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Towards spatially smart abatement of human pharmaceuticals in surface waters: Defining impact of sewage treatment plants on susceptible functions

Lieke J.C. Coppens [a, b, *], Jos A.G. van Gils [c], Thomas L. ter Laak [a, d], Bernard W. Raterman [a], Annemarie P. van Wezel [a, b]

[a] KWR Watercycle Research Institute, P.O. Box 1072, 3430 BB, Nieuwegein, The Netherlands
[b] Utrecht University, Copernicus Institute of Sustainable Development, P.O. Box 80.115, 3508 TC, Utrecht, The Netherlands
[c] Deltares, P.O. Box 177, 2600 MH, Delft, The Netherlands
[d] Wageningen University, Sub-department Environmental Technology, P.O. Box 17, 6700 AA, Wageningen, The Netherlands

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ABSTRACT

For human pharmaceuticals, sewage treatment plants (STPs) are a major point of entry to surface waters. The receiving waters provide vital functions. Modeling the impact of STPs on susceptible functions of the surface water system allows for a spatially smart implementation of abatement options at, or in the service area of, STPs.

This study was performed on a nation-wide scale for the Netherlands. Point source emissions included were 345 Dutch STPs and nine rivers from neighboring countries. The Dutch surface waters were represented by 2511 surface water units. Modeling was performed for two extreme discharge conditions. Monitoring data of 7 locations along the rivers Rhine and Meuse fall mostly within the range of modeled concentrations. Half of the abstracted volumes of raw water for drinking water production, and a quarter of the Natura 2000 areas (European Union nature protection areas) hosted by the surface waters, are influenced by STPs at low discharge. The vast majority of the total impact of all Dutch STPs during both discharge conditions can be attributed to only 19% of the STPs with regard to the drinking water function, and to 39% of the STPs with regard to the Natura 2000 function.

Attributing water treatment technologies to STPs as one of the possible measures to improve water quality and protect susceptible functions can be done in a spatially smart and cost-effective way, using consumption-based detailed hydrological and water quality modeling.

1. Introduction

1.1. Pharmaceuticals in the water cycle

Human pharmaceuticals and their transformation products are ubiquitously present in the water cycle. They are found in sewage effluents, surface waters and drinking water (Benotti et al., 2009; Daughton and Ternes, 1999; De Jongh et al., 2012; Houtman et al., 2014; Mompelat et al., 2009; Monteiro and Boxall, 2010; Ter Laak et al., 2010; Verlicchi et al., 2012). The majority of produced pharmaceuticals are consumed, partly metabolized and excreted into the sewage system. Sewage treatment plants (STPs) are a major point of entry to surface waters (Michael et al., 2013). Depending on their physico-chemical properties, up to 70% of the consumed medicines end up in the surface water (Ter Laak et al., 2010).

Surface waters provide vital functions to humans such as drinking water, nature, recreation and food production. These functions are susceptible to quality degradation by anthropogenic contaminants and are defined here as “susceptible functions”. Water quality regarding emerging substances has been monitored widely (e.g. Loos et al., 2013, 2010, 2009; Monteiro and Boxall, 2010). Modeling efforts are somewhat more scarce (Aldekoa et al., 2013; Alder et al., 2010; Johnson et al., 2013; Ort et al., 2009; Pistocchi et al., 2012; Ter Laak et al., 2010; Verlicchi et al., 2014). As concentrations of individual compounds and their derivatives are relatively low in drinking water and its sources (ng/L to

* Corresponding author. Nelen & Schuurmans, P.O. Box 1219, 3500 BE, Utrecht, The Netherlands.
E-mail address: lieke.coppens@gmail.com (L.J.C. Coppens).

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μg/L), for individual compounds or simple mixtures of known compounds adverse human health effects are not expected (Bruce et al., 2010; De Jongh et al., 2012; Houtman et al., 2014; Schriks et al., 2010). Adverse health effects on organisms in the ecosystem are considered more likely (Blair et al., 2013; Brodin et al., 2013; Corcoran et al., 2010; Escher et al., 2011). Concerns remain on the human and ecosystem health relevance of the total complex mixtures occurring in the environment, in which the pharmaceuticals and their transformation products are part (Bergman et al., 2013; Diamond et al., 2015; Kortenkamp et al., 2007; Malaj et al., 2014).

1.2. Abatement options

Abatement strategies are various types of interventions with the objective to reduce the chemical load of water systems, thus reducing exposure and adverse effects on man and environment. Several reviews address the challenges regarding emission reduction of micropollutants, or pharmaceuticals in particular (Eggen et al., 2014; Schwarzenbach et al., 2006). Options to abate adverse environmental and human health impacts can be taken during the whole chemical life cycle, i.e. design, market authorization, production, prescription, use, disposal and ultimately technological interventions in the water cycle at point-of-use, point of environmental entry or at the susceptible function (Khetan and Collins, 2007; Le Corre et al., 2012; Schirmer and Schirmer, 2008).

STPs are generally not designed to remove pharmaceuticals or other emerging chemicals. Depending on the physico-chemical properties of the pharmaceutical and the exact treatment process in a traditional STP, pharmaceuticals' removal efficiencies vary widely from <24% to 99% (Gros et al., 2010; Jelic et al., 2011). Emission reduction of pharmaceuticals is not required according to EU legislation, and environmental water or drinking water quality standards do not exist for these compounds. However, many European drinking water and wastewater utilities invest in advanced water treatment technology to increase removal efficiencies, e.g. by advanced oxidation, activated carbon filters or membrane reactors. In Switzerland for example, the implementation of additional wastewater treatment techniques at selected STPs is considered to be scientifically, technically, socially and financially feasible (Eggen et al., 2014).

