Science of the Total Environment 640–641 (2018) 1489–1499

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Impact of industrial waste water treatment plants on Dutch surface waters and drinking water sources

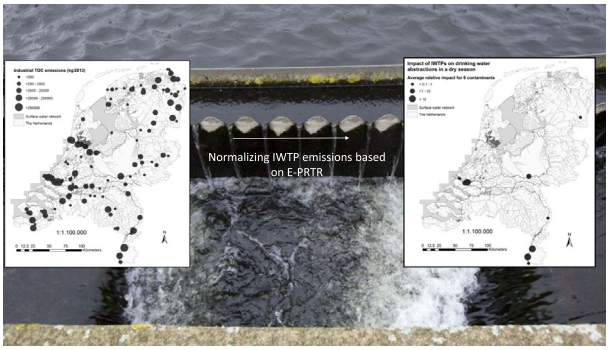
Annemarie P. van Wezel [a,b,*], Floris van den Hurk [a], Rosa M.A. Sjerps [a], Erwin M. Meijers [c], Erwin W.M. Roex [c], Thomas L. ter Laak [a]

[a] KWR Watercycle Research Institute, Nieuwegein, the Netherlands
[b] Copernicus Institute of Sustainable Development, Utrecht University, Utrecht, the Netherlands
[c] Deltares, Utrecht, the Netherlands

HIGHLIGHTS

- Discharge of treated industrial wastewater to surface waters affect CEC water quality.
- We model industrial emissions of all Dutch industrial water treatment plants (IWTP).
- We coupled European Pollutant Release Transfer Register data to a hydrological model.
- Information on industrial effluent concentrations is lacking for many chemicals.
- From all Dutch 182 IWTPs only 15 highly impact drinking water production.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 16 January 2018
Received in revised form 25 May 2018
Accepted 26 May 2018
Available online xxxx

Editor: Thomas Kevin V

Keywords:
Industrial waste water
Effluent
Drinking water

ABSTRACT

Direct industrial discharges of Chemicals of Emerging Concern (CEC) to surface water via industrial wastewater treatment plants (IWTP) gained relatively little attention compared to discharges via municipal sewage water treatment plants. IWTP effluents may however seriously affect surface water quality. Here we modelled direct industrial emissions of all 182 Dutch IWTP from 19 different industrial classes, and derived their impact on Dutch surface water quality and drinking water production. We selected industrial chemicals relevant for drinking water production, however a lack of systematic information on concentrations in IWTP effluents for many chemicals of interest was found. Therefore, we used data from the European Pollutant Release and Transfer Register and data on Dutch IWTP as surrogate. We coupled these to a detailed hydrological model under two extreme river discharge conditions, and compared the predicted and measured concentrations. We derived relative impact factors for the IWTP based on their contribution to concentrations at surface water locations with a drinking water function. In total, a third of the abstracted water for drinking water production is influenced by the IWTP. From all Dutch 182 IWTP, only a limited number has - based on the model approach using surrogate parameters - a high impact on surface waters with a drinking water function. Mitigation measures can be taken cost-efficiently, by placing extra treatment technologies at the IWTP with high impact. Finally, we propose recommendations for licensing and controlling industrial aqueous emissions and give suggestions to fill the currently existing knowledge gaps and diminish uncertainties in the approach.

* Corresponding author at: KWR Watercycle Research Institute, Nieuwegein, the Netherlands.
E-mail address: annemarie.van.wezel@kwrwater.nl (A.P. van Wezel).

https://doi.org/10.1016/j.scitotenv.2018.05.325
0048-9697/

1. Introduction

The production and use of chemicals continues to increase at a speed that outpaces other agents of global change. This holds for both the number of authorized chemicals as for the volumes produced and used (Bernhardt et al., 2017; Wilson and Schwarzman, 2009). Currently worldwide over 348.000 chemicals are registered and regulated (CHEMLIST, 2018). Chemicals of emerging concern (CEC) are measured ubiquitously in low concentrations (mostly ng/L range) in European surface waters, effluents and groundwaters (Loos et al., 2009, 2010a, 2010b, 2013). CEC comprehend a large group of compounds that are not commonly monitored, for which there is scarce information on possible effects, and for which no regulatory criteria or quality standards exist while they potentially might pose risks (Halden, 2015). Example CEC are pharmaceuticals, personal care products, plasticizers, surfactants and pesticides, and industrial chemicals. After incidental releases, CEC concentrations in rivers can be orders of magnitude higher, up to μg/L levels (De Hoogh et al., 2006; Rebelo et al., 2014). Climate change and thus more frequent and severe low river discharges, leads to periods with increased surface water concentrations of synthetic chemicals (Delpla et al., 2009; Petrovic et al., 2011; Sjerps et al., 2017; Van Vliet and Zwolsman, 2008). Chemical pollution of our waters is a global public concern (Malaj et al., 2014, Richardson and Ternes, 2014, Schwarzenbach et al., 2006). Since surface waters provide vital functions such as drinking water production, nature, recreation and food production, it is fundamental to localize and control areas with potential risk associated to CECs (Van Wezel et al., 2017).

Direct industrial discharges of CEC to surface water via industrial wastewater treatment plants (IWTP) gained relatively little attention compared to discharges via municipal sewage water treatment plants (STP). Several reviews stress the importance of STP effluents to surface water quality (e.g. Luo et al., 2014; Tran et al., 2018). IWTP effluents however may also seriously affect surface water quality (Boiteux et al., 2017, Hu et al., 2016, Lee et al., 2011, Lindim et al., 2015, Loos et al., 2007, Ruff et al., 2015, Salgueiro-González et al., 2015). For example, industrial effluents from textile, chemical or pharmaceutical manufacture industries with endocrine activity are found across Europe (Eggen et al., 2003, Schriks et al., 2010, Vethaak et al., 2005). In the Netherlands the number and total capacity of IWTP is comparable to that of STP (CBS Statline, 2017). Industrial plants may directly discharge via IWTP, or discharge indirectly via STP. In the latter case, the resulting effluent is from mixed municipal and industrial origin. The composition of industrial effluent is expected to vary more in time than the composition of municipal effluent, related to changes in the exact industrial production processes, batch-wise production, and maintenance. In Europe, IWTP emissions have to comply to the Industrial Emissions Directive (IED, 2010/75/EU). The IED establishes a procedure for authorising industrial activities, sets minimum requirements to be included in permits and prescribes the application of Best Available Techniques (BAT) (Evrard et al., 2016). BAT imply good industrial processes, such as storing waste or cleaning and rinsing baths (Derden and Huybrechts, 2013; Ozturk et al., 2016), but also the application of effective waste water treatment technologies.

In the Netherlands, 40% of the total drinking water production originates from surface water. Dutch drinking water companies that rely on surface water as a source frequently stop their surface water intake because of problems with industrial emissions (RIWA, 2017). For example, during the summer of 2015, an IWTP emission of amongst others pyrazole resulted in a long-term stop of surface water intake for drinking water production (Baken et al., 2016). In 2018, the license for industrial emission of 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoic acid (FRD-903, 'GenX') is debated in the Dutch court because of expected future problems for drinking water production. Also other examples of industrial emissions giving rise to water quality problems (Boiteux et al., 2017; Kosaka et al., 2014; Van Leerdam et al., 2014), portray the relevance of industrial emissions on surface water quality and drinking water production.

Next to water quality monitoring, modelling may also be used to localize areas with potential risks associated to CEC (Fan et al., 2015). Water quality modelling is fast and cheap compared to monitoring, and has a high spatial and temporal resolution. Various approaches have been developed to model concentrations of CEC (Aldekoa et al., 2013; Coppens et al., 2015; Ippolito et al., 2015; Kapo et al., 2015; Kehrein et al., 2015; Kuroda et al., 2016; Lindim et al., 2016), predominantly applied for specific down-the-drain consumer chemicals such as pharmaceuticals. Water quality models can also be used for an a priori evaluation of mitigation strategies (Coppens et al., 2015; Zijp et al., 2016).

Here we aim to model direct industrial emissions to surface water, their impact on Dutch surface water quality and drinking water production and the options to minimize adverse impacts. After selecting industrial chemicals which are relevant for drinking water production, we used data from the European Pollutant Release and Transfer Register and data on Dutch IWTP, and coupled these to a detailed hydrological model under two extreme river discharge conditions. The predicted concentrations were compared to measured concentrations. We derived relative impact factors for the IWTP, based on their contribution to concentrations at surface water locations with a drinking water function. Finally, we propose recommendations for licensing and controlling industrial aqueous emissions.

2. Material and methods

2.1. Selection of industrial chemicals

To ensure that modelled concentrations can be compared to measured concentrations, chemicals for which monitoring data are available at surface water intake points of Dutch drinking water utilities were selected (total 955 chemicals, RIWA water quality database). These were combined with chemicals earlier prioritized based on their occurrence in Dutch surface waters and drinking water (Sjerps et al., 2016). In addition, literature data on occurrence and prioritization in effluents, surface-, ground- and drinking water were added.

Information on the industrial discharges of these chemicals in the Dutch and European Pollutant Release and Transfer Register (PRTR) was used for water quality modelling. The ultimate selection of chemicals used in the modelling is limited by the data availability in the PRTR database.

Other pathways followed to retrieve information on industrial emissions and concentrations in industrial effluents for the selected chemicals are described in S.I. A, these pathways included searches in databases of permitting authorities, additional inventories amongst industries and case studies on permits for industrial discharges via IWTP.

