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Feasibility and potential of a water quality map of the Netherlands BTO | October 2019

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BTO

Feasibility and potential of a water quality map of the Netherlands

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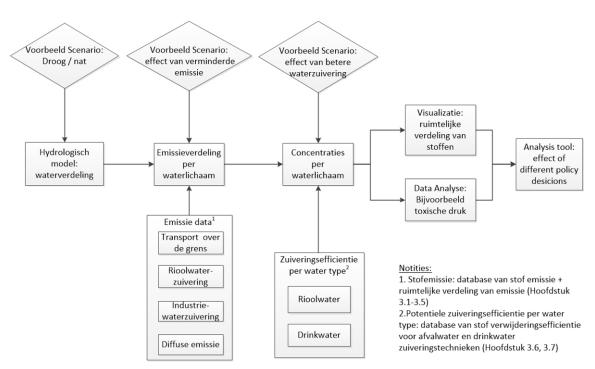
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BTO Management samenvatting

Kwaliteit beschikbare data grootste struikelblok voor creatie Nederlandse waterkwaliteitskaart

Auteur(s) Tessa Pronk, Alex Hockin, Luc Hornstra, Bernard Raterman, Dirk Vries en Geertje Pronk

De verspreiding van diverse soorten verontreinigingen in Nederlands oppervlaktewater kan worden gemodelleerd met behulp van de *KRW-Verkenner*. Op deze manier kunnen verschillende scenario's worden doorgerekend en gecombineerd tot een waterkwaliteitskaart. In dit rapport stellen we vast of data over verontreinigingsbronnen (industriële en rioolwaterzuiveringsinstallaties, landbouw en grensoverschrijdende rivieren) met behulp van een model leiden tot een goede kaart van de Nederlandse waterkwaliteit. Een dergelijke kaart kan worden ingezet om de impact van verontreinigingen ruimtelijk weer te geven. Dit kan bijdragen aan het vormgeven van beleid om verontreinigingen in het water terug te dringen. Op dit moment blijkt de beschikbaarheid, compleetheid, en kwaliteit van de beschikbare gegevens de grootste horde te zijn voor het maken van een complete en accurate waterkwaliteitskaart voor Nederland. Voor de individuele emissies van verontreinigingen via rioolwaterzuiveringsinstallaties en grensoverschrijdende rivieren was het wel mogelijk om met de beschikbare data een waterkwaliteit modellering te doen, deze zijn klaar voor verder analyse op basis van concrete vraagstellingen.



Schematisch overzicht van de Waterkwaliteitskaart

Schematisch overzicht van de waterkwaliteitskaart met scenario's en emissies

Belang: verontreinigingen aanpakken via meer inzicht in bronnen en verspreiding

Emissies van organische, microbiële en chemische vervuilingen uit verschillende bronnen verontreinigen het Nederlandse oppervlaktewater. Om deze verontreinigingen goed te kunnen aanpakken, is kennis nodig over de emissies en de verspreiding. Scenario studies van emissies en potentiele impact van maatregelen op de waterkwaliteit kunnen bijdragen aan het effectief aanpakken van verontreinigingen.

Aanpak: KRW-Verkenner en data combineren

tot een realistische waterkwaliteitskaart Om een realistische waterkwaliteitskaart te maken van verontreinigingen zoals chemische stoffen, micro-organismen en antibioticaresistentie via verschillende emissieroutes zijn gegevens over emissie en zuiveringsefficiëntie van verontreinigingen gecombineerd met een flexibele berekeningsmethode aan de hand van een hydrologisch model van Nederland (KRW-Verkenner). De basis is een matrix met een groot aantal emissiebronnen en eindpunten voor de verontreiniging, met een stofbalans. Zo kan grootschalig worden gerekend aan de verspreiding van verontreinigingen over Nederlandse oppervlaktewateren. Dit geeft inzicht in zowel de transportduur als de geschatte afbraak gedurende het transport. Het model werkt met de emissieroutes (i) rioolwaterzuiverings-installaties, (ii) industriewaterzuiverings-installaties, (iii) diffuse emissie uit landbouw en (iv) bijdragen uit grensoverschrijdende rivieren. Deze routes worden gecombineerd met informatie over de zuiveringsefficiëntie van de waterzuiveringen.

Resultaten: beschikbaarheid, compleetheid en kwaliteit gegevens nog onvoldoende

Voor het modelleren van elke emissiebron, zuivering en verspreiding van chemische stoffen en antibioticaresistentie hebben we mogelijke toepassingen, huidige limiteringen en kennishiaten benoemd en aanbevelingen gegeven voor de toekomst. Voor twee bronnen van verontreinigingen (rioolwaterzuiveringsinstallaties en grensoverschrijdende rivieren) is een daadwerkelijke waterkwaliteitsmodellering uitgevoerd met de beschikbare data. Voor deze bronnen kunnen emissies en resulterende kaarten verder worden geanalyseerd in vervolgprojecten.