Much research focuses on improved understanding of removal efficiencies and on further development of water treatment technologies (Delgado et al., 2012; Lee et al., 2013; Sudhakaran and Amy, 2013). Yet, advanced treatment at the scale of wastewater volumes is relatively costly. Depending on STP size and chosen treatment, costs for advanced treatment mentioned in literature vary widely from 0.64 to 4.00 €/m^3. In Switzerland average costs are expected to increase with 10–50% compared to the current costs for conventional treatment, after upgrading with activated carbon and ozonation. Costs per m^3 drop with increasing size of STPs and further technology development (Eggen et al., 2014; Jones et al., 2007).

Limited attention is paid to smart placement of these technologies within the water system in relation to susceptible functions. In the Netherlands there is much discussion on if, how and where abatement measures would be desirable, and how the option of additional water treatment technology relates to other possible abatement options in the chemical life cycle.

1.3. Aim and scope

The aim is to model the impact of STPs on susceptible functions of the surface water system on a nation-wide scale. This allows for a spatially smart implementation of abatement options at or in the service area of STPs. Because data on use, emission and occurrence of human pharmaceuticals are relatively abundant compared to other emerging substances, pharmaceuticals are useful study compounds and are considered as illustrative for other emerging substances for which STPs are the major route of environmental entry.

We performed a detailed spatial analysis of STP emissions of two pharmaceuticals with different characteristics to the Dutch surface water system. Concentrations of the pharmaceuticals as a result of consumption-based emissions from STPs and rivers from neighboring countries were modeled with a detailed hydrology and water quality model for two extreme discharge conditions. We identified surface waters with a susceptible function, i.e. drinking water abstractions and Natura 2000 areas (European Union nature protection areas).[1] For drinking water abstraction, groundwater abstractions were included. The water quality model provides a quantitative relationship between STP emissions and their impact on susceptible functions of the water system, giving insight in high-impact STPs where placement of abatement options to reduce concentrations of pharmaceuticals will deliver most benefits.

Table 1
Characteristics of the model pharmaceuticals.

	Carbamazepine	Ibuprofen
M, Dutch sales (kg/y)	8,400[a]	28,884[a]
f_e (fraction excreted)	0.26[b]	0.3[c]
f_{STP} (fraction passing STP)	0.91[d]	0.26[d]
half-life times EPI Suite(d)	37.5[e]	15[e]
half-life times in winter at latitude 50° N (d)	450[f]	–
half-life times in summer at latitude 50° N (d)	100[f]	–
log K_{ow}	2.25[e]	3.79[e]
pK_a	n.a.	4.47[g]

[a] Van der Aa et al., 2011.
[b] Besse et al., 2008.
[c] Lienert et al., 2007.
[d] Miege et al., 2009.
[e] EPI Suite Level III Fugacity Model (half-life time in water).
[f] Andreozzi et al., 2003 (interseasonal differences in photodegradation).
[g] Verliefde et al., 2008.

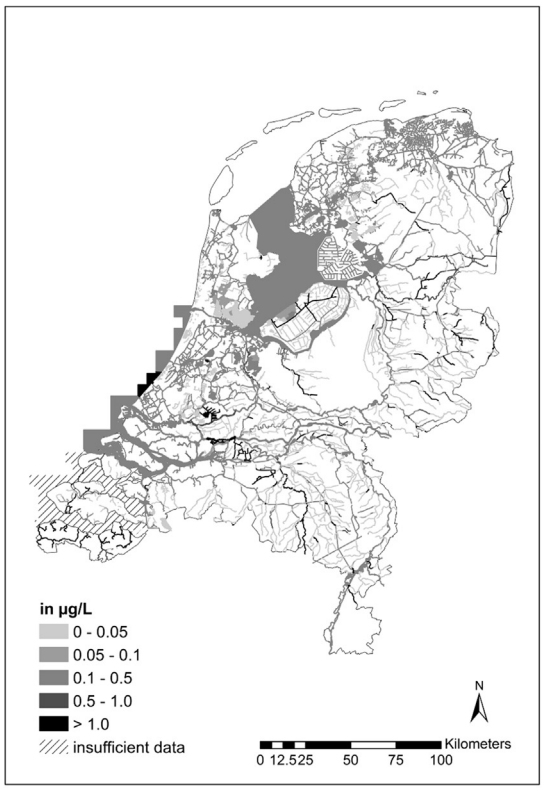
2. Methods

2.1. Modeling surface water concentrations

A water quality model representing the Dutch surface water network and its key hydrology features and a consumption-based emission model were combined to predict how loads from both STPs and inflowing rivers are spread over the Dutch surface water network during low and high river discharges. We studied two pharmaceuticals with different characteristics: the relatively persistent anti-epileptic carbamazepine (Andreozzi et al., 2003) and the relatively unstable anti-inflammatory pharmaceutical ibuprofen (Richardson and Bowron, 1985). Regardless their effect on organisms, these two pharmaceuticals were considered exemplary compounds for other persistent or degradable (emerging) substances that know similar use and environmental entry routes.