2.2. Normalizing IWTP emissions based on PRTR

For specific industrial chemicals Dutch data on emissions are scarce, while for total organic carbon (TOC) Dutch data are highly abundant. Therefore we used European PRTR data to normalize IWTP emissions per industrial class on TOC. Reporting to the European *E*-PRTR is governed by EC directive 166/2006, requiring that annual emissions for 91 chemicals and chemical classes per industrial site are publicly reported if emission surpasses prescribed thresholds. In addition to the 28 EU member states several other countries report according to *E*-PRTR, therefore emission data are available for 39 countries (Sörme et al., 2016). E-PRTR data for compound groups were not used in this study for water quality modelling, as decay rates for chemicals within the group may vary highly. *E*-PRTR data for greenhouse and other gases, heavy metals and inorganic substances were not considered in the current study focusing on organic substances. This leaves *E*-PRTR data for 20 pesticides, 14 chlorinated organic substances and 11 other organic substances. The available E-PRTR data for 2013 for all EU-member states and Iceland, Liechtenstein, Norway, Serbia and Switzerland were used

to estimate emissions per industrial class. The loads for emitted industrial chemical X were based on all *E*-PRTR data normalized to the loads for emitted TOC per NACE-code for a specific industrial sector (Nomenclature statistique des Activités économiques dans la Communauté Européenne, from the Statistical Classification of Economic Activity in the European Community), according to:

$$\frac{X}{TOC} = \frac{\frac{x\ sum}{n_X}}{\frac{TOC\ sum}{n_{TOC}}}$$

The ratio of kg X per kg TOC is estimated by the ratio of the total *E*-PRTR summed emission of X (X_{sum}, kg/yr) divided by the number of IWTP reporting X (n_X) and the summed reported emission of TOC (TOC_{sum}, kg/yr) divided by the number of IWTP reporting TOC (n_{TOC}). The assumption is that the IWTP present in n_X are representative for all IWTP present in n_{TOC}. Based on abundant yearly TOC emissions for the Dutch IWTP and their NACE-code, emissions of the selected industrial chemicals were then estimated per IWTP (g/s). These estimated industrial emissions per IWTP and the decay rates of the chemicals were then used to scale the modelled hypothetical emissions of 1000 g/s (see below).

2.3. Water quality modelling

The Dutch water system is heavily managed, given that large parts of the Netherlands are below sea level. A spatially detailed hydrological water quality model, i.e. the Dutch Water Framework Directive (WFD) model version 2.0 in the WFD-Explorer software, was used to model concentrations from IWTP emissions in analogy to earlier work on pharmaceuticals and STP (Coppens et al., 2015). The WFD Explorer 2.0 software uses a water balance and a pollutant transport model. The Dutch WFD model is based on a network of approximately 17,500 nodes of which 2575 are surface water units (SWU) and approximately 27,000 links represent the routing of the surface water. Quarterly averaged water balance data from an extreme dry and wet season are used to incorporate climate variability, i.e. the 3rd quarter of 2003 and the 4th quarter of 1998 respectively.

A series of water quality tracer computations was performed, assuming complete and instant mixing and first order decay. Data on Dutch IWTP and their NACE-code were retrieved from the Dutch Pollutant Release and Transfer Register (D-PRTR over 2013, www.emissieregistratie.nl). Indirect industrial discharges that take place via STP were not incorporated. IWTP/SWU transfer matrices were made for both a conservative and non-conservative tracer in two extreme discharge conditions, using a hypothetical emission of 1000 g/s per IWTP. The four resulting matrices list contaminant loads (g/s) from each of the 182 IWTP at each of the 2575 SWU. The 182 IWTP are classified in 19 industrial classes and 43 sub-classes, according to the NACE-codes. These matrices were combined with emission data and decay rates of the emitted chemicals, as explained above.

In addition, loads entering the Dutch surface waters via 9 cross-border rivers, i.e. Rhine, Meuse, Scheldt, Sas van Gent (Canal), Roer, Swalm, Niers, Overijsselse Vecht, Mark or Weerijs and Dommel or Tongelreep, were incorporated in the model. RIWA monitoring data were used for Rhine at Lobith and Meuse at Eijsden (1987–2015). Concentrations reported as reporting limit were excluded, except for the lowest reporting limit reported. When the 10th percentile of all RIWA data used equals the lowest reporting limit, half the lowest reporting limit was used. Because of missing monitoring data, concentrations in other cross-border rivers were estimated based on average yearly concentrations from Rhine and Meuse, and corrected for flow rates to obtain loads per cross-border river (see Coppens et al., 2015 for more details).

The sum of the loads from all IWTP and incoming rivers per SWU gives the total mass flux (g/s) at each SWU. When divided by the local discharge (Q in m^3/s), the predicted concentration (C in g/m^3) per SWU is obtained.

Predicted environmental concentrations (PEC) were compared with actually measured environmental concentrations (MEC) between 1989 and 2015 at surface water intake points for the production of drinking water.

2.4. Impact assessment of IWTP on vulnerable drinking water areas

SWU used as source for drinking water production were selected, including surface water intake points, abstraction for river bank filtration and infiltration in the 25-yr protection zone for groundwater abstraction (see Coppens et al., 2015 for further details).

IWTP were ranked based on their impact on SWU hosting a drinking water function. Per IWTP an impact factor (IF) was calculated according to:

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

The $IF_{i,j}$ (g/m^3) of $IWTP_i$ in SWU_j was expressed by the local concentration C_j, multiplied by the load $F_{i,j}$ to the total load of all IWTP in that SWU_j (Q_jC_j), representing the share of $IWTP_i$ in the total impact, and multiplied by a dimensionless weighing factor S/S_{tot} representing the relevance of the SWU for drinking water as represented by the production volume at the production location (m^3/y). For groundwater abstractions with multiple coupled SWU, corresponding abstraction volumes were divided amongst these SWU (Coppens et al., 2015). The summed IF_i over all SWU gives the impact factor per $IWTP_i$. IFs were calculated for both discharge conditions.

The relative impact factor per IWTP rIF_i was calculated according to:

$$rIFi = \frac{IFi}{\Sigma IF}$$

The relative contribution R_j (−) to the concentration in water body j from Dutch IWTP compared to the contribution from abroad was expressed by the concentration originating from Dutch IWTP ($Cx,i_{NL}j$) divided by the total concentration ($Cx,i_{tot}j$):

$$R_j = \frac{C_{X,i_{NL},j}}{C_{X,i_{tot},j}}$$

3. Results

3.1. Selection of industrial chemicals

The selected industrial chemicals of interest are given in Table 1. The majority of these 28 chemicals is produced in volumes above 1000 t/yr and is applied in consumer products, so next to industrial emission also household emissions will be an important route to the water system. An exception might be triphenylphosphine oxide (Schlüsener et al., 2015) which is a unique by-product of the Wittig reaction applied in the chemical industry.

3.2. Normalizing IWTP emissions based on E-PRTR

The Dutch PRTR database is scarce in emission data for specific chemicals, also due to reporting thresholds. Fig. 1 shows TOC normalized European emissions (formula 1) for industrial classes which are relevant for the Netherlands, for six chemicals and four compound groups with relatively high data abundance in *E*-PRTR (see also S.I.B.1.). Highest TOC normalized emissions are reported for the refined

Table 1
Selected industrial chemicals with occurrence in the water cycle and available monitoring data.

CAS-number	Industrial chemical	Uses	REACH >100 ton/yr	REACH >1000 ton/yr	References[a]
112-49-2	1,2-Bis(2-methoxyethoxy)-ethane (triglyme)	Solvent used in ink, paints and cleaners		x	f,i
115-96-8	Tris(2-chloroethyl) phosphate (TCEP)	Reducing agent and flame retardant		x	b,c,e,f,l, m
126-73-8	Tributyl phosphate (TBP)	Solvent and plasticizer in inks, synthetic resins, gums, adhesives, herbicide and fungicide		x	b,c,f,k,l,m
13674-84-5	Tris(2-chloro-1-methylethyl) phosphate (TCPP)	Flame retardant and used in gums and plastics		x	b,c,f,k,l,m
29878-31-7	4-Methyl-1H-benzotriazole	Corrosion inhibitor, drug precursor, heating and cooling	x		f,j
3622-84-2	n-Butylbenzenesulphonamide	Plasticizer		x	f,n
51-03-6	2-(2-Butoxyethoxy)ethyl 6-propylpiperonyl ether	Solvent used in ink, paints and cleaners		x	f
78-40-0	Triethyl phosphate	Industrial catalyst, solvent, plasticizer, flame retardant		x	f,k,l,m
791-28-6	Triphenylphosphine oxide (TPPO)	Crystalizing agent		x	f,k,l,m
80-09-1	4,4′-Sulphonyldiphenol (bisphenol S)	Fast drying epoxy glues, corrosion inhibitor, paper		x	f
826–36-8	2,2,6,6-Tetramethyl-4-piperidone	Drug		x	f
83–15-8	*n*-Acetylaminoantipyrine	Drug	x		d,f
84-69-5	Diisobutyl phthalate	Plastics, nail polish, polish, inks		x	d,f,h
84-74-2	Dibutyl phthalate	Plastics, nail polish, polish, inks		x	d,f,h
95-14-7	Benzotriazole	Corrosion inhibitor, drug precursor, heating and cooling	x		a,b,c,f,k
62-53-3	Aniline	Dyes, medicine, rocket fuel		x	o
608-27-5	2,3-Dichlooraniline	Dyes, medicine, rocket fuel		x	o
95-82-9	2,5-Dichlooraniline	Dyes, medicine, rocket fuel		x	o
126-71-6	Triisobutylphosphate	Plasticizers, solvent, resins, paints, inks, antifoaming	x		b,c
288-32-4	Trifenyl-imidazole-triglycine	Corrosion inhibitor, flame retardant		x	g
80-05-7	Bisfenol a	Fast drying epoxy glues, corrosion inhibitor, paper, thermal paper		x	e
554-00-7	2,4-Dichlooraniline	Dyes, medicine, rocket fuel		x	o
95-76-1	3,4-Dichlooraniline	Dyes, medicine, rocket fuel		x	o
626-43-7	3,5-Dichlooraniline	Dyes, medicine, rocket fuel		x	o
1222-05-5	Galaxolide (HHCB)	Personal care products, cleaning		x	e
117-81-7	Bis(2-ethylhexyl) phthalate (DEHP)	Plastics		x	h
123-91-1	1,4-Dioxane	Stabilizer, aluminium packages, solvent in ink and adhesives		x	h
129-00-0	Pyrene	Stabilizer aluminium packages, solvent in ink and adhesives		x	h,p