Op dit moment zijn de beschikbaarheid, compleetheid, en kwaliteit van de gegevens die nodig zijn bij het modelleren van een waterkwaliteitskaart via alle emissiebronnen nog niet voldoende om een complete en realistische waterkwaliteitskaart voor Nederland te maken. Om de waterkwaliteitskaart succesvol te kunnen inzetten, moet de standaardisering en vindbaarheid van databronnen worden verbeterd. Alleen zo kan relevante data beter en sneller worden gebruikt voor inzicht in de waterkwaliteit. Daarnaast kan de kwaliteit en reproduceerbaarheid van de data-analyse worden verbeterd door kennis van emissiebronnen gecentraliseerd te beheren met behulp van bijvoorbeeld emissiespecialisten.

Implementatie: kwaliteitskaart verder uitwerken

Met dit rapport is een basis gelegd voor een verdere uitwerking van de waterkwaliteitskaart en worden aanbevelingen gedaan voor hoe deze in de toekomst kan worden gerealiseerd. Een voorzet voor mogelijke vraagstellingen en te toetsen scenario's wordt per emissiebron gegeven. Daarnaast wordt apart ingegaan op de mogelijke modellering van verspreiding van antibioticaresistentie.

Rapport

Dit Verkennend onderzoek is beschreven in het Engelstalige rapport *Feasability and potential of a water quality map of the Netherlands* (BTO 2019.054)

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Feasability and potential of a water quality map of the Netherlands

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1 Introduction

1.1 Scope

Emissions of organic, microbial and chemical contaminants from different origin contribute to surface water pollution. Depending on the location or dynamics of the emission, the pollution spreads through the Dutch waterways (van Wezel et al, 2018a,b; Coppens et al., 2015). Emissions can stem from point sources or diffuse sources. Measuring pollution at different sites gives an indication of current pollution. However this is not conclusive for the origin of this pollution. Knowledge on sources and fate of contamination built into a model can optimize monitoring activities. Additionally, such water quality map containing modeled emissions of different origin and the consequent spread to other surface waters can help water managers in targeting problems by tracing them to their source. Furthermore, a water quality map can serve as a basis for evaluating different scenarios on their effectiveness to either reduce concentration of harmful substances, or reduce the spatial spread of these substances. These could be scenarios for climatological variations, increased population (Sjerps et al., 2016) (for instance leading to larger pollution from wastewater treatment plants), increased consumption or alternative applications of substances or abatement options (such as improved treatment) (Vries et al., 2013).

1.2 Objectives

This report has two main objectives.

- To evaluate the feasibility of such a water quality map by identifying sources of information, and evaluating these on correctness, completeness, and usability.
- To identify useful applications of a water quality map.

1.3 Outline

This report describes the results from the exploratory research project (Verkennend Onderzoek) 'water quality map'. A description of the methods and model on which results are based, plus the main outcomes of the work are described in Chapter 2. Chapter 3 provides a systematic description and evaluation of the data collected, sources and calculations for the following topics:

- Pollution from Cross-border Rivers
- Emission by sewage waste water treatment plants (SWWTPs)
- Emission by industrial waste water treatment plants (IWWTPs)
- Diffuse emissions from agriculture
- Antimicrobial resistance spread
- Purification treatment efficiencies for drinking water
- Purification treatment efficiencies for wastewater

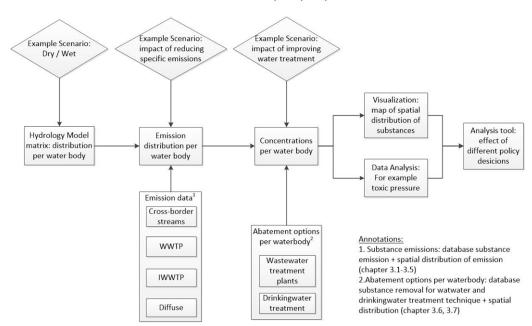
For each of these topics we address the possible applications, current limitations and knowledge gaps of the data available and make recommendations for the future. In Chapter 4, final recommendations are given. The hydrological model matrix used in this project was produced by Deltares based on the NHI LHM (KRW Verkenner). Deltares was only involved in the hydrological aspects of this project.

2 Water quality map

2.1 Methods

The conceptual model behind the water quality map is based on a straightforward linear combination of a hydrological model matrix of the Netherlands, constructed with the WFD-explorer model, data of contaminant loads for known emission sources, and data about treatment efficiencies (see Figure 2.1). The data on emissions was gathered from mostly public sources. The data was combined with the hydrological model matrix by a single script in the programming language 'R'.

This approach allows for combining existing knowledge and data from a large number of sources to create an overview of their combined spatial distribution and impact on water quality. Different scenarios can be implemented by changing the model input; dry or wet years (with respectively resulting low and high discharges) can be simulated and represented in the hydrological model matrix, the impact of reduced emissions and/or improved treatment by changing the respective input files. The combined effect of these scenarios can then be either visualized in a spatial distribution map of substances, or used to calculate e.g. toxic pressure in specific water bodies.