The emissions of the pharmaceuticals were based on Dutch sales M (kg/y). Per pharmaceutical we assumed a total consumption and a fraction excreted after consumption (f_e), and removal efficiencies equal at every STP expressed by the fraction that passes the STP (f_{STP}). Total emissions were distributed over STPs in proportion to their capacity (expressed by inhabitant equivalents (IE)). Therefore, the formula for the emission W (g/s) of pharmaceutical X from STP i

[1] For more information, see: http://ec.europa.eu/environment/nature/natura2000/.


in µg/L
0 - 0.05
0.05 - 0.1
0.1 - 0.5
0.5 - 1.0
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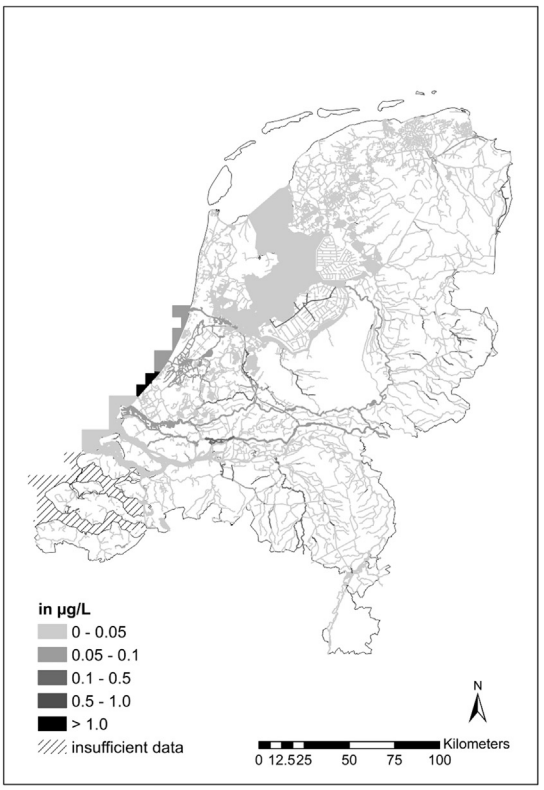
insufficient data
N
Kilometers
0 12.525 50 75 100

a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0 - 0.05
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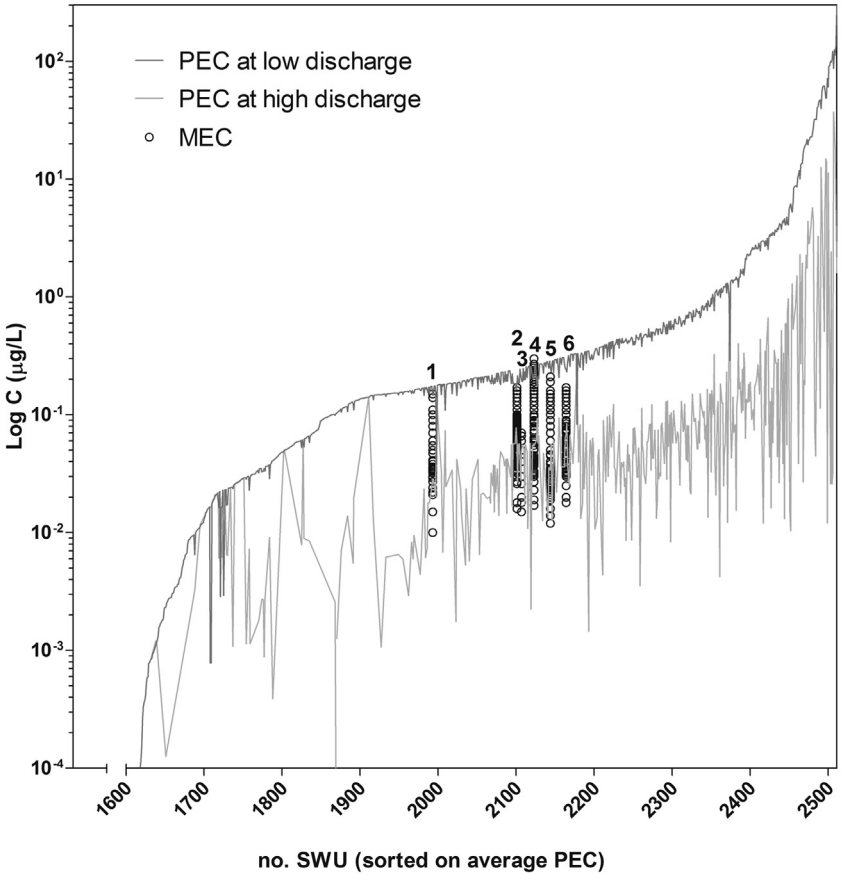
b


PEC at low discharge
PEC at high discharge
MEC
Log C (µg/L)
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
1
2
3
4
5
6
1600
1700
1800
1900
2000
2100
2200
2300
2400
2500
no. SWU (sorted on average PEC)

c

reads:

$$W_{Xi} = \frac{(M_X*1000)/(365*24*60*60)}{IE_{tot}} * f_{e_X} * f_{STP_X} * IE_i$$

Data on 345 Dutch STPs, including their capacity (IE) and geographic coordinates were collected by the Dutch water authorities. The total capacity of the 345 STPs is 24.3 million IE. Dutch sales data and physico-chemical properties of the pharmaceuticals are listed in Table 1.

We accounted for trans-boundary influx of pharmaceuticals via rivers. The related mass flux (g/s) was derived by multiplying the concentration (g/m^3) with the discharge (m^3/s). For the river Rhine at Lobith and the river Meuse at Eijsden, where both rivers enter the Netherlands, empirical data on measured concentrations and discharges were used (Ter Laak et al., 2013, 2010). Data from other (smaller) inflows were estimated based on the averaged Rhine and Meuse concentrations.