[a] a) Loos et al., 2009 b) Loos et al., 2010a, c) Loos et al., 2010b, d) Von der Ohe et al., 2011, e) Lapworth et al., 2012, f) Sjerps et al., 2016, g) Velzeboer et al., 2014, h) Roex, 2003, i) Stepien and Püttmann, 2014, j) Kiss and Fries, 2009, k) Cristale et al., 2013a, l) Cristale et al., 2013b, m) Ding et al., 2015, n) Rider et al., 2012, o) Tas and Pavlostathis, 2014, p) Baldwin et al., 2016.

petroleum and basic chemicals industry, and also for metal and paper industry.

Of the selected industrial chemicals of interest, only for bis(2-ethylhexyl)phthalate (DEHP) TOC normalized emissions based on the *E*-PRTR data could be derived. Next to DEHP, as surrogate parameters therefore also benzene, dichloromethane, toluene, 1,2-dichloroethane and vinylchloride were used for further modelling. These substances were chosen because of data availability and relative high aqueous emissions, and as different industry categories dominate the emission of these model chemicals. Even when using *E*-PRTR data, sufficient sector-specific information on emissions is lacking, also for relevant sectors as producers of dyes and pigments, pesticides or paints and coatings.

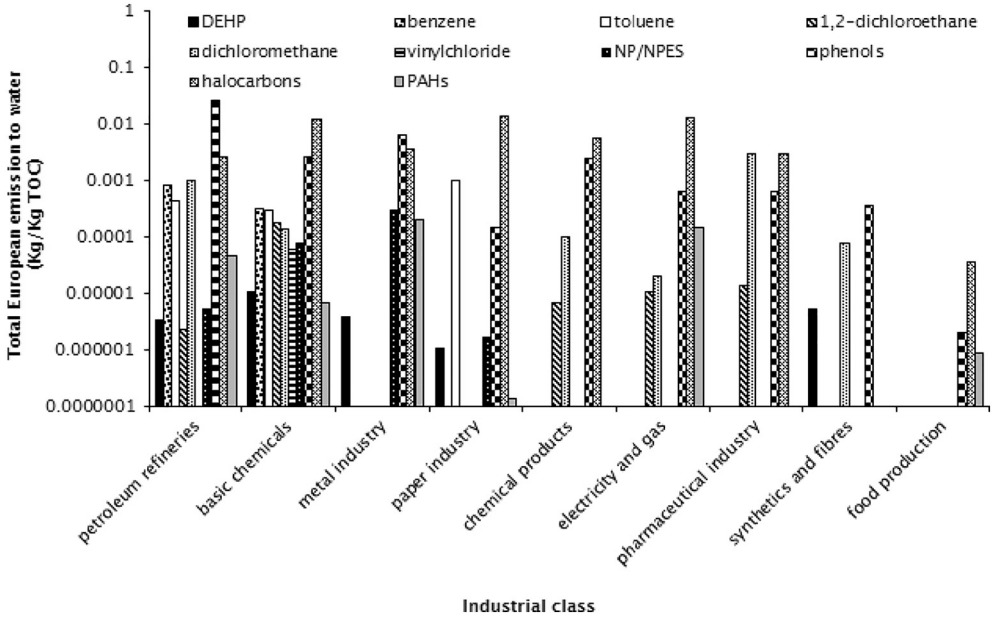


Fig. 1. Total European industrial emissions to water in kg per kg TOC for the year 2013, for all EU-member states and Iceland, Lichtenstein, Norway, Servia and Switzerland, per industrial class (source: E-PRTR). *Zero emissions for the motor-industry, metal coating, beverages industry, paints, coating and ink, machinery and electrics, ship-building, plastic products and glass industry not shown.

Environmental half-lives and corresponding decay rates for the selected chemicals are listed in Table 2, assuming first-order decay. The loads from cross-border rivers Rhine and Meuse are given in Table 3. For DEHP and vinyl chloride monitoring data from both cross-border rivers were too scarce to further model surface water concentrations.

3.3. Predicted surface water concentrations and comparison to monitoring data

Predicted surface water concentrations for benzene, toluene, dichloromethane and 1,2-dichloroethane resulting from the combined industrial emissions and cross-border rivers under low discharge vary highly over the SWU (Fig. 2). The maximum predicted concentrations are in the same range as predicted earlier for the pharmaceuticals carbamazepine and ibuprofen with STPs as a source (Coppens et al., 2015).

From this earlier work on pharmaceuticals which primarily have STP effluents as a source, it is known that the hydrological model approach used is well capable to predict measured concentrations. This despite the complexity of the water management system in the Netherlands. Monitoring data for those pharmaceuticals, which were available at 7 drinking water intake locations along the rivers Rhine and Meuse, were shown to fall within the range of modelled concentrations at two extreme discharge conditions (see Fig. 1.c. of Coppens et al., 2015).

As the current followed a comparable approach, but for industrial chemicals with IWTP as a source, the impacts of the IWTP effluents on water quality are expected to be as well predicted as was the case for pharmaceuticals with STP as a source. In the case of industrial chemicals the measured concentrations of the modelled chemicals (1989 to 2015) often exceed predicted concentrations (Fig. 2). This underestimation was expected beforehand as contrary to the pharmaceuticals model study, for these industrial chemicals other emission sources next to the IWTP emission source modelled will significantly contribute to the surface water concentrations. Examples of other relevant emission sources which are not included in the model approach are indirect industrial emissions via STP, airborne deposition and releases during the use and waste phase in the life cycle of the industrial chemicals. These other relevant emission sources were not taken into account in the current model approach, as central question here focused on the relevance of the IWTP emissions.

The approach to use Rhine and Meuse monitoring data to estimate loads for small cross-border river could be further improved by correcting for the actual presence of IWTP and their NACE-codes in the cross-border river basins. As many large industries are located close to large rivers, our approach might overestimate cross-border inputs by smaller rivers. The Dutch WFD model is to be further improved especially on mass fluxes in the large estuaries such as the Western Scheldt.

3.4. Impact assessment of IWTP on vulnerable drinking water areas

For only a limited share of the SWU with increased concentrations of the modelled industrial chemicals, the contribution of the Dutch IWTP is a dominant factor (Fig. 3). Especially at high river discharges the modelled impact of the IWTP is limited.

The impact of an IWTP on drinking water production is a combined effect of the industrial class concerned, the capacity or the IWTP, the geographic location and the hydrologic coupling to drinking water intake. Based on the described modelling approach from all Dutch 182 IWTP, that cover 43 different industrial (sub-)classes, only a limited number at low discharge or at high discharge have an impact factor higher than 0.1% (Table 4). These IWTP are typically related to the plastic, paper, petroleum or basic chemicals industry (see also S.I.B.2.).

One large IWTP is responsible for a large share of the impact for all six modelled chemicals, which is related to its size and the hydrological relation with drinking water intake. For most chemicals, IWTP in the south-east of the Netherlands show high impact. For dichloromethane which is emitted by the petroleum industry, also the Rotterdam harbour is of importance (Fig. 4).

In total 32% of the abstracted water for drinking water production is affected by IWTP according to the model (Table 5), which is less than the 50% that is impacted by STP (Coppens et al., 2015).

Table 2
Environmental half-life values and corresponding decay rates (Howard, 1991).

Chemical	$t_{1/2}$ Winter (d)	$t_{1/2}$ Summer (d)	kx Winter	kx Summer
DEHP	23	5	−0,030	−0,139
Benzene	16	5	−0,043	−0,139
Toluene	22	4	−0,032	−0,173
1,2-Dichloroethane	180	100	−0,004	−0,007
Dichloromethane	28	7	−0,025	−0,099
Vinyl chloride	180	28	−0,004	−0,025

Table 3
Input loads from cross-border rivers based on RIWA data 1987–2015.

Contaminant	Rhine at Lobith (μg/L)			Meuse at Eijsden (μg/L)		
	90th percentile	10th percentile	n	90th percentile	10th percentile	n
1,2-Dichloorethane	0,063	0,005	318	2000	0,029	763
Benzene	0,040	0,005	290	0,100	0,005	460
Dichloromethane	0,116	0,005	65	4480	0,020	565
Toluene	0,010	0,005	293	0,120	0,005	465

4. Discussion

4.1. Uncertainties of approach

This study reveals that systematic information on concentrations or loads in IWTP effluents for many chemicals of interest is lacking (see also S.I. A). This holds for a wide range of especially relatively hydrophilic chemicals, which are a concern for drinking water production because of their relatively low removal efficiencies by commonly used water treatment technologies.