Schematic overview of water quality map

Figure 2.1. conceptual model of the water quality map and information sources

2.2 The model framework

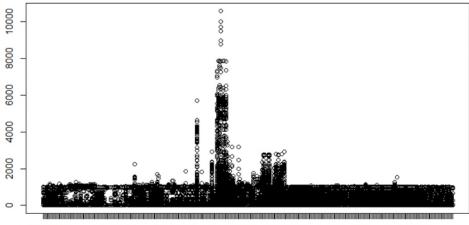
For the hydrological model matrix is a mass transfer matrix that represents the spread of pollution from different sources over the Dutch waterways, we use output from the WFD-explorer model (see reference section). The model itself uses a realistic schematization of the Dutch catchment areas (the NHI LHM Landelijk Hydrologisch Model), which is the basis for a calculated water balance. This is used to calculate how any substance spreads over Dutch surface water, under the assumption that the emission is constant and reaches steady state situation. For any source (diffuse or point), a theoretical substance released at 1000 g/s. The WFD-explorer model calculates the concentration at the receiving surface waters. Distribution of substances is calculated either with or without taking into account the decay of the substance with a standardized decay rate of 0.005 d⁻¹. This end state is converted to two separate matrices both containing all modeled sources (363 SWWTPs, 209 IWWTPS, 8508 Diffuse sources, 65 Cross-border Rivers) in columns and all receiving surface waters (27435) in rows, and per cell the modeled concentrations (g/s).

To recalculate the end state for actual emissions at the sources, the standard flux of 1000 g/s from the matrix can be replaced with actual emission flux (g/s) as obtained from a data source. The standard decay rate can be replaced with the actual decay rate for a compound of interest using measured or predicted decay rates from literature or back calculating decay rates from available monitoring data and known residence times in surface waters (Sjerps et al. 2016). Travel times from source to surface water can be calculated via the matrices for the situation with and without decay (Coppens et al., 2015, Equation 1). Here T is travel time, $C_{nodecay}$ is the flux without decay, and C_{decay} is the flux with decay. The values for C can be found in the respective spreadsheets.

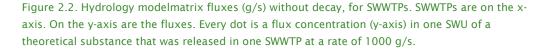
$$T = -\frac{\ln(\frac{c_{nodecay}}{c_{decay}})}{0.005}$$

For interpretation, it is good to realize that in the WFD-explorer some surface water concentrations can accumulate above their initial flux (see Figure 2.2). This is because of the underlying structure of the hydrology model, which can contain circular streams. These circular streams can cause adding of fluxes for some locations. E.g. a stream with a flux in g/s feeds a side stream, but is in turn fed with this side stream causing an additional flux in g/s compound.

(Equation 1)



rwzi_10001.c rwzi_12005.c rwzi_16010.c rwzi_2012.c rwzi_27004.c rwzi_4016.c rwzi_7022.c



Moreover, stochastic elements in the calculation can cause the calculations with decay to incidentally surpass the calculations without decay. Left unattended, this can cause negative travel times and consequently cause substances to increase rather than

5

concentrations altogether, and set any negative travel times to zero.

2.3 State of affairs

Previously, the publication of Coppens et al., (2015) and van Wezel et al., 2018a, 2018b gave examples of the value of the WDF-explorer model matrices. In van Wezel et al. (2018a, 2018b) it was identified on the basis of six modeled compounds that only 15 out of 182 IWWTPs have a large influence on drinking water production sites, of which one had a disproportionally big influence. In Coppens et al. (2015) it was found that for 345 SWWTPs, 65 influenced drinking water sources, and 134 influenced waters with a Natura 2000 function. This finding was based on two modeled substances (Coppens et al., 2015). Both publications stress the possibility of using results to implement water treatment technologies in the relevant locations, as a cost effective measure to improve water quality. This was done for the catchment of the river Dommel by ter Laak et al. (ter Laak et al. 2016).

For the current report, the aim was to evaluate the feasibility to include emission data of more compounds, and for more emission sources, and to combine these into a water quality map. Below we give a short summary of the results and recommendations per emission source.

2.3.1 Summary of modelling emissions

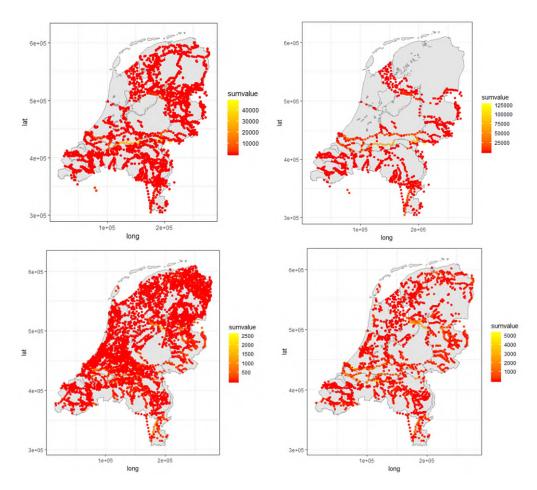
As a proof of concept, we modeled emission sources for cross-border Rivers and SWWTPs. We modeled these with the hydrological conditions for a wet season with high discharges (the first quarter of 2007) and a dry season with low discharges (the second quarter of 2011). For the rest of the topics, we investigated the potential to provide a useful map of pollution in surface waters by emissions from these sources.

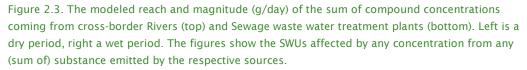
For **Cross-border Rivers** we modeled the impact of compounds for which measurement data *at the source itself* was available (see 3.1). The data consisted of concentrations for 328 compounds for cross-border Rivers. We modeled the emissions and for each receiving surface water calculated the sum of concentrations of compounds that were present, taking degradation rates of compounds into account. This approach was feasible. A point of attention is that the measurements on compounds were available for two rivers only, and had to be extrapolated to model the emissions from other rivers.