For water quality modeling, the WFD Explorer 2.0 was used which includes a schematization of the complete Dutch surface water network (Van den Roovaart et al., 2012). This model makes use of the D-WAQ open source modeling software (Deltares, 2013; Smits and Van Beek, 2013), and relies on the advection-diffusion equation with added source/sink terms (Chapra, 1996), formulated as a mass balance equation. This equation constitutes the mathematical basis for many water quality models, including the GREAT-ER and LF2000-WQX models which have already been successfully used to simulate down-the-drain chemicals (Kehrein et al., 2014; Price et al., 2010). The pre-existing WFD Explorer 2.0 model has been set up on the basis of the Netherlands Hydrological Instrument (NHI, De Lange et al., 2014; Hoogewoud et al., 2012). The schematization includes approximately 8500 sub-catchment areas and approximately 9200 explicitly modeled surface water nodes. Approximately 2500 of these, referred to as surface water units (SWU), represent the surface water bodies as they have been defined in the Water Framework Directive River Basin Management Plans. These SWUs vary in surface area from approximately 750 m^2 to 970 ha. The water fluxes driving the transport of pharmaceuticals in the water quality model have been derived from dynamic NHI v2.2 simulations for the period 1996–2006. These simulations represent all relevant hydrological processes for Dutch conditions, including precipitation, evaporation and evapotranspiration, infiltration and drainage, urban run-off, trans-boundary fluxes, water abstractions and return flows for various agricultural uses, industrial use and drinking water production, and water transfers for maintaining constant polder levels. Based on these results, three-monthly averaged water balances have been compiled. Here, we used the driest and the wettest period encountered in 1996–2006, in particular July–September 2003 and October–December 1998.

We conducted water quality simulations for the geographical distribution of substances in surface waters originating from individual STPs and trans-boundary rivers with D-WAQ, a sub-model of the WFD explorer. All 345 Dutch STPs and nine inflows from abroad, i.e. the inflow of the rivers Rhine, Meuse, Scheldt-Canal Sas van Gent, Roer, Swalm, Niers, Overijsselse Vecht, Mark-Weerijs and Dommel-Tongelreep, were taken into account. The influence of tides is not included in the model. For the Scheldt and Ems estuary no reliable estimation of discharges and thus no reliable estimation of concentrations could be made. At every emission point i a constant unit emission of 1000 g/s was simulated assuming complete mixing in every SWU, and the resulting steady state mass fluxes $F_{i,j}$ (g/s) through all SWUs j were collected in a matrix.

Simple first order decay was included to account for environmental loss processes. For efficiency reasons, we used a method to derive the results for any value of the decay rate k from just two simulations: one for a conservative tracer (F_c, no decay) and one for a non-conservative tracer (F_{nc}, decay rate $k_{nc} = 0.005\ d^{-1}$), making use of the linearity of the underlying mathematical equations. Using the simulated mass fluxes $F_{i,j}$ (g/s) for both tracers, the overall travel time $T_{i,j}$ (d) from a certain STP i to a certain SWU j was derived from the simulation results:

$$T_{i,j} = -\frac{\ln\left(\frac{F_{nc,i,j}}{F_{c,i,j}}\right)}{k_{nc}}$$

We note that this travel time could also have been calculated from the input water volumes and the input water fluxes between those water volumes. Knowing the travel time $T_{i,j}$ allows for calculating $F_{X,i,j}$ for any substance X with an arbitrary k value by:

$$F_{X,i,j} = F_{c,i,j}\exp(-k_X T_{i,j})$$

This is done for both discharge situations alike. The resulting two matrices for substance X for the high and low discharge represent how a unit mass flux originating from one single emission point spreads over the surface water network.

The mass flux of a pharmaceutical X in SWU j, $M_{X,j}$ (g/s), is calculated by scaling all mass fluxes from STP i with the actual load, and calculating the sum over all STPs and rivers. The concentration of a pharmaceutical X in SWU j, $C_{X,j}$ (g/m^3) follows after dividing the mass flux by the local water discharge Q_j (m^3/s):

$$M_{X,j} = \sum_i \frac{W_{X,i}}{1000} F_{X,i,j} \quad C_{X,j} = \frac{M_{X,j}}{Q_j}$$

The relative contribution R of STP i in SWU j compares the mass flux originating from Dutch STPs to the total mass flux in a SWU, and was determined for both discharge conditions.

$$R_{i,j} = \frac{M_{X,i_{NL},j}}{M_{X,i_{tot},j}}$$

For carbamazepine, resulting predicted environmental concentrations (PEC_{min} and PEC_{max}) were compared to measured environmental concentrations in surface waters (MEC) at seven surface water monitoring locations. The MECs were obtained from a dataset provided by the Dutch Association of River Water Companies (RIWA) with measurements taken in 2003–2013 (except for Keizersveer: 2004–2013 and Stellendam: 2010–2013). For sampling methods and analysis, see Houtman et al. (2014). Only measurement data above the detection limit were used.

2.2. Spatial selection of SWUs hosting susceptible functions

SWUs hosting susceptible functions were selected using a Geographical Information System (ArcGIS 10.1). We restricted the analysis to the susceptible functions of drinking water abstraction and Natura 2000 areas, based on the relevance for human and ecosystem health and data availability.

For drinking water, GIS data of all drinking water abstractions in

Fig. 1. ab. Predicted environmental concentrations at low (left) and high (right) discharge for carbamazepine; c. Predicted environmental concentrations at high (light grey) and low (dark grey) discharge versus measured environmental concentrations at seven monitoring stations for carbamazepine, namely 1: Andijk, 2: Lobith, 3: Stellendam (Scheelhoek), 4: Nieuwegein, 5: Keizersveer and Brakel, 6: Nieutversluis.

the Netherlands, the abstraction rate and source type (e.g. surface water, bank filtrate, groundwater) were provided by the Dutch drinking water utilities. Most groundwater abstractions and some bank filtrate abstractions are covered by a 25-year protection zone, i.e. an area around the abstraction from which water will recharge the abstraction within 25 years. GIS data on 25-year protection zones were provided by the Dutch provinces.

