other pollutants. Nevertheless, there are many challenges to face. Firstly, to develop a trustworthy model, it is essential that more empirical research is performed to obtain more reliable data as input data, e.g., population and land use within a catchment, actual basin discharge and location of dams. These data are available for many river basins in GlobalNEWS, but not for all. In some regions, e.g., in Africa and South America, also the accuracy of the given data is questionable. Field data are also important to validate a model, making it trustworthy enough to use it for future predictions.

Secondly, GlobalNEWS was developed for nutrients. The case studies on triclosan and microplastics show that to model other pollutants additional model parameters have to be included, for example to account for temporal processes that determine their river retention, such as degradation and sedimentation. In addition, there are many uncertainties about the processes on land that determine the input of pollutants in surface water, such as the transport of macroplastics from the land into rivers and the conversion of macroplastics into microplastics. Another challenge is formed by wastewater treatment along the rivers. Especially for catchments in Africa, Asia and South America, the removal of pollutants like triclosan and microplastics in wastewater treatment facilities is often unknown. Because this concerns the properties of individual installations, it is also important in the future to know more about the location of various wastewater treatment sites along the river, and to take this knowledge into account in the models.

Thirdly, to model river export of different pollutants, various sources have to be taken into account, i.e., diffuse sources and point sources, as implemented for nutrients in GlobalNEWS. Many emerging pollutants are distributed in the environment by sewerage. In GlobalTCS sewerage is considered the only source of triclosan. To extend GlobalTCS for other chemicals, other sources certainly need to be reflected upon, leading to the implementation of new model parameters. For example, when modelling river export of pesticides, diffuse sources, e.g., agriculture, have to be considered as well. The case study on microplastics shows that including a diffuse source of microplastics, i.e., badly managed plastic waste, is

possible, but complicates the modelling considerably. Therefore, more research has to be done to select the most important model processes that determine the modelling of diffuse sources of different chemicals.

Finally, to predict future riverine export of pollutants, it is important to use a number of scenarios, in which possible future developments, regarding socio-economic development, climate and land use in the future, are outlined. These scenarios can be translated into data, and implemented in the model. The millennium ecosystem assessment scenarios, formulated in 2005 (Alcamo et al., 2005), have since become outdated on several points. For example, compared with the implemented scenarios in GlobalNEWS, the estimated population growth for the year 2050 has increased by about 10-30% (Seitzinger et al., 2010; United Nations, 2017). Not only the future socio-economic parameters such as the population density have changed over the last decades, but also the parameters regarding land use, e.g., percentage of agricultural land within a river basin, climate and hydrology, e.g., basin discharge. Therefore, the millennium ecosystem scenarios should be updated or replaced by another set of future scenarios, such as the Representative Concentration Pathways (RCPs) and the Socioeconomic Shared Pathways (SSPs). These scenarios were already implemented for nutrients in different studies on a global scale (van Puijenbroek et al., 2015) and for China (Wang et al., 2017).

6.7 General conclusions

The aim of this thesis was *“to explore the possibilities to expand GlobalNEWS to address the environmental impact of new water pollution challenges like those triggered by large scale biofuel production, contaminants of emerging concern and microplastics”*, motivated by the idea that river export of different pollutants is, at least partly, determined by similar processes and parameters.

As illustrated by the case studies, a GlobalNEWS-like approach can be used to predict nutrient export in new scenarios and for other compartments (i.e., atmosphere) and to predict riverine export of other pollutants (i.e., triclosan and microplastics).

New scenarios were developed as variants of the, in GlobalNEWS implemented, Millennium Ecosystem Assessment scenarios. In the case study on biofuels, scenarios were developed in which different assumptions about appropriate energy crops and the synthetic fertiliser use that goes with it were included. Also in the other case studies, the Millennium Ecosystem Assessment scenarios were adapted to predict future triclosan and microplastics export.

Although GlobalNEWS focuses on the water compartment, there are possibilities for modelling export into other compartments, as demonstrated in the case study on nitrous oxide emissions. In this case study, the nitrous oxide emissions were calculated using the input of synthetic fertiliser from agriculture, both directly from agricultural soils and indirectly from aquatic systems, after leaching and runoff of nitrogen from fertilised soils. In

a similar way, the atmospheric emissions of other pollutants, e.g., pesticides, could be calculated.

As demonstrated in the case studies on triclosan and microplastics, it is possible to use a GlobalNEWS-like approach to analyse river export of contaminants other than nutrients. Both the sewerage system (triclosan, microplastics) and diffuse sources (microplastics) were taken into account. Therefore, river export of other contaminants from these sources, e.g., pesticides and antibiotics, could also be analysed with similar models. Despite these promising options, extending GlobalNEWS for contaminants other than nutrients remains a challenge. Temporal processes, like degradation and sedimentation, important for river retention of chemicals, are rather difficult to implement, because model parameters that describe river characteristics like pH, temperature and sediment type are lacking in GlobalNEWS. Wastewater treatment plants are not described in sufficient detail and typical rate constants of these processes for each river are unknown. In GlobalTCS and GREMiS, these characteristics are only moderately taken into account, making these models suitable for calculating global trends. Moreover, also input data, as for example typical rate constants for pollutants in the river are largely unknown.

As a result of the relatively simple design, GlobalNEWS is suitable for future predictions on river export of contaminants. In the last decades, the vision of socio-economic and climatological development in the future has changed from that expressed in the Millennium Ecosystem Assessment scenarios, implemented in GlobalNEWS. Therefore, these future scenarios should perhaps be replaced by more contemporary ones, such as the Representative Concentration Pathways (RCPs) and the Socioeconomic Shared Pathways (SSPs). In addition, more up to date hydrological and socio-economic data could be used.

The case studies show that a GlobalNEWS-like approach could deliver promising results for a relatively quick screening of river export of different pollutants on a global scale. Drawback of this approach is that it is difficult to implement temporal processes, e.g., degradation, agglomeration, sedimentation and fragmentation, that are important for the river export of many contaminants. The water quality debate will benefit from multi-pollutant modelling, since the output for different pollutants will be easier to interpret by the members of different societal groups than when using multiple models. A multi-pollutant model offers better opportunities to assess water quality as a whole, making the interpretation of the results less complex. A possible approach for the development of such a model is described by Stokal et al. (2019), addressing various challenges, such as the complexity of the behaviour of various contaminants in rivers, possible interaction between different contaminants and the general lack of input data for such a model.