For **SWWTPs** we modeled the impact of compounds for which measurement data *at the source itself* was available (see 3.2). The data consisted of concentrations for a total of 914 compounds for SWWTPs. We modeled the emissions and for each receiving surface water calculated the sum of concentrations of compounds that were present, taking degradation rates of compounds into account. This approach was feasible. A point of attention is that not all compounds are measured at all SWWTPs. Emission had to be extrapolated from data on individual SWWTPs to all SWWTPs. Not all SWWTPs emissions are necessarily comparable, for instance because of specific industries or medical institutions that are present in the catchment of some SWWTPs (Vergouwen et al. 2011). Additionally, differences in the use of chemicals, personal care products and pharmaceuticals between populations of SWWTP catchments can result in deviations from the presumed similarity of emissions (van Batenburg-Eddes et al. 2002), and even if raw wastewater is of the same composition, differences in treatment efficiency can result in qualitatively and quantitatively differing emissions (Pieters 2011). This needs

to be evaluated on a compound by compound, and treatment by treatment, to refine future calculations.

The preliminary results for cross-border Rivers and SWWTPs (Figure 2.3) show that SWWTPs (lower plots) have a wider spatial reach than cross-border Rivers (top plots). More surface waters are affected by emissions from SWWTPs than by cross-border Rivers . This has to do with the connectedness of the affected surface waters, and the amount of individual sources considered in SWWTPs and cross-border Rivers.





In addition what can be seen in general is that, under dry conditions with low discharge, more surface waters contain some concentration of different compounds. This applies to both emission sources, cross-border Rivers and SWWTPs. The rationale behind this phenomenon is that under dry conditions, locks are opened to keep discharge leveled at the various locations (pers. comm, J. van Gils), or prevent salt water intrusion. In addition there is a difference in draining/feeding of polders under dry conditions. This enables the emission to travel where previously it could not. The total load of pollution does tend to be lower at low discharge. This is because water travels slower at low discharge (pers. comm, J. van Gils) and substances decline by natural biodegradation. Having reliable estimations of biodegradation thus proves to be important. The pollution load from emissions in cross-border Rivers is generally higher than those from SWWTPs. This is counterintuitive because less parameters were modeled in Cross-border Rivers than for SWWTPs (328 vs. 914). However, some parameters in the emissions from cross-border Rivers, mostly inorganic parameters, were present at very high concentrations. The sum of concentrations can be best expressed as a quality indicator such as total deviation from regulatory threshold values, total risk or toxic pressure, depending on the intended use of the indicator in future calculations.

For **IWWTPs** data was obtained on emissions per industry type (European and Dutch) and for specific industrial installations in the Netherlands. Also a search was performed for Dutch public permits for industrial emissions (See **3.3**). The data proved scarce. A lot of data concerned sum-parameters for multiple pollutants. These are less suitable for the model as no specific concentrations and degradation rate can be appointed to an undefined mixture. As a consequence the emissions from IWWTP were not modeled in this study. The recommendation is that, for now, this emission can only be modeled for some selected compounds for which explicit data are available.

For **diffuse emissions from agriculture** (see **3.4**) data was collected on type of culture, compounds used per culture, and the culture area was overlaid with the basins areas for diffuse emission, therewith obtaining the culture per basin area. Although this emission source would be a novel and important additional modeled source of emissions, it was decided that additional modelling is needed in order to estimate the emission fluxes to the water bodies. Namely, only a percentage of compounds used in a culture will end up in surface water via surface runoff and subsurface transport. For surface runoff, land management practices have to be taken into account to estimate the emission flux. Subsurface transport will strongly depend on the local soil properties and the chemical behavior of each substance. It is possible to model this (for example using the model GeoPEARL, <u>https://www.pesticidemodels.eu/pearl/pearl-model</u>; Lahr and van den Berg, 2009) but this would have to be done on a case by case basis both for particular substances, local application rates and soil conditions. It is not yet possible to model subsurface transport for the entire Netherlands, but this could be done for example for drinking water protection areas.

With regards to modelling spread and presence of **genes and antibiotics resistance** (see 3.5), data on the released concentrations from SWWTP and the removal rates for different SWWTP processes was collected via a search in literature. Limited concentration data were available for the Netherlands and no location specific data for the removal rates of resistant genes and bacteria in Dutch SWWTP were available. While removal rates from SWWTP processes from other countries were collected, because the treatments may differ substantially from Dutch processes, the data were determined to be incompatible. Measurement of the retention of genes and antibiotic resistant specific for the Dutch situation are needed in order to make a realistic prediction.

The **drinking water treatment capacity** (Bertelkamp et al, 2019; Hofs, 2014) was evaluated by calculating removal efficiencies for treatment clusters, based on data as supplied by Evides and Vitens. These ranged from 16 % to 83% per treatment cluster (see **3.6**). In addition, reliability estimates were calculated. This data is useful to estimate the removal efficiencies of compounds that are present at the intake points, to assess their potential influence on local drinking water quality. The data was limited by the number of compounds present in the database. Compounds not present in the dataset need assessment of their removal efficiency in another way (Vries et al., 2013).