Because of this information gap, we relied on EU-wide *E*-PRTR data and included surrogate parameters for which information was available, normalized per industrial sector on TOC. This was combined with available data on Dutch industrial emissions for TOC per IWTP per NACE-code. Several uncertainties can be mentioned, e.g.;

- the approach relies on the limited number of relatively well-known chemicals that are reported in the E-PRTR, which are surrogate parameters for the chemicals of interest except for DEHP.
- during normalization of chemical to TOC emissions, it is assumed that the selection of IWTP for which chemical emission data are available is an a-select representation of the broader set of IWTP for which TOC emissions are available.
- an industrial area served by a IWTP may contain a mix of industrial sectors, in our approach we use available statistical data where the various activities are often combined to one NACE-code.

For STP effluents more information on CEC is available then for IWTP effluents, both in scientific literature and in databases. In the Netherlands for example, as part of the PRTR, the Watson database on STP effluents contains over 900 chemicals. These includes 86 industrial chemicals with ≥10 measurements in the period 1990–2015 with concentrations in STP effluent above 0.01 μg/L (see S.I. C), including flame retardants and endocrine disrupting chemicals. Most STP effluents result from mixed input by households and industrial waste water, e.g. 93% of Dutch STP treat mixed household and industrial waste water and the proportion of industrial influent is on average 24% with a maximum of 90% (personal communication Kees Baas Central Bureau for

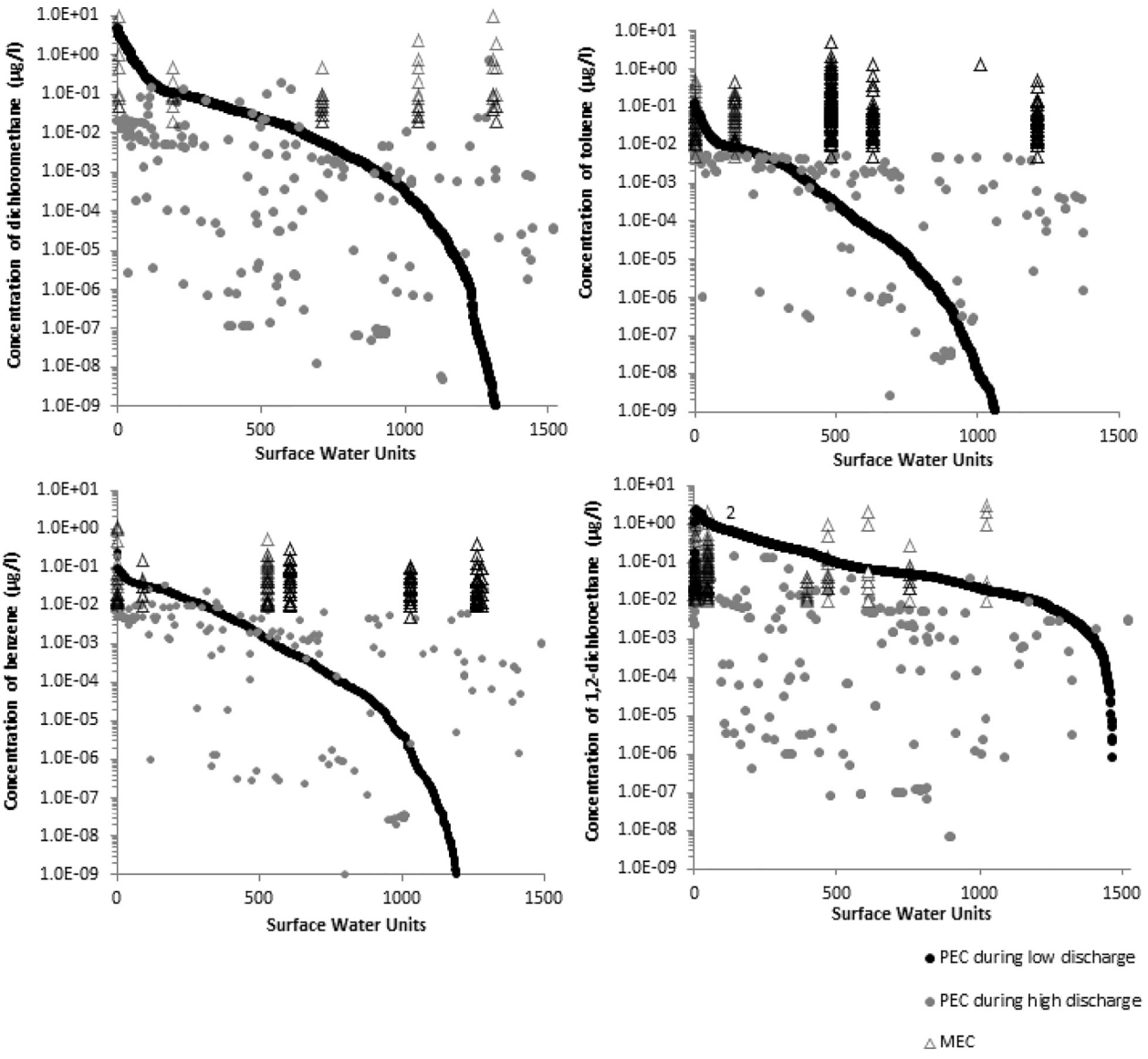


Fig. 2. Predicted surface water concentrations per surface water unit at low and high discharge (PEC) compared to measured concentrations at drinking water intake points (MEC, data RIWA 1989–2015). Surface water units are ranked according to PEC during low discharge.

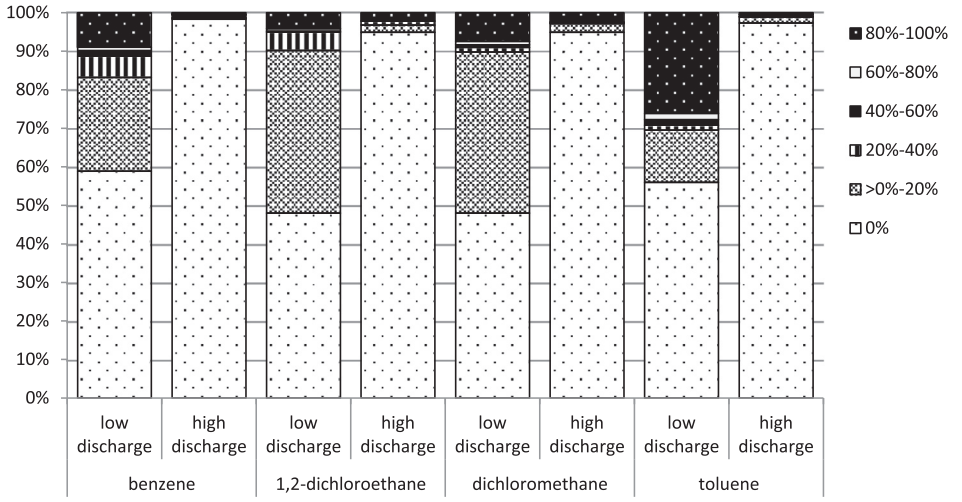


Fig. 3. Relative contribution from Dutch IWTP to the concentration in SWU compared to the contribution from abroad, shading indicates the impact expressed as percentage by Dutch IWTP.

Table 4
Percentage of impactful IWTP with relative impact factor (rIF) >0,1, based on averaged impact of the modelled six contaminants.

Discharge condition	Percentage of impactful IWTP	% of total Dutch TOC emission	% cumulative impact Dutch IWTP by the impactful IWTP
Low	8,2%	44,7	99,9
High	5,5%	28,6	100

Statistics). A third of the total capacity of Dutch STP (24.2 million inhabitant equivalents) is used to treat industrial waste waters. The volume of industrial waste water treated via STP, equalling 7.9 million inhabitant equivalents, is in the same range as industrial waste water directly treated via Dutch IWTP equalling 13.8 million inhabitant equivalents (data by CBS Statline, 2017 and personal communication Kees Baas Central Bureau for Statistics). However, as NACE-codes of the industries of which the waste waters are treated by the STP are not centrally registered, the STP effluent data in the Watson database cannot be generalized for modelling purposes.

Incidental high industrial releases are not explicitly modelled, as the approach here is based on the yearly averages in which emission data are reported. Aqueous concentrations as a result of incidental high IWTP emissions will in case of the incidents be temporarily higher than modelled. Also short periods of extremely low river discharge are not covered, as 3 monthly averages periods are used in the modelling. These short periods of extremely low discharge also will temporarily result in higher concentrations than currently modelled.

4.2. European studies on industrial chemical emission to surface water

Currently approximately industrial substances are registered under REACH regulation (Registration, Evaluation and Authorization of CHemicals), 15% of which are produced in volumes over 1.000 t per year. For the majority of registered compounds the tonnage is either confidential, or the compounds are only registered as being for intermediate use. REACH focuses on the PBT criteria (persistent, bioaccumulation and toxic). From a drinking water perspective, the persistent, mobile and toxic organic compounds (PMOC) are more relevant (Reemtsma et al., 2016, Sjerps et al., 2016).