First, SWUs with surface water abstractions for direct treatment or for artificial infiltration (e.g. in the dunes) were selected. Bank filtrate abstractions were linked to the nearest SWU, and if applicable to its 25-year protection zone. If applicable, groundwater abstractions were also linked to their 25-year protection zone. Groundwater abstractions concerned were 'phreatic', 'semi-confined' or 'from limestone', all types that have the characteristics to potentially be fed by surface water because they are not (fully) confined. Confined groundwater abstractions were assumed to not be influenced by surface water. All SWUs crossing a 25-year protection zone were selected. Following a worst case approach, there was no correction for dilution or mixing within the abstraction, and it was assumed that the flow occurs from SWU to the aquifer.

For Natura 2000 areas, GIS data on the 163 Natura 2000 areas in the Netherlands were taken from National Georegister (National Georegister, 2014). SWUs that overlap with Natura 2000 areas were selected. No distinction was made between Habitats Directive and Birds Directive.

2.3. Prioritizing impact of STPs on SWUs hosting susceptible functions

To prioritize the STPs at or in the service area where abatement of pharmaceuticals would be most efficient, STPs were ranked for their impact on SWUs hosting susceptible functions. Per STP an impact factor (*IF*) was calculated by the following formula:

$$IF_i = \sum_j C_j \frac{F_{i,j}}{Q_j C_j} \frac{S_j}{S_{tot}}$$

The *IF* (g/m^3) of STP i in SWU j is expressed by the local concentration C_j to represent the total impact on that SWU, multiplied by $F_{i,j}$ to the total flux $Q_j * C_j$ representing the share of STP i within the total impact, and multiplied by a dimensionless weighing factor S/S_{tot} representing the relevance of the SWU for the function of interest. For the analysis of drinking water abstractions S is represented by the production volume (m^3/y), while for Natura 2000 areas S is represented by the surface area (km^2). When groundwater abstractions with multiple coupled SWUs were concerned, corresponding abstraction volumes were divided amongst the SWUs. The sum of *IF* over the SWUs gives IF_i, the impact factor of a certain STP i. *IF*s were calculated for both susceptible functions concerned, as well as both discharge conditions. The relative impact factor per STP (rIF_i) is defined as follows:

$$rIF_i = \frac{F_i}{\sum_i F_i}$$

All STPs having a rIF_i higher then 0.1% were selected and ranked.

3. Results and discussion

3.1. Surface water concentrations

Based on estimated emissions from 345 Dutch STPs and nine rivers from neighboring countries, concentrations for carbamazepine and ibuprofen were modeled on a detailed spatial scale for 2511 individual SWUs at high and low discharges. Fig. 1ab and 1c show results for carbamazepine. Of the 2511 SWUs, 83% and 65% is not influenced by STPs at high and low discharge respectively. The 95-percentile of the modeled concentrations in SWUs vary up to 88 ng/L at high discharge and up to 1.4 μg/L at low discharge. For ibuprofen modeled concentrations in SWUs vary up to 95 ng/L and 0.65 μg/L at high and low discharge respectively. At low discharge there is less dilution, but travel times are longer with a higher degradation as a result. At low discharge water is retained and redistributed in the low western part of the Netherlands, so then certain areas are influenced by STPs which are not influenced at high discharges. Due to the higher degradation rate of ibuprofen spatial differences in concentrations are higher with longer travel times from STP to SWU.

The ratio between measured and predicted environmental concentrations (MEC/PEC) indicates the predictive performance of consumption-based hydrological modeling. Available monitoring datasets at the national scale are mainly measured in large water bodies and gathered during non-extreme conditions (Houtman et al., 2014; Ter Laak et al., 2010). Monitoring data for extreme conditions as used for modeling in this study are hardly available.

Consumption-based modeling, also for the selected pharmaceuticals, was performed in earlier studies (Aldekoa et al., 2013; Celle-Jeanton et al., 2014; Hut et al., 2013; Johnson et al., 2013; Kehrein et al., 2014; Kugathas et al., 2012; Oosterhuis et al., 2013; Ter Laak et al., 2014, 2010; Verlicchi et al., 2014), mostly on a river-basin scale and for non-extreme conditions. MEC/PEC ratios in these studies are generally in good agreement. For carbamazepine ratios were within a factor 2 or 3, for the Rhine and smaller rivers in the Netherlands respectively (Ter Laak et al., 2014, 2010). Predicted concentrations for ibuprofen were overestimated with a factor 7, possibly due to an underestimation of environmental loss and or partial consumption of sales data (Ter Laak et al., 2010). A modeling effort on local scale for the Po river (Italy), from a single STP, resulted in predicted surface water concentrations of amongst others carbamazepine within a range of 0.5–2 of measured concentrations (Verlicchi et al., 2014). In a study on the Llobregat river (Spain), measured diclofenac concentrations were within a factor 0.9 – 1.6 to predicted results (Aldekoa et al., 2013).

Our study knows a comparable approach to determine PECs, however calculated for extreme discharge conditions. Because of the national scope boundary points are included instead of including whole river basins, which introduced additional uncertainties (Van den Roovaart et al., 2012). Nevertheless, monitoring datasets from 2003 to 2014 which are available for seven monitoring stations do mostly fall within the range of modeled concentrations for the extreme discharge conditions (see Fig. 1c).