Implementing the sustainable development goals (SDGs) will require considerable effort in the near future. Concerning SDG6 ('Clean water and sanitation'), the UN is committed to enhanced monitoring and to promoting 'good government' practices to improve water quality. Modelling could be a useful addition to the measures already proposed. By using

models to identify future hot spots, it will be possible to extend monitoring practices in a more targeted way. Furthermore, multi-pollutant modelling could also play a role supporting the linking of the different SDGs, to prevent passing on environmental problems from one SDG to another. The case studies in this thesis show that global modelling of various contaminants is possible with a relatively simple tool. Hopefully they contribute to the development of future modelling and, with that, to a more sustainable world.

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Supplementary Materials

Chapters 4 and 5 of this thesis refer to supplementary materials, that contain additional information. Originally, these supplementary materials were available with the published articles. In the following paragraphs these materials, adjusted to the thesis' format, are included.

Supplementary materials to chapter 4

Global TCS

The GlobalTCS model was based on the Global NEWS model (Mayorga et al., 2010; Seitzinger et al., 2010) which calculates the river export of nutrients to coastal seas worldwide. The equations and data sets of Global NEWS were used and modified as a starting point for calculating the river export of triclosan.

Equations

Nutrients can enter rivers in different ways. In Global NEWS two main groups of sources are distinguished, i.e. point sources and diffuse sources. Each source has its own set of equations in Global NEWS.

Triclosan was assumed to be emitted to rivers by point sources only, i.e. through sewage systems. Therefore three Global NEWS equations for point sources were used and adapted as described in chapter 4 of this thesis. These three equations (Table S4.1) define the river export of triclosan (Yld_{TCS}).

Table S4.1.

Comparison of equations in GlobalTCS and GlobalNEWS

Global TCS		Global NEWS ¹
Equation number	Equation	Equation number
1	$Yld_{TCS} = RS_{pnt,TCS} \times FTCS_{riv}$	1
2	$RS_{pnt,TCS} = (1-hw_{frem,TCS}) \times I \times WShw_{TCS}$	2
3	$FTCS_{riv} = (1-L_{TCS})(1-D_{TCS})(1-FQ_{rem})$	17

¹Mayorga et al. (2010)

Data sets

Several data sets of the Global NEWS model were used in GlobalTCS, such as population density, sewerage connectivity and hydrology data (Table S4.2). Data was used for the year 2000 and for the year 2050. The latter data originated from different future scenarios implemented in Global NEWS by Seitzinger et al. (2010).

Removal of triclosan at WWTPs

The amount of triclosan entering rivers depends on the sewerage connectivity in a river basin and the fraction of triclosan removed in WWTPs. We estimated the WWTP treatment efficiency of triclosan using the removal efficiency of phosphate available in Global NEWS (Table S4.3).

Table S4.2*Data sets used in GlobalTCS*

Model inputs	
Population	Global NEWS ¹ - Gridded (0.5°x0.5°) - Per river basin
Sewerage connectivity	Global NEWS ¹
Consumptive water use	Global NEWS ¹
WWTP treatment efficiency	Global NEWS ^{1,2} (for Phosphate) (avg per basin)
Hydrology	Global NEWS ¹ (data generated by the Water Balance Plus model ³)
Damming	Global NEWS ¹ (for TSS) (avg per basin)

¹Mayorga et al. (2010) ²Van Drecht et al. (2009) ³Fekete et al. (2010)

Table S4.3.*Removal of triclosan at WWTPs in GlobalTCS*

Average removal of phosphate ¹	Description of the removal process	Assumed average removal of triclosan
<20 %	The dominant way to discharge sewage into the river is without treatment	0%
20-80 %	The WWTPs are equipped with primary and secondary treatment; triclosan is mainly being removed by settling during these two steps	60%
>80%	The majority of the WWTPs will perform extensive biological treatment	90%

¹Mayorga et al. (2010)

Supplementary materials to chapter 5

Macroplastics as a diffuse source of microplastics

The amount of plastic waste was estimated per economic region (Figure 5.1). We used the amount of municipal waste (kg/cap/day) as assumed by the World Bank (Hoornweg and Bhada-Tata, 2012) to estimate the average amount of plastic per capita in each region, taking into account the average percentage of plastic in waste, the average collection rate of the waste and the different waste disposal ways. Inadequately managed waste is waste that is: (1) collected, but not properly processed, or (2) not collected at all. Collected waste is inadequately managed if it is dumped or, in low income and lower middle income countries, is sent to landfills (Hoornweg and Bhada-Tata, 2012). Therefore, in high income countries all collected waste is adequately managed and in other countries only part of the collected waste is processed properly (Table S5.1).

Table S5.1.

Macroplastics input from municipal waste

Region	Average amount of municipal waste (kg/cap/day) (kg/cap/y)	% plastics in waste	Average amount of plastics (kg/cap/y)	Average collection rate (%)	Inadequately managed waste (%)	Average plastics input (kg/cap/y)
OECD	2.2 (803)	11	88.3	90	0	8.8
AFR	0.65 (237)	13	30.8	45	70	26.6
MENA	1.1 (402)	9	36.1	80	30	15.9
EAP	0.95 (347)	13	45.1	70	30	23.0
ECA	1.1 (402)	8	32.1	75	30	15.2
LAC	1.1 (402)	12	48.2	75	30	22.9
SAR	0.45 (164)	7	11.5	65	60	8.5

Conversion of macroplastics into microplastics

Arising on land, macroplastics that were not collected and properly processed, can be distributed in the environment through different pathways. On land, these macroplastics can be converted into microplastics that can enter streams and rivers or become part of the soil (Sundt et al., 2014), or they can enter the aquatic environment directly and be converted there. Once in the surface waters, macroplastics that were not converted into microplastics will reach coastal areas and become part of the macroplastics load there. Eventually, these macroplastics will be converted into microplastics in the oceans and seas.