Several cases based on analytical measurements of industrial effluents are described. In Belgian and Italian textile industrial effluents octyl- and nonylphenol, their ethoxylates and carboxylates were measured (Loos et al., 2007). The use and production of nonylphenol ethoxylates have been banned in EU countries. Perfluorinated compounds (PFCs) are detected in numerous industrial waste water treatment plants during the last ten years (Arvaniti and Stasinakis, 2015; Castiglioni et al., 2015; Gebbink et al., 2017; Loos et al., 2013). In Italy, STP effluent with a large proportion of textile and furniture industry wastewater contained short and long chained perfluorinated carboxylic acids and perfluorooctanoic acid (PFOA) in concentrations varying from 37 to 786 ng/L (Castiglioni et al., 2015). In industrial waste water treatment plants, Loos et al. (2013) detected PFOA at the highest median concentration levels (12.9 ng/L), followed by other perfluorinated compounds (PFOS, PFHxA, PFHpA, PFHxS, PFDA, and PFNA). The PFOA replacer GenX was detected downstream from a chemical production plant in The Netherlands up to concentrations of 812 ng/L (Gebbink et al., 2017), the same pattern was followed by 11 emerging PFASs. The Dutch Watson database mentions STP effluent concentrations up to 0,74 μg/L for PFOS and 0,062 μg/L for PFOA. Estrogenic activity in industrial effluents is found widely across Europe, in particular in effluents from textile, chemical or pharmaceutical manufacture industries (Eggen et al., 2003), related to the presence of nonyphenol, nonylphenol ethoxylates, hydroxyphenyl hexanoic acid or bisphenol A. Also Van der Linden et al. (2008) found high activities of estrogen (ERα), progesterone (PR), glucocorticoid (GR) and androgen (AR) in the industrial effluent compared to STP effluents in the Netherlands, which were found to be partly explained by synthetic hormones up to a concentration of 247 ng/L for prednisolone (Schriks et al., 2010). In Croatian pharmaceutical industry effluents, veterinary antibiotics (fluoroquinolones, trimethoprim, sulfonamides and tetracyclines) ranged up to approx. 200 μg/L (Bielen et al., 2017). In a German study on paper industrial effluents photoinitiators, ink and thermal paper constituents were present such as Bisphenol A up to 6,1 μg/L (Dsikowitzky et al., 2015).

However, this information remains sparse and further confirms that systematic information on composition of industrial effluents is lacking.

4.3. Suggestions for regulation regarding industrial aqueous emissions

A public and register of all chemicals and by-products produced per industrial site, and thus possibly emitted via the industrial wastewaters, by CAS numbers is currently not available in the Netherlands nor other European member states. Such a register, ideally including production volume ranges at the site, would be informative to other water users in the river basin, to focus their monitoring, modelling, risk assessment and risk management efforts. It would also be in line with the Aarhus convention as implemented in the EU in Directive 2003/4/EC on public access to environmental information.

Available information in the public REACH dossiers on all produced chemicals and by-products in an industrial process is to be implemented by the competent authorities in the licensing of industrial discharges. In the REACH dossiers restrictions can be prescribed, e.g., a product should not be allowed to enter water courses, or precautionary measures should be taken to prevent accidental spillages. Currently this implementation of REACH restrictions is not systematically taken into account during the licensing process (S.I. A). It would be helpful if in the public REACH dossiers information is provided on the NACE-codes of the industrial sectors where the chemical is produced or used, as currently only total European tonnage bands are given in the REACH dossiers.

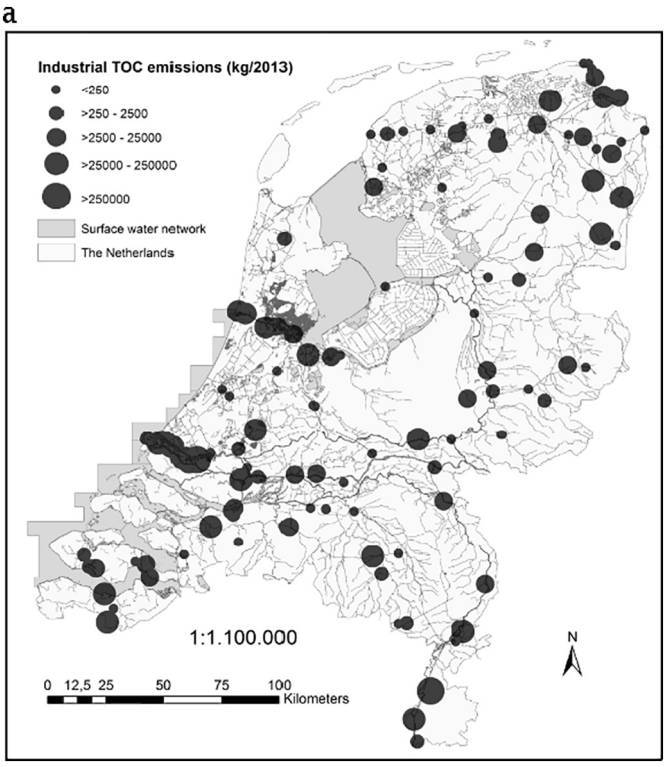


Fig. 4. Overview of a) all 182 Dutch IWTP with variable TOC emissions over the year 2013, and IWTP having a relative impact higher than 0,1 b) for the six modelled industrial chemicals and c) for the averaged chemicals at low discharge.

b

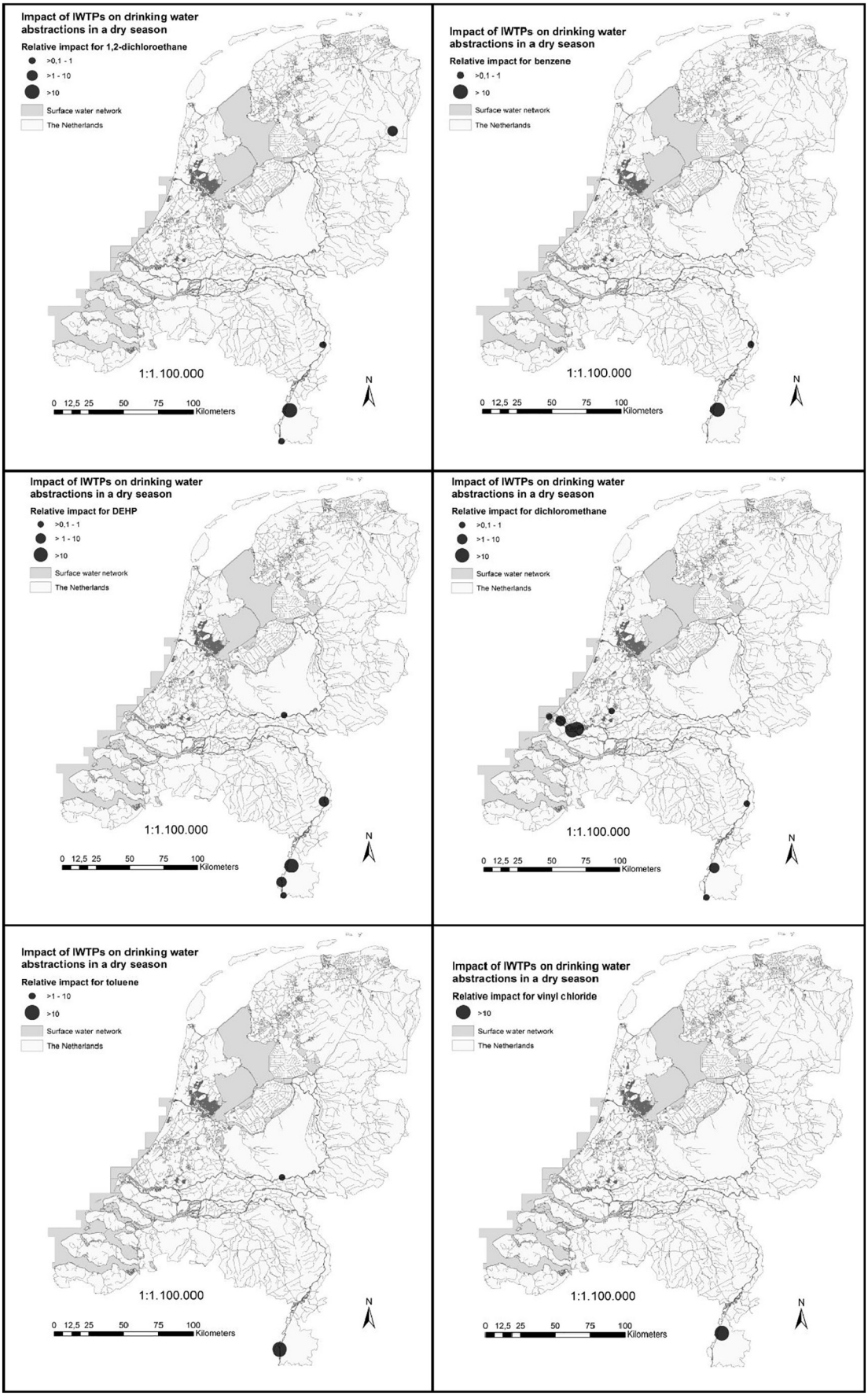


Fig. 4 (*continued*).

C

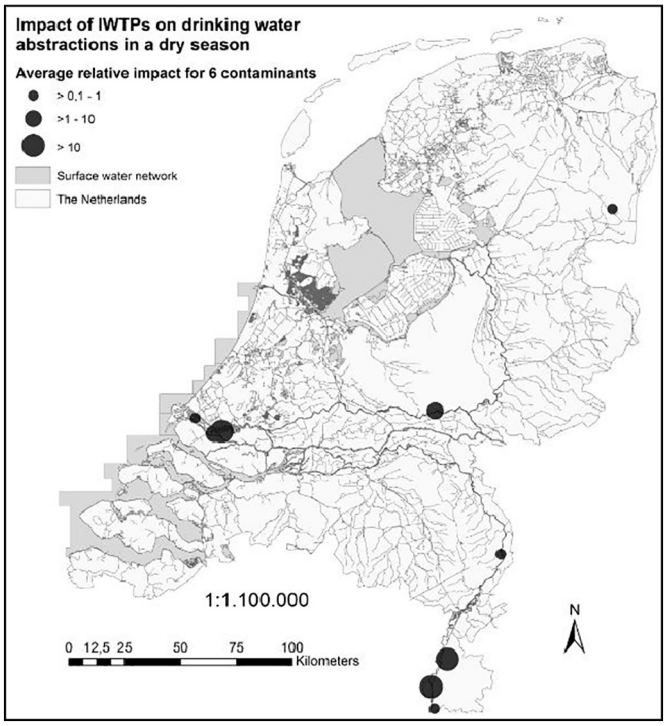


Fig. 4 (*continued*).