3.2. Relative contribution of Dutch STPs to the surface water concentration

The relative contribution *R* from Dutch STPs to the total modeled surface water concentration for both discharge conditions is shown in Fig. 2ab and 2c. For most impacted SWUs, the Dutch contribution to the surface water concentrations dominates compared to the contribution by neighboring countries, although the last contributions contain uncertainties as they are partly estimated (Fig 2c). *R* is in general high in small streams and in SWUs with a large travel distance from the (upstream) country borders. Respectively, this can be explained by less dilution in smaller streams and more degradation of substances with longer travel times. Besides variation in *R* amongst the SWUs, there is variation between the two discharge extremes as *R* is generally higher at low discharge. Also,

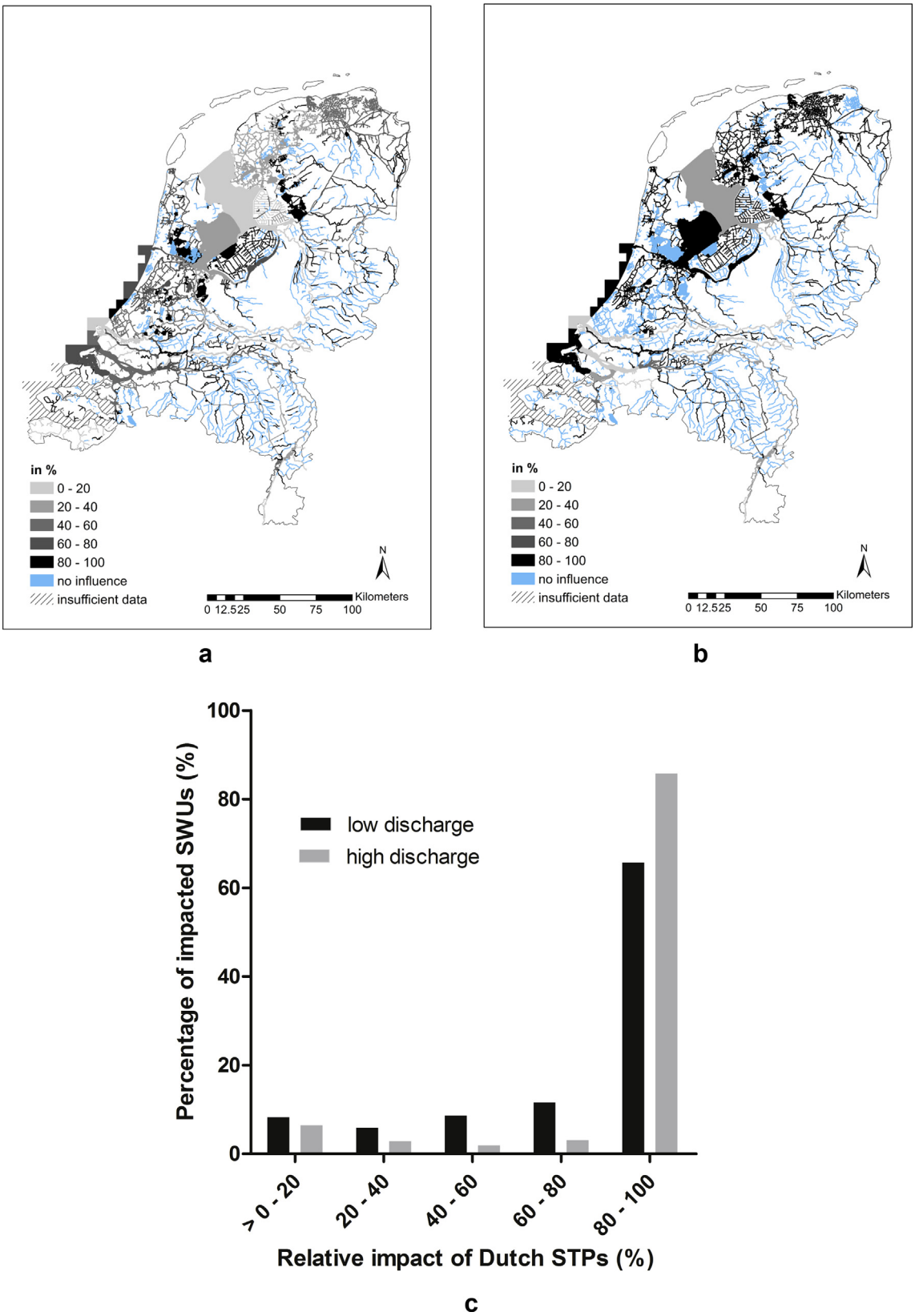


Fig. 2. ab. Modeled relative contribution of Dutch STPs to carbamazepine concentrations at low (left) and high (right) discharge. c. Relative contribution of Dutch STPs to predicted surface water concentrations at low (black) and high (grey) discharge for carbamazepine.

for the more degradable pharmaceutical ibuprofen *R* is higher (data not shown). In the near future, as a result of the planned upgrading of wastewater treatment plants in Switzerland (Eggen et al., 2014) and parts of Germany, the relative share of Dutch STPs to the surface water concentration in the shared river basins, such as the Rhine basin, is expected to increase.

3.3. Selected SWUs hosting susceptible functions

In total, 499 out of the total 2511 SWUs have either only a drinking water function (92) or a Natura 2000 function (377) or both (30).

The majority of the abstracted volumes of surface water and bank filtrate for drinking water production are influenced by STP effluent at low discharge (Table 2). This also holds for a minority of the abstracted volumes of groundwater. In total, half of the abstracted volume of raw water for drinking water production is influenced by STPs at low discharge. At high discharge, eight additional bank filtrate abstraction locations are in contact with a SWU influenced by STP, and one bank filtrate location appears not to be influenced. Besides, four groundwater abstractions influenced at low discharges are not influenced by STP effluent at high discharges. This shows that modeling for both extreme hydrologic conditions is relevant.