For the **sewage waste water treatment capacity (see 3.7**), data was obtained using data on influent and effluent per SWWTP. It was found that on average 75.5 % of the individual concentrations of compounds was removed in SWWTPs. Data was available for quite a lot of compounds, however not for all SWWTPs. There is data available on the type of treatment per SWWTP. With this, the data would for instance be suitable for determining the 'best' sewage wastewater treatment installations to prevent high concentrations of compounds emitting to surface waters, and where to place these for the highest impact on surface water quality. The fact that the quality of influent and the retention times of water and sludge play a role as well in treatment capacity, has to be kept in mind, though.

3 Emissions from different sources

This chapter provides an overview of available and required data/information to define and evaluate sources of various types of pollution. It provides an overview of resources for future work on this topic, a list of the data sources, methods, calculations and assumptions made, and an overview of further resources/information needed for a complete story. Locations for each emission source were provided by either KWR or Deltares. Locations were input to the WFD-explorer model to calculate hydrology matrices of these emission sources at these locations, in relation to receiving surface waters.

3.1 Cross-border Rivers

Contribution to pollution

There are several entry points where water from abroad reaches the Netherlands. This water has travelled though different countries, accumulating pollution from emissions by for instance Industry, waste water, and agriculture. The pollution coming from other countries is, for a part, out of reach for Dutch abatement options and abatement should be sought in international (European) cooperation.

Relevance and pathways

Data obtained from Deltares reveals that 65 entry points from abroad are present, 63 of which have an inflow (m3/s). Three of those are very large in the wet period (3144 for the Rhine, 637 for the Meuse, 227 m3/s for the Schelde. The rest is under 44 m3/s (Figure 3.1). In a dry period, the discharge drops heavily for the Rhine, and the Meuse, and a little for the Schelde. Knowing the pollution coming from other countries is important because this will limit the efficiency of local Dutch abatement options.

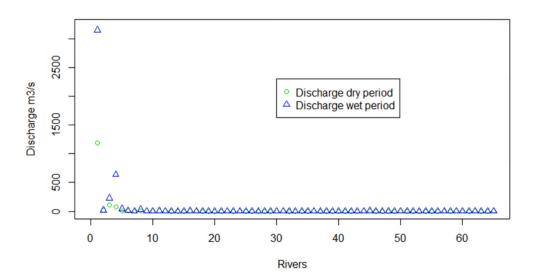
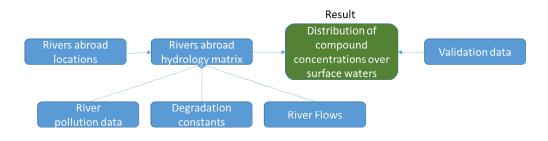


Figure 3.1 The inflow (in m3/s) (y-axis) of rivers that enter the Netherlands from abroad (x-axis).

For two of the large entry points, data on actual pollution is available. For Eijsden (Meuse river) and Lobith (Rhine river) an extensive monitoring program is in place, measuring over 500 parameters monthly (stored in the 'RIWA base', access on request at RIWA). Considering the contribution in volume of these two rivers, probably a large fraction of the contamination entering the Netherlands is covered.

3.1.1 Methods

For calculating emission exposure on the Dutch surface waters via water entering from abroad, it was identified that the following data is needed (figure 3.2):



Figuur 3.2 schematic overview of the relation between different required data sources.

Table 3.1 description of the data sources that are needed in the emission source of cross-border Rivers.

Description	Dataset Code (see Attachment)	Status
Locations of inflowing rivers	BLRLOC01	Proprietary data, Deltares
River flows: Discharge of the rivers in a wet and a dry season	BLRFLOW01	Proprietary data, Deltares
River pollution data: data on reported compounds in the inflowing rivers	VALIDAT01	RIWA data, permission needs to be obtained on a case by case basis
Degradation rate constants of compounds	CDC01	Public data, model predictions
Affected surface waters per abroad stream (hydrology matrix)	HMBLR01 HMBLR02	Proprietary data
Validation data: data on reported compounds in surface waters	VALIDAT01	RIWA data, permission needs to be obtained on a case by case basis

From the monitoring data from RIWA, a selection was made for data of 2016 (most recent). The measurement values of parameters over the year per location were

averaged per parameter. The assumption was made that all parameters with a CASnumber are relevant, combined with units of ' μ g/l' or 'mg/l'. For the location Lobith (Rhine) this resulted in 328 parameters. For location Eijsden (Meuse) this resulted in 253 parameters.

In the paper of Coppens et al. (2015) the data (concerning two compounds) were treated as such: averaged Rhine and Meuse concentrations were allotted to nine specific smaller inflows. We did the same. So for the remaining streams, the data per parameter was averaged over Eijsden and Rhine. This might deviate from the actual situation. Small rivers and streams can contain high concentrations of contaminants (ter Laak et al., 2014) because they are in some cases mainly fed by wastewater treatment plants under dry conditions. By a lack of data, this was not further investigated. This resulted in a total of 328 measured parameters with concentration (μ g/l) data for all inflowing rivers (figure 3.3). These were matched to the fluxes (g/s) in the hydrology matrices HMBLR01/02 (Table 3.1).