Only a small part of the microplastics formed on the land will run off into surface waters and therefore, we assumed this fraction to be negligible (Sundt et al., 2014). Macroplastics that end up in surface waters will undergo different processes that can result in microplastics. These time-dependent processes, like fragmentation, embrittlement, UV degradation and biodegradation, vary in time and place, depending on the characteristics of the

macroplastics and environmental conditions. Fragmentation can already take place after 4 weeks (e.g., polyurethane foam; (Andrady, 2017), but it can also take much longer.

For the conversion of macroplastics into microplastics in the ocean, Sundt et al. (2014) estimated an annual conversion rate of 1-5% of the total so called ‘standing mass’ of macroplastics in the ocean. Using this estimation, we assumed macroplastics conversion in rivers. We assumed a macroplastics mass present in rivers as a result of annual macroplastics input, that is divided over two riverine fractions (i.e., a ‘fast’ fraction and a ‘slow’ fraction). We estimated the amount of microplastics annually formed as 3% of this mass, taking into account the average residence time of both macroplastics fractions (for the fast fraction we used the residence time estimated for each river in van Wijnen et al. (2018), and for the slow fraction we assumed a residence time of 5 years). Further, we assumed that 50% of the poorly processed macroplastics ended up in the river, following Sundt et al. (2014).

Microplastics from tyre wear

One of the sources of microplastics discharged by WWTPs is the wear of care tyres, typically resulting in 60-80 µm particles of styrene-butadiene that end up in the sewerage system. We made a first estimation of the global input of microplastics from tyre wear using the average number of cars owned per capita in each region (data extracted from (Hoornweg and Bhada-Tata, 2012)) and the average car tyre wear assumption of Siegfried et al. (2017) for Europe (0.18 kg/cap/y) (Table S5.2).

Table S5.2.

Microplastic input as a result of car tyre wear.

Region	Number of motor vehicles/ 1000 cap	Microplastics Tyre wear (kg/cap/y) ¹
OECD	500	0.18
AFR	20	0.0072
MENA	190	0,068
EAP	50	0.018
ECA	213	0.077
LAC	115	0.041
SAR	20	0.0072

¹calculated proportional to OECD numbers

Microplastics from laundry fibers

Another source of microplastics discharged by WWTPs is fiber from clothing. The NOVA institute estimated the amount of microplastics originating from clothing being 1% of the synthetic fibers produced (Essel et al., 2015). In 2011, the FAO published a survey on global apparel fiber consumption (Shui and Plastina, 2013), in which they distinguished between natural fibers (e.g., cotton, wool, flax) and synthetic fibers (Table S3). In 2007, the worlds’ most consuming countries of synthetic fibers were: China, USA, India, Japan, The Russian Federation. Also Pakistan is in the top ten. Laundry fibers are collected in the sewerage

system before entering the environment. In this study we used the microplastics input estimated for 2005 (Table S5.3).

Table S5.3.

Microplastics input from synthetic fibers in clothing

Region	Synthetic fibers (kg/cap/y) 2005	Synthetic fibers (kg/cap/y) 2008
OECD	4.7	4.0
AFR	0.7	0.6
MENA	4.7	4.0
EAP	4.1	5.6
ECA	4.9	6.0
LAC	2.8	3.0
SAR ¹	3.6	4.6

¹In the FAO survey (Shui and Plastina, 2013) SAR is not an individual region, therefore we chose the average for developing countries.

Microplastics from personal care products (PCPs)

The most discussed sources of microplastics in rivers are personal care products (PCPs), e.g. , facial and body scrubs, toothpaste, shaving cream, peeling products, and make up, which contain plastic microbeads. Little is known about the global amount of these microbeads that ends up in rivers. For Europe, an estimate of 8 g/cap/y was made by Sundt et al. (2014). The NOVA institute (Essel et al., 2015) estimated the use of personal care products 6.25 g/cap/y for Germany. Therefore, Siegfried et al. (2017) used for Europe an estimate of 7.1 g/cap/y. To estimate the amount of microplastics from personal care products for the different global regions, we compared the sale of cosmetic products (in billion USD) from 2007 (Łopaciuk and Łoboda, 2013) with that of Europe and assumed a proportionate share of the 7.1 g/cap/y for that region (Table S5.4).

Table S5.4.

Microplastics input from personal care products

Region	Sale of cosmetic products (billion USD) 2007 ¹	Microbeads (kg/cap/y)
OECD	28.9	0.0055
AFR	3.6	0.0007
MENA	3.6	0.0007
EAP	25.9	0.0049
ECA	25.9	0.0049
LAC	12.9	0.0025
SAR	3.6	0.0007
EUR	37.3	0.0071 ²

¹ Łopaciuk and Łoboda (2013), ²Siegfried et al. (2017)

Removal of microplastics at waste water treatment plants

Microplastics that enter the environment by sewerage systems are partly removed in WWTPs. We estimated the average microplastics removal for each river basin using the average phosphorus removal for each river basin from Global NEWS (Van Drecht et al., 2009). Thus, we distinguished four classes of river basins with respect to microplastics removal at WWTPs (Table S5.5).

Table S5.5.

Estimated average microplastics removal at WWTPs in the river basins.

Riverbasin class	Description	Phosphate removal efficiency (GlobalNEWS¹)	Microplastics removal efficiency (estimated)
Class 1	Almost all of the WWTPs are equipped with primary and secondary treatment.	>70%	95%
Class 2	A lot of WWTPs have primary and secondary treatment	30 – 70 %	75%
Class 3	Most of the WWTPs have no secondary treatment and only (poor) primary treatment.	10 – 30%	50%
Class 4	On average, only few WWTPs exist. No extensive settling takes places.	<10%	0%

¹Mayorga et al. (2010), ²Van Drecht et al. (2009)

Summary

The quality of fresh water is of great importance for our society and the natural environment. Pollutants from agricultural, industrial and urban areas threaten the quality of surface waters including rivers, seas and oceans. New environmental issues are constantly arising, some of which are directly or indirectly influencing water quality. An example is the energy transition, aiming at the use of a more sustainable energy mix to lower carbon dioxide (CO₂) emissions. The energy transition may lead to an increased production of energy crops. This could, in turn, result in increased agricultural fertiliser use, leading to increased nutrient export by rivers and eutrophication of already vulnerable coastal areas. Moreover, increased nitrogen levels can make rivers important sources of nitrous oxide (N₂O). N₂O is a powerful greenhouse gas that contributes to climate change. Other water quality related environmental issues are those of contaminants of emerging concern (CECs) and of the growing amount of plastic pollution in the aquatic environment.