When industry applies for a license to discharge their (treated) wastewater to the aqueous environment, they are obliged to provide specific information with regard to the relevant chemicals and by-products produced and the production processes to the competent authority. In the Netherlands, these are the national water authority Rijkswaterstaat or the regional water authorities for industrial discharges via privately owned IWTP, or the provinces for indirect industrial discharges via public STP. This specific information is then often translated in more general terms in the license, in accordance to the IED and associated 'Best available techniques reference documents' (BREFs).

In the Netherlands a limited general location-specific risk assessment is compulsory, this discharge test assesses the impact of the discharge upon the receiving surface water (http://www.immissietoets.nl/#version=nl-en). However, a refined location-specific risk assessment may be needed for the license, as the risks of the industrial discharge may influence specific downstream river basin water uses, such as drinking water production, food production, nature or recreation. Location-specific risks of an industrial discharge further depend on the hydrological situation including climate variability, and on contamination with other available sources. This location-specific risk assessment cannot be fulfilled by general legislation such as REACH, the Water Framework Directive (WFD, 2000/60/EC) or the Drinking Water Directive (DWD, 98/83/EC).

Table 5
Number of SWU with drinking water function and abstraction volumes influenced by aqueous industrial emissions; based on all six model compounds during low discharge.

	Number of SWU with drinking water function	Influenced by IWTP	Total abstraction volume (million m^3/y)	Abstraction volume influenced by IWTP (million m^3/year)
Surface water	9	8	416	415 (99%)
Bank filtrate	20	13	108	62 (57%)
Groundwater	180	18	739	114 (15%)
Total	209	39	1262	415 (32%)

Currently, there is hardly any obligation for the industry or IWTP owner to report on CECs emitted, unless they are emitted in relevant concentrations. The term "relevant" has a subjective tone to it, and is also influenced by practical and financial constraints with respect to the monitoring of CECs. For example in the Netherlands the competent water authority, in cooperation with the licensee, is responsible for the compliance monitoring of the industrial effluent and thus has an interest to keep monitoring costs low. Therefore compliance monitoring is often only targeted on a number of benchmark substances, which in the permitting phase are assessed to cover most of the substances emitted. Within this approach, relevant substances will be missed in the regular compliance monitoring. However, nowadays monitoring techniques like high-resolution screening techniques and whole effluent toxicity assessment have evolved in such a way that emittance of a broad set of relevant chemicals and their effects can be followed (Hollender et al., 2017; Chapman, 2000, 2007).

For drinking water and drinking water sources, in the Netherlands a signalling parameter for 'other anthropogenic substances' has been introduced of respectively 1 and 0,1 μg/L as a top-up on the EU Drinking Water Directive and EU Water Framework Directive. When a concentration of a synthetic chemical in drinking water or drinking water sources exceeds this signalling parameter, further research is carried out on the environmental and health risks, the sources and removal efficiencies during water treatment. This signalling value draws attention of water managers and drinking water utilities to the presence of the chemicals, also resulting in evaluation of their risks (Baken et al., in press). Such a 'signalling value' for anthropogenic substances could also be implemented with regards to industrial effluents to further increase awareness of emitting industries.

4.4. Further mitigation of emissions by IWTP

In this modelling study a limited number of IWTP, typically serving plastic, paper, petroleum or basic chemicals industry, drive the impact with regard to drinking water production. This implies that mitigation measures can be taken cost-efficiently, when extra treatment technologies are placed at the IWTP with high impact. Waste water treatment technologies generally are available (Van Wezel et al., 2017). Treatment technologies are well established for more general classical water quality parameters (Evrard et al., 2016; Polders et al., 2012), and are laid down in the BREF documents established for the different industrial sectors (available via http://eippcb.jrc.ec.europa.eu/reference/). Conventional biological treatment is most widely used, which removes hydrophobic and well biodegradable substances. More advanced technologies are available for treatment of industrial effluents to remove specific chemicals, examples are electrochemical methods (Brillas and Martínez-Huitle, 2015; Niu et al., 2016), membranes (Caldwell et al., 2016; Kanakaraju et al., 2014) or electrocoagulation (Dasgupta et al., 2015; Khandegar and Saroha, 2013), TiO_2 photocatalytic methods, advanced oxidation processes. It depends on the chemicals to be removed and their physical-chemical properties, the industrial effluent matrix involved and the particular operational settings, which treatment technology or combination of treatment techniques gives the highest removal efficiencies (Fischer et al., 2017; Van Wezel et al., 2017). Besides treatment technologies, the BREF documents describe strategies for cleaner production to prevent pollution and reduce risks.

5. Conclusion

- A public register of all chemicals and by-products produced and used per industrial site is currently not available in European member states. Such a register would be in line with the Aarhus convention and informative to water users in the river basin.
- Systematic information on concentrations in IWTP effluents for many chemicals of interest for drinking water production is lacking, also for relevant chemical-intensive sectors. Of 28 selected industrial

chemicals of interest, only for bis(2-ethylhexyl)phthalate (DEHP) TOC normalized emissions based on the *E*-PRTR data can be derived. Therefore, as surrogate parameters, data from the European Pollutant Release and Transfer Register were used and coupled to TOC data on Dutch IWTP. These emissions were used as input to a detailed hydrological model under two extreme river discharge conditions.

- Predicted surface water concentrations for benzene, toluene, dichloromethane and 1,2-dichloroethane vary by over 3 orders of magnitude over the surface water bodies. Maximum predicted concentrations are in the same range as predicted earlier for some pharmaceuticals.
- Measured concentrations often exceed predicted concentrations, explained since both indirect industrial emissions via STP and releases during the use and waste life cycle phase of industrial chemicals were not included in the model.
- From all Dutch 182 IWTP a limited number has according to our model approach using surrogate parameters a high impact on surface water with a drinking water function. These IWTP typically serve plastic, paper, petroleum or basic chemicals industry. In total a third of the abstracted water for drinking water production is affected by the IWTP. The results of the modelling approach are uncertain due to a lack of information, these uncertainties could be diminished when industrial emission data on more chemicals would be public.
- Available information in the public REACH dossiers, such as restrictions for safe use or precautionary measures, is currently not systematically implemented by the competent authorities in the licensing of industrial discharges.

Acknowledgements

Nanette van Duijnhoven on behalf of the Dutch Pollutant Release and Transfer Register, Kees Baas from the Dutch Central Bureau of Statistics, Rob Berbee from Rijkswaterstaat and Idzi Hubrecht from the Flanders Environment Agency are acknowledged for their help regarding statistics on industrial emissions. RIWA and the Dutch drinking water utilities producing drinking water from surface water, are acknowledged for their support for this project. The Dutch Drinking water utilities shared their data on raw drinking water abstraction, the Dutch provinces shared their data on 25-year ground water protection zones, and RIWA shares their data (www.riwa.org) on surface water quality. The Dutch ministry of Infrastructure and Environment provided the national application of the WFD-explorer 2.0. The work is co-funded by the European Commission, FP7 project SOLUTIONS, contract number 603437.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.05.325.

References

Aldekoa, J., Medici, C., Osorio, V., Pérez, S., Marcé, R., Barceló, D., Francés, F., 2013. Modelling the emerging pollutant diclofenac with the GREAT-ER model: application to the Llobregat River Basin. J. Hazard. Mater. 263, 207–213.

Arvaniti, O.S., Stasinakis, A.S., 2015. Review on the occurrence, fate and removal of perfluorinated compounds during wastewater treatment. Sci. Total Environ. 524–525, 81–92.

Baken, K., Kolkman, A., Van Diepenbeek, P., Ketelaars, H., Van Wezel, A., 2016. Signallling 'Other Antropogenic Susbstances', and Then? The Pyrazole Case. (H_2O online, September 2016 In Dutch).

Baken, K.A., Sjerps, R.M.A., Schriks, M., Van Wezel, A.P., 2018. Toxicological relevance and Threshold of Toxicological Concern (TTC) for drinking water relevant contaminants of emerging concern. Environ. Int. (accepted).

Baldwin, A.K., Corsi, S.R., De Cicco, L.A., Lenaker, P.L., Lutz, M.A., Sullivan, D.J., Richards, K.D., 2016. Organic contaminants in Great Lakes tributaries: prevalence and potential aquatic toxicity. Sci. Total Environ. 554–555, 42–52.

Bernhardt, E.S., Rosi, E.J., Gessner, M.O., 2017. Synthetic chemicals as agents of global change. Front. Ecol. Environ. 15, 84–90.

Bielen, A., Šimatović, A., Kosić-Vukšić, J., Senta, I., Ahel, M., Babić, S., Jurina, T., González Plaza, J.J., Milaković, M., Udiković-Kolić, N., 2017. Negative environmental impacts of antibiotic-contaminated effluents from pharmaceutical industries. Water Res. 126, 79–87.

Boiteux, V., Dauchy, X., Bach, C., Colin, A., Hemard, J., Sagres, V., Rosin, C., Munoz, J.F., 2017. Concentrations and patterns of perfluoroalkyl and polyfluoroalkyl substances in a river and three drinking water treatment plants near and far from a major production source. Sci. Total Environ. 583, 393–400.