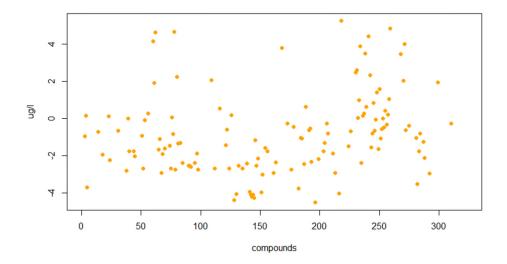


Figure 3.3 Log10 Concentrations μ g/l averaged between Lobith and Eijsden for 328 measured parameters. Some parameters (mainly metals and nutrients) have high concentrations.

Degradation rate constants used for compounds were those as calculated by the OPERA (OPEn (quantitative) structure-activity Relationship Application) model. It provides a suite of QSAR models to predict physicochemical properties and environmental fate of organic chemicals (Mansouri et al., 2018) (CDC01 in Table 3.1). For 311 of the parameters we could match degradation rate constants (CDC01 in Table 3.1). The other parameters were assigned the average of all matched degradation rate constants. This was 0.11 and this is corresponding to a half-life of 6.3 days, so half of the concentration is degraded in 6.3 days. We would expect that remaining substances that are in the river a long time are those with slow degradation. The standard degradation rate constant as handled by Deltares is 0.005 per day. It could be that a different data source with degradation rate constants gives a different picture. The fact that degradation rates can differ depending on temperature (e.g. slow degradation in winter, fast in summer) (Howard, 1991), was not included.

The modeling resulted in a table with steady state distributions over the surface water units for the 328 substances emitted from Cross-border Rivers. Two situations were modeled, a dry situation (low discharge) and a wet situation (high discharge). The results are shown in Chapter 2, Figure 2.3 (top plots).

3.1.2 Results

Which research questions can we answer, based on the available information?

- What types of pollution in Dutch surface waters stem from streams coming in from abroad? If in the Netherlands we want to improve water quality, the cross-border Rivers need a different (political) approach (Munthe et al., 2017). Measures to alleviate chemical pressure within the Netherlands will be less efficient if these come partly from abroad.
- What is the effect of low discharge on pollution in the Netherlands stemming from cross-border Rivers (see Figure 2.3, top plots)?
- What part of the composition of mixtures of compounds in Dutch surface waters stems from abroad?
- What are the surface waters that are affected by cross-border Rivers ?

3.1.3 Recommendations

What information is needed to get a complete picture?

- A datasheet with discharges of the abroad rivers (wet and dry season) was provided with the hydrology model matrices of Deltares. These data are in house data on discharges in waterways determined by monitoring devices and theoretical mass balances of water discharges.
- Information on actual measured parameters for two locations for cross-border Rivers are present. For the other locations, there is no data. We extrapolated the pollution from the two locations to the others, however it might be that these are influenced by other sources of pollution than the two locations that were used. Although the flow (m3/s) is smaller in these locations, and these are a less important source, they influence particular surface waters. It can be evaluated based on their influence, how accurate the extrapolations need to be and if a study should be performed towards the accuracy of these extrapolations. Also an effort could be directed to obtaining measurement data from abroad measuring points for these rivers.
- Aside from using measured values as input, estimated values based on emission of sources along the cross-border Rivers (industries, sewage) could be used. This approach is being adopted in the 'Kennisimpuls Waterkwaliteit' (KIWK) project 'mengseltoxiciteit' within a collaborative framework with Deltares, WUR and RIVM.

3.2 Sewage waste water

3.2.1 How does this contribute to pollution?

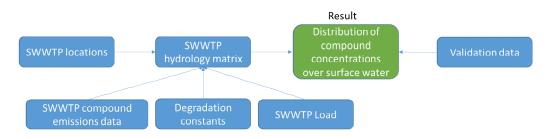
Sewage waste water contains a plethora of chemical and biological contamination stemming from its producers. On average, it consists of 35% communal wastewater: greywater (from sinks, tubs, showers, dishwashers, and clothes washers), blackwater (from toilets, combined with the human waste that it flushes away), waste waters from small industries (15%), rain (30%) and infiltrating ground or surface water (20%) (Lieftink en de Man, 2017). Exactly what industries or companies add their waste water to the sewage waste water , where and when they do this, is largely unknown.

Sewer waste water treatment plants are the points of entry where this waste water enters the surface water, after some level of purification treatment.

The compounds entering the waters via SWWTPs could cause harm to the aquatic ecosystem in receiving surface waters (Coppens et al., 2015) by toxic effects to organisms in the ecosystem and oxygen depletion. Moreover, the compounds may compromise or limit reuse of these waters in agriculture or in the production of drinking water. Another risk is the exposure to pathogenic bacteria during recreational activities. This is becoming more problematic due to increased popularity of for instance city swim fundraising events.

3.2.2 Methods

For obtaining a view of emission exposure from SWWTPs on the Dutch surface waters the following data is needed (figure 3.4, table 3.2):



Figuur 3.4 schematic overview of the relation between different required data sources.

Table 3.2 Description of the data sources that are needed in the emission source SWWTPs.

Description	Dataset code (see Attachment)	Status
Locations of SWWTPs	SWWLOC01	Public data
	SWWLOC02	Public data
Recent load of SWWTPs	SWWLOAD01	Proprietary data
Compound emissions data: data on compounds in the effluent of SWWTPs	SWWCOMP01	Public data
Compound emissions data: which Industry types emit what compounds to which SWWTPs		Data not identified
Degradation rate constants of compounds	CDC01	Public data
Affected surface waters per SWWTP (hydrology model)	HMSWW01 HMSWW02	Proprietary data
Validation data (how accurate are the calculations)	VALIDAT01	Proprietary data, permission required on a case by case basis.