To manage water quality, monitoring programs have been set up to measure concentrations of contaminants in surface waters and to ensure safe drinking water. However, in many regions of the world, monitoring activities are scarce because of the costs of water sampling and analysis. As a result, the sources of pollution are often not well understood. For these regions water quality modelling offers a solution. Models can help to estimate pollution loads when environmental measurements are scarce. In addition, modelling can be used to simulate loads of emerging contaminants which are not included in ongoing monitoring programs, and to explore trends in water quality as a result of future developments. In the last decades, many water quality models have been developed, modelling various pollutants on different spatial and temporal scales. One of these models is GlobalNEWS, a global spatially explicit model that analyses the river export of nutrients to coastal seas in the past, present and future. This model calculates, in a steady state approach, annual average pollutant loads for over 6000 river basins, and is therefore suitable for analysing global trends.

This thesis aims to explore the possibilities to expand GlobalNEWS to address the environmental impact of new water pollution challenges like those triggered by large scale biofuel production, contaminants of emerging concern and microplastics. To this end, GlobalNEWS was adapted in different ways: (1) by developing new scenarios, i.e., for large-scale production of energy crops, (2) by including a new environmental compartment in the model, i.e., to account for nitrous oxide emissions to the atmosphere, and (3) by including process formulations in the model for new substances, such as triclosan and microplastics.

These goals were elaborated in four case studies, each with their own objectives:

- To explore possible effects of large-scale biodiesel production from energy crops on coastal eutrophication in European seas through enhanced nutrient losses from agricultural land to rivers in the year 2050.

- To quantify future N₂O emissions from European river basins that are associated with the cultivation of energy crops.
- To quantify future trends in global river export of triclosan from personal care products to coastal seas.
- To contribute to a better understanding of river export of microplastics from land to sea and to explore trends in global river export of microplastics for three future scenarios (year 2050), that differ in assumed levels of environmental control.

The possibilities to expand GlobalNEWS were evaluated based on the findings from the case studies, that are described in Chapters 2-5 of this thesis. The findings are summarised in the following paragraphs.

The implementation of the Millennium Ecosystem Assessment scenarios makes it possible to analyse future nutrient export to coastal areas with GlobalNEWS. These scenarios, however, do not explicitly take into account new developments, such as the large-scale cultivation of energy crops. This was the focus of the first case study, in which the possible effects of increased biodiesel use on coastal eutrophication in Europe in the year 2050 were explored. The large-scale production of biofuels from energy crops is expected to lead to an increase in the use of synthetic fertilisers and an increased nutrient export by rivers. Six illustrative scenarios in which the biodiesel production increases to about 10 to 30% of the diesel use in 2010 were defined. As a baseline for the scenarios, an existing Millennium Ecosystem Assessment scenario for 2050, i.e., Global Orchestration, was used. The scenarios differed with respect to the assumptions on where the energy crops were cultivated: either on land that is currently used for agriculture, or on land used for other purposes. The scenarios were analysed with the GlobalNEWS model for nitrogen (N) and phosphorus (P). GlobalNEWS distinguishes inorganic and organic forms from these nutrients. In the baseline scenario the dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphorus (DIP) export by rivers to European coastal waters decreases between 2000 and 2050 by about 5% (DIN) and 15% (DIP) as a result of environmental and agricultural policies. In the six scenarios with increased biodiesel production, the DIN export increases by about 20–35% and the DIP export by about 10–20% compared to the baseline scenario, thus counteracting the decrease in the baseline scenario. Spatial patterns of nutrient export showed large differences between the different European regions. Largest increases in nutrient export were predicted for the Mediterranean Sea and the Black Sea.

This case study shows that large scale biofuel production in the European Union is not without dispute. The results indicate that it is difficult to produce considerable amounts of biodiesel without affecting nature and food and feedstock production. As a result of increased nutrient export, large-scale cultivation of energy crops may lead to eutrophication of the European coastal waters, especially in the Mediterranean Sea and the Black Sea.

The second case study focussed on the formation of N₂O as a result of growing first generation energy crops for biodiesel production in Europe. Biodiesel is considered a fuel

with a low greenhouse gas potential, since the amount of CO₂ that is released as a result of combustion of biofuels is compensated by the uptake of CO₂ by the crop previously. However, for the production of biodiesel various processes are responsible for additional CO₂ emissions, which negatively influences the greenhouse gas balance. In addition, many synthetic nitrogen-containing fertilisers are used for the cultivation of energy crops, which can be partially converted into N₂O, which has a large global warming potential, about 298 times larger than CO₂. Fertiliser use not only increases the direct agricultural soil emissions, but also the indirect N₂O emissions from aquatic systems, after leaching and runoff of nitrogen from fertilised soils. This second case study quantified future N₂O emissions associated with the cultivation of energy crops in European river basins. To this end, three future scenarios for biodiesel production in Europe, also used in the first case study, were analysed using Global NEWS and the IPCC guidelines for N₂O emission factors, for both direct and indirect N₂O emissions.

The results of this case study indicate that increased biodiesel production may increase N₂O emissions in Europe by about 25-45% relative to the baseline scenario without a growth in biodiesel production. In 2050 total N₂O emissions from European river basins as result of energy crop growing may be as large as 220 – 260 Gg N₂O/y.