Brillas, E., Martínez-Huitle, C.A., 2015. Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review. Appl. Catal. B Environ. 166–167, 603–643.

Caldwell, D.J., Mertens, B., Kappler, K., Senac, T., Journel, R., Wilson, P., Meyerhoff, R.D., Parke, N.J., Mastrocco, F., Mattson, B., Murray-Smith, R., Dolan, D.G., Straub, J.O., Wiedemann, M., Hartmann, A., Finan, D.S., 2016. A risk-based approach to managing active pharmaceutical ingredients in manufacturing effluent. Environ. Toxicol. Chem. 35, 813–822.

Castiglioni, S., Valsecchi, S., Polesello, S., Rusconi, M., Melis, M., Palmiotto, M., Manenti, A., Davoli, E., Zuccato, E., 2015. Sources and fate of perfluorinated compounds in the aqueous environment and in drinking water of a highly urbanized and industrialized area in Italy. J. Hazard. Mater. 282, 51–60.

CBS Statline, 2017. Emission to Water; Type of Discharge, Sector of Industry. Statistics Netherlands (CBS) (In Dutch). http://statline.cbs.nl/Statweb/publication/?DM=SLNL&PA=81353ned&D1=0-23,26-28&D2=0-2&D3=0,4-5,17,19-21,28,31-34,37&D4=l&VW=T.

Chapman, P.M., 2000. Whole effluent toxicity testing—usefulness, level of protection, and risk assessment. Environ. Toxicol. Chem. 19, 3–13.

Chapman, P.M., 2007. Determining when contamination is pollution - weight of evidence determinations for sediments and effluents. Environ. Int. 33, 492–501.

CHEMLIST, 2018. Regulated Chemicals Listing.Available via. http://support.cas.org/content/regulated-chemicals (visited May 24, 2018).

Coppens, L.J.C., Van Gils, J., Ter Laak, T., Raterman, B., Van Wezel, A., 2015. Towards spatially smart mitigation of human pharmaceuticals in surface waters: defining impact of sewage treatment plants on susceptible functions. Water Res. 81, 356–365.

Cristale, J., García, V., Barata, C., Lacorte, S., 2013a. Priority and emerging flame retardants in rivers: occurrence in water and sediment, *Daphnia magna* toxicity and risk assessment. Environ. Int. 59, 232–243.

Cristale, J., Katsoyiannis, A., Sweetman, A.J., Jones, K.C., Lacorte, S., 2013b. Occurrence and risk assessment of organophosphorus and brominated flame retardants in the River Aire (UK). Environ. Pollut. 179, 194–200.

Dasgupta, J., Sikder, J., Chakraborty, S., Curcio, S., Drioli, E., 2015. Remediation of textile effluents by membrane based treatment techniques: a state of the art review. J. Environ. Manag. 147, 55–72.

De Hoogh, C.J., Wagenvoort, A.J., Jonker, F., Van Leerdam, J.A., Hogenboom, A.C., 2006. HPLC-DAD and Q-TOF MS techniques identify cause of *Daphnia* biomonitor alarms in the River Meuse. Environ. Sci. Technol. 40, 2678–2685.

Delpla, I., Jung, A.V., Baures, E., Clement, M., Thomas, O., 2009. Impacts of climate change on surface water quality in relation to drinking water production. Environ. Int. 35, 1225–1233.

Derden, A., Huybrechts, D., 2013. Brominated flame retardants in textile wastewater: reducing Deca-BDE using best available techniques. J. Clean. Prod. 53, 167–175.

Ding, J., Shen, X., Liu, W., Covaci, A., Yang, F., 2015. Occurrence and risk assessment of organophosphate esters in drinking water from Eastern China. Sci. Total Environ. 538, 959–965.

Dsikowitzky, L., Botalova, O., Illgut, S., Bosowski, S., Schwarzbauer, J., 2015. Identification of characteristic organic contaminants in wastewaters from modern paper production sites and subsequent tracing in a river. J. Hazard. Mater. 300, 254–262.

Eggen, R.I.L., Bengtsson, B.E., Bowmer, C.T., Gerritsen, A.A.M., Gibert, M., Hylland, K., Johnson, A.C., Leonards, P., Nakari, T., Norrgren, L., Sumpter, J.P., Suter, M.J.F., Svenson, A., Pickering, A.D., 2003. Search for the evidence of endocrine disruption in the aquatic environment: lessons to be learned from joint biological and chemical monitoring in the European project COMPREHEND. Pure Appl. Chem. 75, 2445–2450.

Evrard, D., Laforest, V., Villot, J., Gaucher, R., 2016. Best available technique assessment methods: a literature review from sector to installation level. J. Clean. Prod. 121, 72–83.

Fan, F.M., Fleischmann, A.S., Collischonn, W., Ames, D.P., Rigo, D., 2015. Large-scale analytical water quality model coupled with GIS for simulation of point sourced pollutant discharges. Environ. Model. Softw. 64, 58–71.

Fischer, A., Ter Laak, T., Bronders, J., Desmet, N., Christoffels, E., Van Wezel, A., Van der Hoek, J.P., 2017. Decision support for water quality management of contaminants of emerging concern. J. Environ. Manag. 193, 360–372.

Gebbink, W.A., Van Asseldonk, L., Van Leeuwen, S.P.J., 2017. Presence of emerging per- and polyfluoroalkyl substances (PFASs) in river and drinking water near a fluorochemical production plant in the Netherlands. Environ. Sci. Technol. 51, 11057–11065.

Halden, R.U., 2015. Epistemology of contaminants of emerging concern and literature meta-analysis. J. Hazard. Mater. 282, 2–9.

Hollender, J., Schymanski, E.L., Singer, H.P., Ferguson, P.L., 2017. Nontarget screening with high resolution mass spectrometry in the environment: ready to go? Environ. Sci. Technol. 51, 11505–11512.

Howard, P.H., 1991. Handbook of Environmental Degradation Rates. CRC Press.

Hu, X.C., Andrews, D.Q., Lindstrom, A.B., Bruton, T.A., Schaider, L.A., Grandjean, P., Lohmann, R., Carignan, C.C., Blum, A., Balan, S.A., Higgins, C.P., Sunderland, E.M., 2016. Detection of poly- and perfluoroalkyl substances (PFASs) in U.S. drinking water linked to industrial sites, military fire training areas, and wastewater treatment plants. Environ. Sci. Technol. Lett. 3, 344–350.

Ippolito, A., Kattwinkel, M., Rasmussen, J.J., Schäfer, R.B., Fornaroli, R., Liess, M., 2015. Modeling global distribution of agricultural insecticides in surface waters. Environ. Pollut. 198, 54–60.