The locations of 363 SWWTPs were publicly available and were stored as SWWLOC01 (see Attachment I). These locations were set as a point source in the WFD-explorer model, and the model was run by Deltares to get the hydrology matrices (HMSWW01 and 02 in Table 3.2).

Measured values of effluent concentrations (μ g/l) where publicly available for 214 SWWTPs for in total 913 substances, identified by CAS-number. Not all substances are measured for all SWWTPs (Figure 3.5). These data were stored as SWWCOMP01.

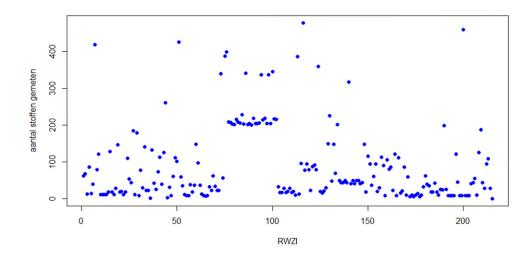
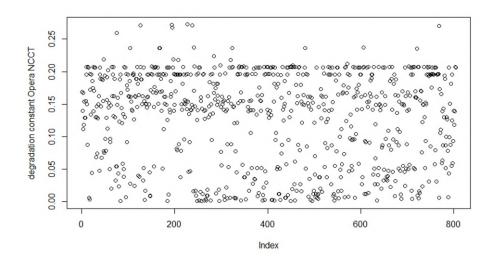


Figure 3.5 The number of parameters measured (y-axis) per SWWTP (x-axis) in the Watson database.

The SWWTPs in the hydrology matrices HMSWW01/02 and the measurement data SWWTPs from SWWCOMP01 were connected by their "RWZI code". To gain a more complete picture of the pressure of SWWTPs on surface waters, we used average measured values for compounds of SWWTPs and assigned these to all SWWTPs where no value was measured for that substance. This approach was possible as SWWTPs for a large part receive their water from the same source (household water) and are therefore expectedly rather similar. This was not tested, but should be in a future analysis as this could be untrue for some compounds (ter Laak et. al., 2014). The SWWTPs without measurement data were assigned the average measured values. It would be advisable to adjust this extrapolation in future work, and have at least two or three sources if an average is awarded. This will prevent incidental emissions from a single SWWTP being extrapolated over all SWWTPs. Alternatively, ratios between chemicals can be used to predict concentrations of other chemicals that are not measured. If consumption patterns are similar, emissions can have similar ratios for individual chemicals.

To get from measured concentrations to actual grams per second of compounds entering the surface waters, we used SWWLOAD01 for the quantity of water coming from SWWTP's. This data is not publicly available, and was originally obtained from CBS microdata by a third party. The loads from SWWTPs from SWWLOAD01 and the point sources SWWTPs in the hydrology matrices were again connected via their "RWZI code". This code was partly incompatible because it was not fully standardized and this had to be adjusted.

For degradation rate constants we use values from Mansouri et al. (2018) (CDC01 in Table 3.1) and link these to the substances via their CAS numbers. See Figure 7 for how the degradation rate constants are for the compounds measured at the SWWTPs. For 50 compounds no degradation constant could be linked. The average of all the other compounds in the Watson database (~0.13) was used for these. This is slightly higher than the average found in cross-border River compounds (0.11). This makes sense because for compounds in a SWWTP the actual residence time shall expectedly be shorter than in a River system, and fast degrading compounds will potentially still be present.





This resulted in a table with steady state distributions over the surface water units for the 914 substances emitted from the SWWTPs. Two situations were modeled, a dry situation (low discharge) and a wet situation (high discharge). The results are shown in Chapter 2, Figure 2.3 (bottom plots).

3.2.3 Results

Based on the results we have now, which research questions can we answer?

- How does SWWTP effluent influence water quality over the Netherlands? For this
 question the exceedance from regulatory thresholds for compounds for each
 surface water unit, such as the Water framework Directive (2000) or Dutch drinking
 water directive (2018), or suggested by non-governmental organizations, can be
 calculated. This provides water utilities the information where problems arise that
 stem from SWWTPs.
- The distributions of the 914 compounds over surface water units could be restructured to compound groups such as 'pharmaceuticals', 'detergents', etc. to get an idea of the type of pollutants.