As a general and worst-case approach applied to all groundwater abstractions, it was assumed that the groundwater abstractions are influenced by intrusion of SWUs crossing their 25-year protection zone. A more detailed location-specific modeling per individual groundwater abstraction might give insight if mixing-up with cleaner water types occurs or if groundwater feeds SWUs instead of vice versa, both leading to less influence by STPs than modeled here using worst-case assumptions. On the longer run however, STP influences outside the 25-year protection zones might influence the groundwater abstractions, which is not taken into account in the present study.

There are 163 Natura 2000 areas in the Netherlands, of which 108 are in contact with a SWU. Of these, 85 Natura 2000 areas are influenced by STPs at low discharge. The total area of the Dutch Natura 2000 areas is 17,365 km^2, about a quarter (4576 km^2) of which is influenced by STPs.

3.4. Prioritizing impact of STPs on SWU hosting susceptible functions

STPs influencing SWUs hosting susceptible functions were prioritized according to their relative impact factor, based on modeled carbamazepine concentrations both at high and low discharges. The impact factor is based on the fraction of the emission of an STP that reaches SWUs with a susceptible function. This prioritization gives insight where abatement options are most efficient with regard to susceptible functions of the water system.

Table 3 shows the results for STPs ($rIF_i > 0.1\%$) influencing SWUs with a drinking water or Natura 2000 function. Note that the cumulative impact factor, to which the *rIF* is scaled, differs between drinking water and Natura 2000 because of the quantities used for the weighing (abstraction volume versus surface area). At low discharge a slightly higher number of STPs impact SWUs hosting susceptible functions. In total, 19% of all STPs is prioritized for the drinking water function, and 39% is prioritized with regard to the Natura 2000 function. Of all STPs 13% impact both a drinking water and a Natura 2000 function.

The cumulative impact of the prioritized STPs is 94–96% with respect to the total impact of Dutch STPs. Including the impact through rivers from abroad, the prioritized STPs have a cumulative impact ranging from 45 to 78% (Table 3).

A comparison between the percentage of selected STPs to the corresponding percentage of the total IE, indicates that STPs with a higher capacity (IE) are relatively more prioritized. Fig. 3abc shows all Dutch STPs including their capacity, and those STPs prioritized for the two selected susceptible functions at low discharge. The spatial distribution of prioritized STPs for the drinking water or Natura 2000 function differs substantially; for drinking water STPs especially in the south-east of the Netherlands are prioritized while for Natura 2000 these are all over the Netherlands. This difference relates to the spatial distribution of both drinking water abstractions and Natura 2000 functions of the surface water. Some STPs having a high capacity in the western part of the Netherlands are not relevant for the susceptible functions selected – they might nevertheless be very relevant to susceptible functions in the marine environment.

3.5. Possibilities for further improvement of the analysis tool

This study is as far as known the first spatial analysis on the impact of STP emissions on susceptible functions of surface waters, based on hydrology and consumption-based modeling. The focus is on the Dutch scale; a detailed surface water network in a densely populated country with many STPs and susceptible functions.

Some further refinements of the model may contribute to a sounder STP prioritization.

For drinking water the purification technologies in place at drinking water production locations, especially where drinking water is produced from surface water, might be accounted for. For Natura 2000, the interaction with the groundwater system could be incorporated in the analysis.

Refinements of assumptions concerning spatial variation in demography, including socio-cultural background, and seasonal variation in pharmaceutical consumption might improve the estimated loads. Spatial differentiation of removal efficiencies between STPs and temporal variation in percentage of capacity used improves the reliability of predicted impacts per STP. No regional difference in the presence of hospitals and care homes was assumed (Batenburg-Eddes et al., 2002), as hospitals are for most pharmaceuticals a minor point of emission (Le Corre et al., 2012).

Photo- and biodegradation and sorption are dominant environmental reduction processes (Andreozzi et al., 2003; Kümmerer, 2009; Packer et al., 2003), which depend on environmental circumstances as radiation, temperature and the presence and type of sorbents. A dynamic value for environmental decay might better account for this variability.

Future changes in demography, (medical) technologies and pharmaceutical use could be taken into account (Green et al., 2013; Van der Aa et al., 2011). Real data on extreme discharges were used here, which might be expanded by future scenarios for hydrology (Fowler et al., 2007; Whitehead et al., 2009). Major changes in Dutch water management practices (Vink et al., 2013) are to be included in the underlying hydrological WFD explorer

Table 2
Drinking water abstractions in contact with SWU and influenced by STPs: amounts and abstraction volumes.[a]

Drinking water abstraction	Total	In contact with SWU	Influenced by STP	Abstraction volume (million m^3/y)	Abstraction volume influenced by STP (million m^3/y)
Surface water[b]	9	9	8	416	411 (99%)
Bank filtrate	20	20	11	108	74 (68%)
Groundwater	180	40	26	739	144 (19%)
Total	209	69	45	1.262	629 (50%)

[a] Based on carbamazepine at low discharge.
[b] Including abstraction for artificial recharge in dunes.

Table 3
Numbers of selected STPs influencing surface waters hosting susceptible functions, including their share in cumulative impact.

	Selected STPs	% of total IE	% cumulative impact Dutch STPs only	% cumulative impact Dutch STPs and neighboring countries
Drinking water				
High discharge	58 (17%)	30.7	96	78
Low discharge	65 (19%)	33.4	96	76
Natura 2000				
High discharge	108 (31%)	43.7	94	57
Low discharge	133 (39%)	51.2	95	45

schematization.

To overcome uncertainties at regional or local scale, a nested model on continental, river basin, national and regional scale can be considered. Including estuarine effects in the Scheldt and Ems rivers would improve the results.

Furthermore the model can be broadened towards other susceptible functions of the water system such as recreation or food production. An analogous model could be developed for more diffuse emission sources, e.g. pesticides or veterinary pharmaceuticals, or for other contaminants such as pathogens (Hofstra et al., 2013). This study modeled single parent compounds. In the future also transformation products and mixtures might be considered, relevant for a comprehensive risk assessment (De Jongh et al., 2012; Escher and Fenner, 2011; Ter Laak et al., 2014).