Pollutants other than nutrients are also transported by rivers to the sea. Therefore, a first step to expand GlobalNEWS beyond nutrients was taken by developing GlobalTCS. GlobalTCS is a global, spatially explicit model, that calculates river export of triclosan, an antibacterial agent. Triclosan is, for instance, added to commonly used personal care products. In GlobalTCS, triclosan is assumed to enter the aquatic environment through sewage effluent, which simplifies the modelling considerably. With this model annual, basin-wide triclosan export for the year 2000 and two future scenarios for the year 2050 (the Millennium Ecosystem Assessment scenarios Global Orchestration and Adapting Mosaic) were analysed, based on assumptions about worldwide triclosan use.

The analyses for 2000 indicate that triclosan export to coastal areas in Western Europe, Southeast Asia and the east coast of the USA is higher than in the rest of the world. For the future scenarios, the GlobalTCS model predicts an increase in river export of triclosan in Southeast Asia as a result of fast population growth, increasing urbanisation and increasing numbers of people connected to sewerage systems with poor waste water treatment.

In the last case study, the Global Riverine Export of Microplastics into Seas (GREMiS) model was developed; a global spatially explicit model for analysing annual microplastics export to coastal seas. Like GlobalTCS, GREMiS is based on GlobalNEWS. Microplastics are plastic particles with a typical size between 1 µm and 5 mm, which may vary in composition. Important sources of microplastics in the aquatic environment are badly managed plastic waste, e.g., municipal and agricultural plastic waste, car tyres, synthetic laundry fibres and personal care products. Microplastics can be intentionally put into a product (primary

microplastics), for example in personal care products or abrasives, or be formed by fragmentation and degradation of larger plastic items (secondary microplastics). They are distributed in the environment in various ways. Microplastics of personal care products, laundry fibres and a large part of the car tyre wear enter the aquatic environment via sewage, while the microplastics that result from fragmentation of larger plastic items are spread in a diffuse way. This study focused on river export of microplastics from land to sea and on exploring trends in global river export of microplastics for three future scenarios (year 2050) that differ in the assumed level of environmental control. GREMiS was used to locate hotspots and to identify the most important sources and to predict future trends of microplastics export. For this, four different sources of microplastics were included, i.e., personal care products, synthetic laundry fibres, car tyre wear and mismanaged municipal plastic waste. The input of these sources was estimated for seven different world regions.

The results indicate that river export of microplastics varies among world regions, with hotspots in South East Asia and South America. The 'Business as usual' scenario for 2050 (BAU) assumes an increasing world population, GDP and sewage treatment, but no specific measures to reduce plastic in the environment. As a result, the global microplastics export may increase by 50% compared with the recent past. Improved waste management and removal of microplastics in waste water treatment plants, as assumed in the 'Environment profits' scenario (ENV) may halve the riverine microplastics export. Globally, fragmentation of macroplastics is modeled to be the main source of microplastics in rivers. About 20% of the total microplastics river export is from discharges of sewerage systems carrying microplastics from car tyre wear, laundry fibers and personal care products. This percentage varies among regions, ranging from 1% in Africa to 60% in the OECD countries. In the BAU scenario, the share of these sewerage discharges globally decreases to 10% (varying among regions, from 2 to 32%) by 2050 as a result of improved sanitation and sewerage connection. In the ENV scenario, the share of sewerage discharges decreases a little further in most regions.

Summarising, in this thesis GlobalNEWS has been successfully extended, covering new scenarios, a new compartment, i.e., atmosphere, and new pollutants, i.e., triclosan and microplastics.

The Millennium Ecosystem Assessment scenarios, implemented in GlobalNEWS, could be used as a basis for new scenarios, to analyse nutrient export by rivers as a result of new environmental issues in the future. The study on first generation energy crops in Europe illustrated this. The introduction of new scenarios in GlobalNEWS was done by extending existing scenarios. To calculate a new scenario with GlobalNEWS, new input data were required, while the modelled processes remained unchanged. Inclusion of a new compartment in the model, as is described in the second case study, was achieved by a relatively simple addition to the model: the fate processes in that compartment were not explicitly modelled, only the emission to that compartment. Extending GlobalNEWS to

simulate river export of micro-pollutants turned out to be more complex, and required the development and implementation of new model equations.

Using one modelling approach to study multiple pollution problems has several advantages, such as an improved consistency (e.g., in global hydrology), the potential to study trade-offs between different problems and easier communication with external parties. GlobalNEWS can be compared with other large scale water quality models for nutrients, pathogens, and plastics. All models have their own typical strengths and weaknesses, and very few examples exist of modelling nutrients, organic pollutants and microplastics simultaneously. Some models have the advantage of modelling processes like degradation and sedimentation in a dynamic way, but this comes at the cost of increasing complexity, data need and computation time. The choice for a model depends on the purpose of the intended study and considerations like the desired level of accuracy and the available input data. The main advantage of GlobalNEWS is its limited complexity and need for input data which makes it relatively easy to apply and especially useful to simulate global trends in future scenarios. As such, the extension of GlobalNEWS is a good step forward in the development of a global platform to explore future trends in water quality.

Water is one of the main topics of the 2030 Agenda of Sustainable Development Goals (SDGs). Each SDG has a specific objective, such as poverty, inequality, environment and human rights, health and peace. There are two SDGs on water issues, i.e., SDG 6, 'Clean water and sanitation' and SDG 14, 'Life below water'. Concerning SDG 6, the UN is committed to enhanced monitoring and to promoting 'good government' practices to improve water quality. Modelling could be a useful addition to these measures, especially when predicting future scenarios and to prevent passing on environmental problems from one SDG to another. Multi-pollutant modelling will become increasingly relevant to deal with emerging water quality issues. This thesis shows that global modelling of different pollutants is possible with a relatively simple modelling approach. In this way it contributes to the development of water quality modelling, so that, hopefully, future water issues can be prevented and resolved better.