Kanakaraju, D., Glass, B.D., Oelgemöller, M., 2014. Titanium dioxide photocatalysis for pharmaceutical wastewater treatment. Environ. Chem. Lett. 12, 27–47.
Kapo, K.E., Mcdonough, K., Federle, T., Dyer, S., Vamshi, R., 2015. Mixing zone and drinking water intake dilution factor and wastewater generation distributions to enable probabilistic assessment of down-the-drain consumer product chemicals in the U.S. Sci. Total Environ. 518–519, 302–309.
Kehrein, N., Berlekamp, J., Klasmeier, J., 2015. Modeling the fate of down-the-drain chemicals in whole watersheds: new version of the GREAT-ER software. Environ. Model. Softw. 64, 1–8.
Khandegar, V., Saroha, A.K., 2013. Electrocoagulation for the treatment of textile industry effluent - a review. J. Environ. Manag. 128, 949–963.
Kiss, A., Fries, E., 2009. Occurrence of benzotriazoles in the rivers Main, Hengstbach, and Hegbach (Germany). Environ. Sci. Pollut. Res. Int. 16, 702–710.
Kosaka, K., Asami, M., Ohkubo, K., Iwamoto, T., Tanaka, Y., Koshino, H., Echigo, S., Akiba, M., 2014. Identification of a New N-nitrosodimethylamine precursor in sewage containing industrial effluents. Environ. Sci. Technol. 48, 11243–11250.
Kuroda, K., Itten, R., Kovalova, L., Ort, C., Weissbrodt, D.G., Mcardell, C.S., 2016. Hospital-use pharmaceuticals in Swiss waters modeled at high spatial resolution. Environ. Sci. Technol. 50, 4742–4751.
Lapworth, D.J., Baran, N., Stuart, M.E., Ward, R.S., 2012. Emerging organic contaminants in groundwater: a review of sources, fate and occurrence. Environ. Pollut. 163, 287–303.
Lee, I.S., Sim, W.J., Kim, C.W., Chang, Y.S., Oh, J.E., 2011. Characteristic occurrence patterns of micropollutants and their removal efficiencies in industrial wastewater treatment plants. J. Environ. Monit. 13, 391–397.
Lindim, C., Cousins, I.T., Van Gils, J., 2015. Estimating emissions of PFOS and PFOA to the Danube River catchment and evaluating them using a catchment-scale chemical transport and fate model. Environ. Pollut. 207, 97–106.
Lindim, C., Van Gils, J., Cousins, I.T., 2016. A large-scale model for simulating the fate & transport of organic contaminants in river basins. Chemosphere 144, 803–810.
Loos, R., Hanke, G., Umlauf, G., Eisenreich, S.J., 2007. LC–MS–MS analysis and occurrence of octyl- and nonylphenol, their ethoxylates and their carboxylates in Belgian and Italian textile industry, waste water treatment plant effluents and surface waters. Chemosphere 66, 690–699.
Loos, R., Gawlik, B.M., Locoro, G., Rimaviciute, E., Contini, S., Bidoglio, G., 2009. EU-wide survey of polar organic persistent pollutants in European river waters. Environ. Pollut. 157, 561–568.
Loos, R., Locoro, G., Comero, S., Contini, S., Schwesig, D., Werres, F., Balsaa, P., Gans, O., Weiss, S., Blaha, L., Bolchi, M., Gawlik, B.M., 2010a. Pan-European survey on the occurrence of selected polar organic persistent pollutants in ground water. Water Res. 44, 4115–4126.
Loos, R., Locoro, G., Contini, S., 2010b. Occurrence of polar organic contaminants in the dissolved water phase of the Danube River and its major tributaries using SPE-LC-MS(2) analysis. Water Res. 44, 2325–2335.
Loos, R., Carvalho, R., António, D.C., Comero, S., Locoro, G., Tavazzi, S., Paracchini, B., Ghiani, M., Lettieri, T., Blaha, L., Jarosova, B., Voorspoels, S., Servaes, K., Haglund, P., Fick, J., Lindberg, R.H., Schwesig, D., Gawlik, B.M., 2013. EU-wide monitoring survey on emerging polar organic contaminants in wastewater treatment plant effluents. Water Res. 47, 6475–6487.
Luo, Y., Guo, W., Ngo, H.H., Nghiem, L.D., Hai, F.I., Zhang, J., Liang, S., Wang, X.C., 2014. A review on the occurrence of micropollutants in the aquatic environment and their fate and removal during wastewater treatment. Sci. Total Environ. 473-474, 619–641.
Malaj, E., Von Der Ohe, P.C., Grote, M., Kühne, R., Mondy, C.P., Usseglio-Polatera, P., Brack, W., Schäfer, R.B., 2014. Organic chemicals jeopardize the health of freshwater ecosystems on the continental scale. Proc. Natl. Acad. Sci. U. S. A. 111, 9549–9554.
Mank, R., Hoff, E., 2017. Raadsinformatie Inzake Bodemrapporten PFOA (Letter by the city of Dordrecht SO/1861552 in Dutch).
Niu, J., Li, Y., Shang, E., Xu, Z., Liu, J., 2016. Electrochemical oxidation of perfluorinated compounds in water. Chemosphere 55, 526–538.
Ozturk, E., Koseoglu, H., Karaboyaci, M., Yigit, N.O., Yetis, U., Kitis, M., 2016. Sustainable textile production: cleaner production assessment/eco-efficiency analysis study in a textile mill. J. Clean. Prod. 138, 248–263.
Petrovic, M., Ginebreda, A., Acuña, V., Batalla, R.J., Elosegi, A., Guasch, H., de Alda, M.L., Marcé, R., Muñoz, I., Navarro-Ortega, A., Navarro, E., Vericat, D., Sabater, S., Barceló, D., 2011. Combined scenarios of chemical and ecological quality under water scarcity in Mediterranean rivers. TrAC 30, 1269–1278.
Polders, C., Van Den Abeele, L., Derden, A., Huybrechts, D., 2012. Methodology for determining emission levels associated with the best available techniques for industrial waste water. J. Clean. Prod. 29-30, 113–121.
Rebelo, A., Ferra, I., Gonçalves, I., Marques, A.M., 2014. A risk assessment model for water resources: releases of dangerous and hazardous substances. J. Environ. Manag. 140, 51–59.
Reemtsma, T., Berger, U., Arp, H.P.H., Gallard, H., Knepper, T.P., Neumann, M., Quintana, J.B., Voogt, P.D., 2016. Mind the gap: Persistent and Mobile Organic Compounds - Water contaminants that slip through. Environ. Sci. Technol. 50, 10308–10315.
Richardson, S.D., Ternes, T.A., 2014. Water analysis: Emerging contaminants and current issues. Anal. Chem. 86, 2813–2848.
Rider, C.V., Dourson, M.L., Hertzberg, R.C., Mumtaz, M.M., Price, P.S., Simmons, J.E., 2012. Incorporating nonchemical stressors into cumulative risk assessments. Toxicol. Sci. 127, 10–17.
RIWA, 2017. Yearly report 2016. The Meuse Available via. www.riwa.org (In Dutch).
Roex, E.W.M., 2003. TEB praktijkonderzoek. Deel T-1: Evaluatierapport Meten TEB-Parameters. Ministerie van Verkeer en Waterstaat. Directoraat-Genraal Rijkswaterstaat, Lelystad.
Ruff, M., Mueller, M.S., Loos, M., Singer, H.P., 2015. Quantitative target and systematic non-target analysis of polar organic micro-pollutants along the river Rhine using high-resolution mass-spectrometry – identification of unknown sources and compounds. Water Res. 87, 145–154.
Salgueiro-González, N., Turnes-Carou, I., Besada, V., Muniategui-Lorenzo, S., López-Mahía, P., Prada-Rodríguez, D., 2015. Occurrence, distribution and bioaccumulation of endocrine disrupting compounds in water, sediment and biota samples from a European river basin. Sci. Total Environ. 529, 121–130.
Schlüsener, M.P., Kunkel, U., Ternes, T.A., 2015. Quaternary triphenylphosphonium compounds: a new class of environmental pollutants. Environ. Sci. Technol. 49, 14282–14291.
Schriks, M., Van Leerdam, J.A., Van der Linden, S.C., Van der Burg, B., Van Wezel, A.P., De Voogt, P., 2010. High-resolution mass spectrometric identification and quantification of glucocorticoid compounds in various wastewaters in the Netherlands. Environ. Sci. Technol. 44, 4766–4774.
Schwarzenbach, R.P., Escher, B.I., Fenner, K., Hofstetter, T.B., Johnson, C.A., Von Gunten, U., Wehrli, B., 2006. The challenge of micropollutants in aquatic systems. Science 313, 1072–1077.
Sjerps, R.M.A., Vughs, D., Van Leerdam, J.A., Ter Laak, T.L., Van Wezel, A.P., 2016. Data-driven prioritization of chemicals for various water types using suspect screening LC-HRMS. Water Res. 93, 254–264.
Sjerps, R.M.A., Ter Laak, T.L., Zwolsman, G., 2017. Projected impact of climate change and chemical emissions on the water quality of the European rivers Rhine and Meuse: a drinking water perspective. Sci. Total Environ. 601–602, 1682–1694.
Sörme, L., Palm, V., Finnveden, G., 2016. Using E-PRTR data on point source emissions to air and water—first steps towards a national chemical footprint. Environ. Impact Assess. Rev. 56, 102–112.
Stepien, D.K., Püttmann, W., 2014. Source identification of high glyme concentrations in the Oder River. Water Res. 54, 307–317.
Tas, D.O., Pavlostathis, S.G., 2014. Occurrence, toxicity, and biotransformation of Pentachloronitrobenzene and Chloroanilines. Crit. Rev. Environ. Sci. Technol. 44, 473–518.
Tran, N.H., Reinhard, M., Gin, K.Y.H., 2018. Occurrence and fate of emerging contaminants in municipal wastewater treatment plants from different geographical regions - a review. Water Res. 133, 182–207.
Van der Linden, S.C., Heringa, M.B., Man, H.Y., Sonneveld, E., Puijker, L.M., Brouwer, A., Van der Burg, B., 2008. Detection of multiple hormonal activities in wastewater effluents and surface water, using a panel of steroid receptor CALUX bioassays. Environ. Sci. Technol. 42, 5814–5820.
Van Leerdam, J.A., Vervoort, J., Stroomberg, G., De Voogt, P., 2014. Identification of unknown microcontaminants in Dutch river water by liquid chromatography-high resolution mass spectrometry and nuclear magnetic resonance spectroscopy. Environ. Sci. Technol. 48, 12791–12799.
Van Vliet, M.T.H., Zwolsman, J.J.G., 2008. Impact of summer droughts on the water quality of the Meuse river. J. Hydrol. 353, 1–17.
Van Wezel, A.P., Ter Laak, T.L., Fischer, A., Bäuerlein, P.S., Munthe, J., Posthuma, L., 2017. Operationalising solutions-focused risk assessment; mitigation options for chemicals of emerging concern in surface waters. Environ. Sci.: Water Res. Technol. 3, 403–414.
Velzeboer, I., 2014. Threats for the water quality of Dunea; Evaluation and update 2014. The Water Laboratory, Haarlem (In Dutch).
Vethaak, A.D., Lahr, J., Schrap, S.M., Belfroid, A.C., Rijs, G.B.J., Gerritsen, A., de Boer, J., Bulder, A.S., Grinwis, G.C.M., Kuiper, R.V., Legler, J., Murk, T.A.J., Peijnenburg, W., Verhaar, H.J.M., de Voogt, P., 2005. An integrated assessment of estrogenic contamination and biological effects in the aquatic environment of The Netherlands. Chemosphere 59, 319–331 (511–524).
Von der Ohe, P.C., Dulio, V., Slobodnik, J., De Deckere, E., Kühne, R., Ebert, R.U., Ginebreda, A., De Cooman, W., Schüürmann, G., Brack, W., 2011. A new risk assessment approach for the prioritization of 500 classical and emerging organic microcontaminants as potential river basin specific pollutants under the European Water Framework Directive. Sci. Total Environ. 409, 2064–2077.
Wilson, M.P., Schwarzman, M.R., 2009. Toward a new U.S. chemicals policy: rebuilding the foundation to advance new science, green chemistry, and environmental health. Environ. Health Perspect. 117, 1202–1209.
Zijp, M.C., Posthuma, L., Wintersen, A., Devilee, J., Swartjes, F.A., 2016. Definition and use of solution-focused sustainability assessment: a novel approach to generate, explore and decide on sustainable solutions for wicked problems. Environ. Int. 91, 319–331.