In finding high impact STPs an algorithm could be used (Moschet et al., 2013; Ort et al., 2009), for which criteria can be derived from this study. Characteristics such as the STPs capacity, the discharge of the receiving SWU, travel times from STP to SWUs hosting a susceptible function, connectivity of the surface water network, and management practices influencing flow direction together determine a STP's impact factor.

In view of implementation of the developed tool within a policy or legislative context, it can be questioned what level of accuracy is required to base decisions upon. A further sensitivity analysis to the factors mentioned here might be helpful to explore which refinements are needed.

3.6. Possibilities for implementation by stakeholders

Prioritization of STPs minding susceptible functions in the surface water supports cost-efficient implementation of local abatement options. Whether these abatement options are water treatment technologies at the STP, options before the entrance at the STP or a combination; emissions are ideally to be reduced in an integrated way in different stages in the chemical life cycle from production to consumption and emission (Schirmer and Schirmer, 2008). Current or future investments at STPs might be assessed in planning when to implement abatement measures to reduce costs.

Depending on the various interests of stakeholders considered, results of prioritization may differ. Alternative choices are to weigh all SWUs hosting a certain susceptible function alike, without applying a correction for volume or area as was done here. Also, one might assign more weight to one type of susceptible function compared to another, or concentrate actions solely on SWUs with a concentration above a certain threshold.

Impact factors and the resulting prioritization of STPs are thus partly steered by interests.

4. Conclusion

For consumed pharmaceuticals, sewage treatment plants (STPs) are a major point of entry to surface waters which provide vital functions. Modeling the impact of STPs on susceptible functions of the surface water system on a nation-wide scale allows for a spatially smart implementation of abatement options.

We performed a detailed spatial analysis of STP emissions of two pharmaceuticals into the Dutch surface water system and modeled concentrations for two extreme discharge conditions. We identified surface waters with a susceptible function, i.e. drinking water, Natura 2000 or both.

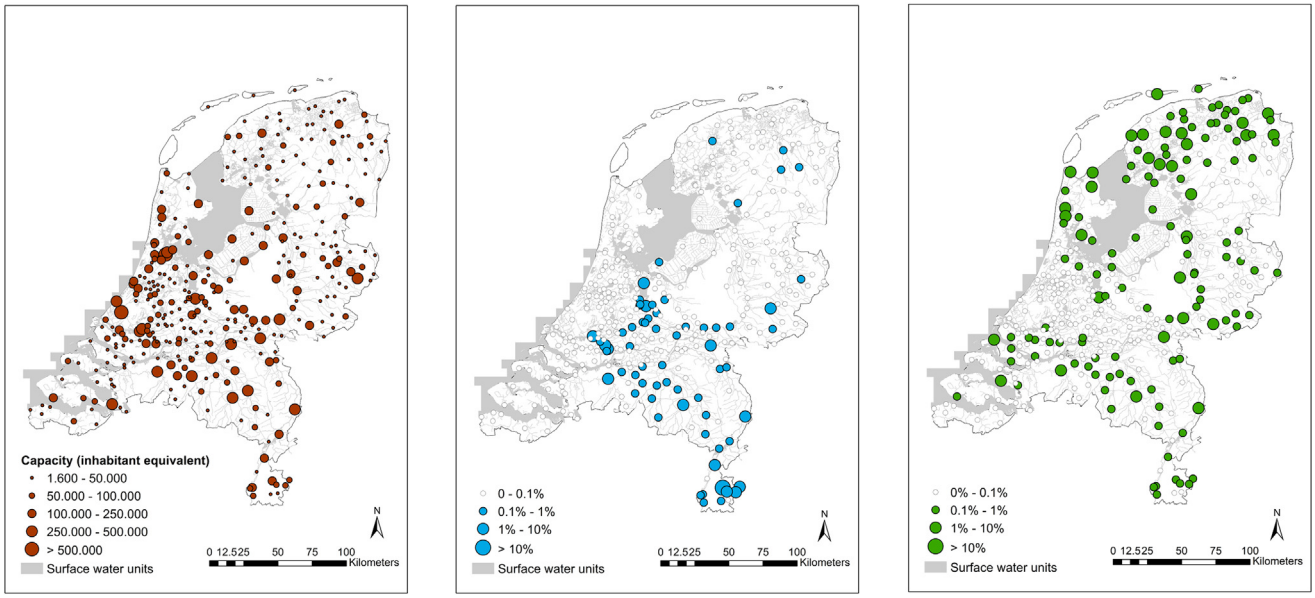


Fig. 3. abc. Overview of all Dutch STPs and a) the STP's capacity in IE, b) the STP's relative impact on SWUs hosting a drinking water function and c) the STP's relative impact on Natura 2000 functions, scaled to the cumulative impact of all Dutch STPs at low discharge conditions and based on data for carbamazepine.

Significant amounts of water abstracted for drinking water production, and of water in Natura 2000 zones, appeared to be influenced by STPs at low discharge. One out of five of all Dutch STPs are prioritized with regard to their impact on the drinking water function, whereas two out of five of all STPs are prioritized with regard to the Natura 2000 function of the surface water. The prioritized STPs together bear the majority of the cumulative impact. Including the impact through rivers from abroad, impact of prioritized Dutch STPs is still significant.

The model gives insight in the spatial distribution of STPs across the Netherlands that have a relatively high impact, where local abatement options to reduce concentrations of pharmaceuticals deliver most benefits for susceptible functions in surface waters.

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