Samenvatting

Water is belangrijk voor het milieu en onze samenleving. De kwaliteit van water in rivieren, zeeën en oceanen wordt bedreigd door verontreinigen afkomstig van landbouw, industrie en stedelijke gebieden. In onze samenleving ontstaan voortdurend nieuwe milieuproblemen, waarvan sommige direct of indirect betrekking hebben op de waterkwaliteit. Voorbeelden hiervan zijn het gebruik van biobrandstoffen als alternatief voor fossiele brandstoffen, opkomende stoffen (contaminants of emerging concern, CECs) en (micro)plastics in het aquatisch milieu.

Om de waterkwaliteit te garanderen zijn monitoringprogramma's opgezet waarin concentraties van verontreinigingen in het oppervlaktewater worden gemeten. Wereldwijd wordt monitoring helaas nog weinig toegepast, vanwege de kosten van waterbemonstering en –analyse. Het gebruik van modellen kan in dit geval uitkomst bieden. Ook voor opkomende stoffen, die niet zijn opgenomen in lopende monitoringprogramma's en om trends in waterkwaliteit als gevolg van toekomstige ontwikkelingen te voorspellen, kunnen modellen worden gebruikt. In de afgelopen decennia zijn veel waterkwaliteitsmodellen ontwikkeld die geschikt zijn om verschillende verontreinigende stoffen op verschillende ruimtelijke en temporele schaal te modelleren. Een van deze modellen is GlobalNEWS, een ruimtelijk expliciet model dat opereert op mondiale schaal en waarmee riviertransport van nutriënten naar kustzeeën in het verleden, heden en toekomst geanalyseerd kan worden. Dit steady-state model berekent de jaarlijks gemiddelde vracht aan nutriënten voor meer dan 6000 stroomgebieden en is geschikt voor het analyseren van wereldwijde trends.

Doel van het onderzoek in dit proefschrift is het verkennen van de mogelijkheden om GlobalNEWS zo aan te passen dat het model kan worden gebruikt om de milieu-impact van nieuwe uitdagingen op het gebied van watervervuiling in kaart te brengen, bijvoorbeeld veroorzaakt door grootschalige productie van biobrandstoffen, opkomende stoffen en microplastics. Om dit te bereiken werd GlobalNEWS op verschillende manieren aangepast: 1) door nieuwe scenario's te ontwikkelen, onder andere voor grootschalige productie van energiegewassen, 2) door een nieuw milieucompartiment in het model op te nemen, waardoor rekening gehouden kan worden met lachgas (N_2O) emissies naar de atmosfeer, en 3) door nieuwe procesformuleringen op te nemen voor opkomende stoffen, zoals triclosan en microplastics.

Deze aanpassingen werden uitgewerkt in vier case studies, elk met specifieke doelstellingen:

- Het verkennen van de mogelijke effecten van grootschalige biodieselproductie uit energiegewassen op eutrofiëring van Europese kustzeeën in het jaar 2050, als gevolg van de verhoogde emissie van nutriënten van landbouwgronden naar rivieren.
- Het kwantificeren van toekomstige N_2O -emissies van Europese stroomgebieden als gevolg van grootschalige teelt van energiegewassen.
- Het kwantificeren van toekomstige trends in het wereldwijde riviertransport van triclosan uit persoonlijke verzorgingsproducten naar kustzeeën.
- Het bijdragen aan een beter begrip van het riviertransport van microplastics van land naar zee en het verkennen van trends in het mondiale riviertransport van microplastics voor drie scenario's voor 2050.

Op grond van de conclusies van de case studies, die in de hoofdstukken 2-5 van dit proefschrift worden beschreven, worden in hoofdstuk 6 de mogelijkheden om GlobalNEWS uit te breiden geëvalueerd. Die conclusies worden hieronder samengevat.

De implementatie van de Millennium Ecosystem Assessment-scenario's in GlobalNEWS maakt dit model geschikt om toekomstige export van nutriënten naar kustgebieden te analyseren. Deze scenario's houden echter niet expliciet rekening met nieuwe ontwikkelingen, zoals de grootschalige teelt van energiegewassen. In de eerste case study werden daarom de mogelijke effecten van toenemend biodieselgebruik op eutrofiëring van Europese kustwateren in het jaar 2050 onderzocht. Grootschalige productie van biobrandstoffen uit energiegewassen zal naar verwachting leiden tot een toename van het gebruik van synthetische meststoffen en een verhoogde export van nutriënten door rivieren. Er zijn zes illustratieve scenario's ontworpen waarin een biodieselproductie van 10 tot 30% van het dieselgebruik in 2010 kan worden gerealiseerd. Als basis voor de scenario's is een bestaand Millennium Ecosystem Assessment-scenario voor 2050 (Global Orchestration) gebruikt. De scenario's verschillen met betrekking tot de aannames over waar de energiegewassen worden verbouwd: op bestaande landbouwgrond of op land dat nu nog wordt gebruikt voor andere doeleinden. De emissiescenario's voor stikstof (N) en fosfor (P) zijn met GlobalNEWS geanalyseerd. GlobalNEWS onderscheidt anorganische en organische vormen van deze nutriënten. In het basisscenario daalt de export van opgelost anorganisch stikstof (DIN) en opgelost anorganisch fosfor (DIP) naar Europese kustwateren tussen 2000 en 2050 met ongeveer 5% (DIN) en 15% (DIP) als gevolg van milieu- en agrarisch beleid. In de zes scenario's met een verhoogde productie van biodiesel neemt de DIN-export toe met ongeveer 20-35% en de DIP-export met ongeveer 10-20% vergeleken met het basisscenario, waardoor de daling in het basisscenario meer dan gecompenseerd wordt. Verschillende Europese regio's laten grote verschillen in nutriëntenexport zien. De grootste toename wordt voorspeld voor de Middellandse Zee en de Zwarte Zee.

Deze case study laat zien dat grootschalige productie van biobrandstoffen in de Europese Unie niet onomstreden is. Het blijkt moeilijk om aanzienlijke hoeveelheden biodiesel te produceren zonder de natuur en voedsel- en grondstofproductie (negatief) te beïnvloeden. Als gevolg van de toegenomen export van nutriënten kan grootschalige teelt van energiegewassen leiden tot eutrofiëring van de Europese kustwateren.

De tweede case study richtte zich op de vorming van N_2O als gevolg van de teelt van eerste generatie energiegewassen voor de productie van biodiesel in Europa. Biodiesel wordt beschouwd als een brandstof die relatief weinig bijdraagt aan het broeikaseffect, omdat de hoeveelheid koolstofdioxide (CO_2) die vrijkomt als gevolg van de verbranding van biobrandstoffen wordt gecompenseerd door de opname van CO_2 die nodig is voor de groei van het gewas. Bij de productie van biodiesel zijn echter verschillende processen verantwoordelijk voor extra CO_2 -uitstoot, waardoor de broeikasgasbalans negatief wordt beïnvloed. Bovendien wordt veel stikstofhoudende kunstmest gebruikt voor de teelt van energiegewassen, die gedeeltelijk wordt omgezet in N_2O , een gas met een hogere broeikaspotentie dan die van CO_2 (265- 298 keer zo groot). Het gebruik van kunstmest verhoogt niet alleen de directe N_2O -emissies van landbouwgrond, maar ook de indirecte

N₂O-emissies van watersystemen, na uitspoeling en afvoer van stikstof uit bemeste bodems. In deze tweede case study zijn toekomstige N₂O-emissies gekwantificeerd die verband houden met de teelt van energiegewassen in Europese stroomgebieden. Daartoe werden drie toekomstscenario's voor de productie van biodiesel in Europa, ook gebruikt in de eerste case study, geanalyseerd met behulp van Global NEWS en de IPCC-richtlijnen voor N₂O-emissiefactoren, voor zowel directe als indirecte N₂O-emissies.

De resultaten van deze case study laten zien dat bij verhoogde biodieselproductie de N₂O-uitstoot in Europa met ongeveer 25-45% kan toenemen ten opzichte van het basisscenario zonder groei van de biodieselproductie. In 2050 kan de totale N₂O-uitstoot van Europese stroomgebieden als gevolg van de teelt van energiegewassen oplopen tot 220 - 260 Gg N₂O/jaar.

Naast nutriënten worden vele andere verontreinigende stoffen via rivieren naar de zee getransporteerd. Daarom werd in de derde case study het GlobalTCS model ontwikkeld; een mondiaal, ruimtelijk expliciet model dat is gebaseerd op GlobalNEWS en rivierexport van triclosan berekent. Triclosan is een stof met een antibacteriële werking die bijvoorbeeld wordt toegevoegd aan veelgebruikte producten voor persoonlijke verzorging, zoals zeep en tandpasta. In GlobalTCS wordt ervan uitgegaan dat triclosan via rioolwater in het aquatisch milieu terecht komt, wat de modellering aanzienlijk vereenvoudigt. GlobalTCS werd gebruikt om de triclosan-export voor het jaar 2000 en twee toekomstscenario's voor het jaar 2050 (de Millennium Ecosystem Assessment-scenario's Global Orchestration en Adapting Mosaic) te analyseren.

Uit de analyses voor 2000 blijkt dat de export van triclosan naar kustgebieden in West-Europa, Zuidoost-Azië en de oostkust van de VS hoger is dan in de rest van de wereld. Voor de toekomstscenario's voorspelt GlobalTCS een toename van de triclosan-export in Zuidoost-Azië als gevolg van snelle bevolkingsgroei, toenemende urbanisatie en een toenemend aantal mensen dat is aangesloten op rioleringsystemen, die nog niet in een goede afvalwaterzuivering voorzien.

In de laatste case study is het Global Riverine Export of Microplastics into Seas (GREMiS) model ontwikkeld; een globaal ruimtelijk expliciet model voor de analyse van de jaarlijkse export van microplastics naar kustwateren dat net als GlobalTCS is gebaseerd op GlobalNEWS. Belangrijke bronnen van microplastics in het aquatisch milieu zijn plasticafval, bijvoorbeeld huishoudelijk en agrarisch plastic afval, autobanden, synthetische vezels van kleding en producten voor persoonlijke verzorging. Microplastics kunnen opzettelijk aan een product worden toegevoegd (primaire microplastics), bijvoorbeeld in producten voor persoonlijke verzorging of schuurmiddelen, of ze worden gevormd door fragmentatie en degradatie van grotere plastic voorwerpen (secundaire microplastics). Ze worden op verschillende manieren in de omgeving verspreid. Microplastics uit producten voor persoonlijke verzorging, kledingvezels en bandenslijtstof komen vooral via riolering in het aquatisch milieu, terwijl de microplastics die het gevolg zijn van fragmentatie van grotere plastic voorwerpen diffuus worden verspreid. Deze case study had als doel de trends in de export van microplastics van land naar zee voor drie toekomstscenario's (jaar 2050) te verkennen. GREMiS werd gebruikt om hotspots voor de export van microplastics en de

belangrijkste bronnen van microplastics te identificeren. Er werden vier verschillende bronnen van microplastics beschouwd: producten voor persoonlijke verzorging, synthetische kledingvezels, slijtage van autobanden en stedelijk plastic afval. De input van deze bronnen werd geschat voor zeven verschillende wereldregio's.

Uit de resultaten blijkt dat de export van microplastics varieert per wereldregio, met hotspots in Zuidoost-Azië en Zuid-Amerika. Het scenario 'Business as usual' voor 2050 (BAU) gaat uit van een toenemende wereldbevolking, bruto nationaal product en rioolwaterzuivering, maar geen specifieke maatregelen om plastic in het milieu te verminderen. Als gevolg hiervan kan de mondiale export van microplastics in 2050 met 50% toenemen. Een betere inzameling en verwerking van afval en een betere verwijdering van microplastics in afvalwaterzuiveringsinstallaties, zoals aangenomen in het 'Environment profit' scenario (ENV), kan de export van microplastics halveren. Fragmentatie van macroplastics is volgens GREMiS wereldwijd de belangrijkste bron van microplastics in rivieren. Ongeveer 20% van de totale export van microplastics is afkomstig van slijtage van autobanden en kledingvezels die via het riool op het oppervlaktewater worden geloosd (en slechts 1% van producten voor persoonlijke verzorging). Dit percentage varieert per regio, van 1% in Afrika tot 60% in de landen aangesloten bij de OECD. In het BAU-scenario daalt in 2050 het aandeel van deze lozingen wereldwijd tot 10% (variërend per regio, van 2 tot 32%) als gevolg van verbeterde sanitaire voorzieningen en het aansluiten van een groter deel van de bevolking op het riool. In het ENV-scenario daalt het aandeel van deze lozingen in de meeste regio's nog verder.

De case studies in hoofdstuk 2-5 laten zien hoe GlobalNEWS kan worden uitgebreid met nieuwe scenario's, een nieuw compartiment (de atmosfeer), en voor nieuwe verontreinigende stoffen (triclosan en microplastics). De case study naar energiegewassen in Europa illustreert hoe de Millennium Ecosystem Assessment-scenario's die in GlobalNEWS zijn geïmplementeerd als uitgangspunt kunnen worden gebruikt voor de ontwikkeling van nieuwe scenario's om de export van nutriënten door rivieren als gevolg van toekomstige ontwikkelingen te analyseren. Voor het berekenen van een nieuw scenario in GlobalNEWS zijn alleen nieuwe invoergegevens vereist; de gemodelleerde processen kunnen ongewijzigd blijven. Opname van een nieuw compartiment in het model, zoals beschreven in de tweede case study, werd bereikt door een relatief eenvoudige toevoeging van de emissie naar het compartiment. De processen binnen het compartiment werden niet expliciet gemodelleerd. Uitbreiding van GlobalNEWS om export van andere milieuverontreinigende stoffen te simuleren blijkt complexer en vereist de ontwikkeling en implementatie van nieuwe modelvergelijkingen.

Het gebruik van één modelbenadering om meerdere verontreinigingsproblemen te bestuderen heeft verschillende voordelen, zoals een betere samenhang (bijvoorbeeld in de wereldwijde hydrologie), de mogelijkheid om afwenteling tussen verschillende problemen tegen te gaan en een makkelijkere communicatie met externe partijen. Naast GlobalNEWS zijn er vele andere mondiale waterkwaliteitsmodellen voor voedingsstoffen, ziekteverwekkers en kunststoffen. Al deze modellen hebben hun eigen sterke en zwakke punten, en er zijn maar weinig voorbeelden van het tegelijkertijd modelleren van

voedingsstoffen, organische verontreinigende stoffen en microplastics. Sommige modellen hebben het voordeel dat processen als degradatie en sedimentatie op een dynamische manier zijn geïmplementeerd, maar dit gaat gepaard met een toenemende complexiteit, behoefte aan gegevens en rekentijd. De keuze voor een bepaald model hangt af van de beoogde doelen en van overwegingen zoals de gewenste mate van detail en de beschikbare invoergegevens. Het belangrijkste voordeel van GlobalNEWS is de beperkte complexiteit en behoefte aan invoerdata, waardoor het model relatief eenvoudig kan worden gebruikt. Dit maakt de uitbreiding van GlobalNEWS een goede stap voorwaarts bij het ontwikkelen van een platform om toekomstige mondiale trends in waterkwaliteit te verkennen.

Water is een van de belangrijkste onderwerpen op de 2030 Agenda van de Sustainable Development Goals (SDG's). De VN zet bij SDG 6, 'Schoon water en sanitair', in op verbeterde monitoring en op het bevorderen van 'good government' om de waterkwaliteit te verbeteren. Modelleren kan een nuttige aanvulling zijn op deze maatregelen, vooral bij het voorspellen van toekomstige scenario's en om te voorkomen dat milieuproblemen van de ene SDG op de andere worden afgewenteld. Het tegelijkertijd modelleren van meerdere verontreinigende stoffen zal steeds relevanter worden voor het oplossen van nieuwe problemen met de waterkwaliteit. Dit proefschrift laat zien dat globale modellering van verschillende verontreinigende stoffen mogelijk is met een relatief eenvoudige modelleringsbenadering. Op deze manier draagt het bij aan de ontwikkeling van waterkwaliteitsmodellering, waarmee toekomstige waterproblemen wellicht kunnen worden voorkomen.

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Over de auteur

Jikke van Wijnen werd op 26 december 1961 geboren in Heiloo en groeide op in en om Amsterdam. Na haar middelbare schooltijd op het Stedelijk Gymnasium in Arnhem startte ze in 1980 met een studie scheikunde aan de Universiteit van Amsterdam. In 1986 studeerde ze af met als hoofdvak 'Moleculaire biologie' en als bijvak 'Milieu- en toxicologische chemie'. Na de studie werkte ze van 1986-1989 als docent scheikunde op het Murmellius Gymnasium in Alkmaar. Daarnaast werkte ze vanaf 1987 als studiebegeleidster Natuurwetenschappen op het Studiecentrum Amsterdam van de Open Universiteit. Na drie jaar voor de klas te hebben gestaan, keerde ze terug naar de vakgroep Milieu-en toxicologische chemie van de UvA, waar ze van 1989-2003 onderzoek deed naar de opname van dioxines en dibenzofuranen door zoogdieren. Vanaf 2003 werkt ze alleen nog voor de Open Universiteit, waar ze verantwoordelijk is voor de scheikunde vakken en het geïntegreerd natuurwetenschappelijk practicum en de begeleiding van verschillende andere cursussen van de faculteit natuurwetenschappen en –tot 2019- de faculteit informatica.

Vanaf 2011 maakt ze onderdeel uit van het onderzoeksgroepje dat zich, onder leiding van prof. Carolien Kroeze, bezig houdt met het modelleren van het transport van nutriënten door rivieren naar kustgebieden met behulp van GlobalNEWS. Dit geeft haar de mogelijkheid om in 2013 een promotieonderzoek te beginnen, met Carolien Kroeze als promotor. Als het onderzoek zich gaat richten op het rivier transport van andere stoffen, zoals triclosan en microplastics, sluit prof. Ad Ragas zich, als tweede promotor, bij het onderzoek aan. In 2019 leidt dit tot dit proefschrift met de titel 'River export of pollutants: A global modelling approach'.

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