- Rerouting wastewater to less harmful areas. Emissions into large waterbodies such as rivers ensures the average concentration is low (not considering other emissions, from other sources). However, the reach of this pollution is large as the river flows fast. The WDF-explorer model matrices can identify locations suitable for emissions to smaller, less interlinked waterbodies. These can serve as a pretreatment, where biodegradation can solve part of the problem, and has local influence on water quality (personal communication, Jos van Gils and Erwin Roex).
- Adverse effects of low discharge on water quality via SWWTP effluent. The hydrological mass transfer matrix was calculated for both a dry and a wet period. These can be compared. It can be established where most problems will occur. For example, in Figure 2.3 the SWU's that receive any substance at any concentration from SWWTPs in dry periods (left) with low discharge and wet (right) periods with high discharge are visualised.
- Potential and importance of SWWTP purification treatment for Dutch surface water quality. Dataset SWW01 holds measurement data of compounds of SWWTP effluent and influent. A spatial map could be made of the compounds influencing surface waters *without* any SWWTPs (based on influent values) and this can be compared with the actual compounds influencing surface waters with SWWTP purification (based on effluent values). This gives an indication on the importance of SWWTP's purification treatments. This could be extended by simulating the effects of equipping SWWTPs with an additional treatment step. Would this be enough to have zero exceedance of the thresholds by compounds emitted by SWWTPs? Or will other measures that are targeted to minimize SWWTP influent compound concentrations prove indispensable.
- Forensics to locate sources of pollution. If a problem with pollution is detected in surface waters, the model could be used to locate the potential source of contamination, for example a particular SWWTP.
- Optimal reuse of surface waters omitting hot-spots in pollution, in times of high and low discharge. The spatial spread of mixes of chemicals can give direction to favorable extraction points and guidance for periodic intake of reservoirs (Kroesbergen et al. 2018), in terms of water quality.

3.2.4 Recommendations

What is needed to get a complete picture?

The data on SWWTPs are quite good and usable. There are some points of attention:

- The data on compound concentrations in influent and effluent of SWWTPs are incomplete. Not all SWWTPs are included, and not all SWWTPs measure the same compounds. However, it is an option that concentrations can be extrapolated. The data are averages, and do not reflect temporal changes throughout the year.
- The load of the SWWTPs is not publicly available, which would pose a problem if the most recent loads would have to be acquired. This is necessary as some SWWTPs close down, and would at that point not contribute to pollution. There is an open

• The decay rates of compounds were from a source which based the values on predictions (Mansouri et al., 2018) rather than measurements, and could therefore be inaccurate for some compounds. Moreover, degradation rates are not constant but vary with conditions. Especially degradation rates of compounds with high degradation rate constants need to be accurately established, as they are most prone to variation as a result of environmental conditions. E.g. a variation in half-life of 1 hour to 1 day would make a very large difference in the concentrations an impact of compounds to surface waters. A half-life of 1000 days vs 10000 days would not, as residence times generally are not so large, in the range of days (rivers) to months (lake IJssel, polders, canals). For 50 of 914 compounds, no decay rate was present in dataset CDC01. These would have to be added from another or additional source.

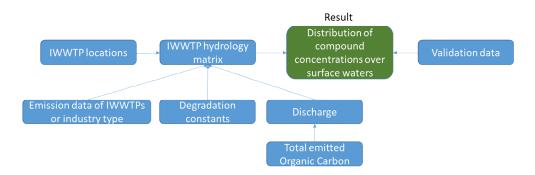
3.3 Industrial waste water

3.3.1 How does this contribute to pollution?

Industrial waste water treatment plants (IWWTPs) are an entry point for a wide variety of substances, depending on industry type. As industries are constantly innovating their products and methods, the chance of emission of novel substances via IWWTPs is substantial. The total capacity of IWWTPs is comparable to that of SWWTPs. Nevertheless, IWWTPs emissions have received much less attention. Possibly this is because of a lack of data. It is hard to get a grip on these emissions because there is a lack of transparency on the substances emitted . The emissions from IWWTPs are less continuous from that of SWWTPs. Emissions depend on the industry type and will differ per location. Industries may also vary their production process and with that their use of specific substances in time. From time to time, incidents with emissions of compounds by IWWTPs occur. For example, the high concentration of pyrazole in surface waters that was observed in 2015 (Baken et al., 2016).

3.3.2 Methods

The data in the following scheme was necessary for the modeling of emissions from IWWTPs on Dutch surface waters (figure 3.7).



Figuur 3.7 schematic overview of the relation between different required data sources.

We identified several sources with possible useful information. These are listed in Table 3.3. In the data sources, for every facility, an industry type is given. This can be a NACE,

SBI, TNO or unknown origin code, depending on the data source. These have to be linked to the 209 IWWTPs modeled in the hydrology matrices. The emission registration databases IWWCOMP01-04 (Table 3.3) hold emission data from European industry facilities, industry types, or specific Dutch facilities via water. These can be extrapolated over Dutch IWWTPs.

Many of the parameters listed in the data sources IWWCOMP01-04 (Table 3.3) are sumparameters. These are of limited use for the water quality map as the degradation rate constants of individual compounds within these grouped parameters can differ. In the report of van Wezel et al. (2018) the emissions from the E-PRTR were per industrial sector normalized based on emissions of total organic carbon. In that report, the data on total organic carbon of the modeled IWWTPs was used to come to a scaling of the emission of the IWWTPs (g/s). This method can be applied.

We also checked if data from permits could in potential be a source of information on emissions of individual industries. To find publicly available permits directly, we searched Google with the search phrase: "watervergunning lozing filetype:pdf". We opened and assessed a small subset of the different files containing permits.

Description	Dataset Code (see Attachment I)	Status
Locations of IWWTP	IWWLOC01 IWWLOC02	Public data
IWWTP loads	IWWLOAD01	Public data
Emissions per IWWTP industry type	or IWWCOMP01 IWWCOMP02 IWWCOMP03 IWWCOMP04	Public data Public data Public data Public data
Degradation rate cons of compounds	stants CDC01	Public data
Affected surface wate per IWWTP (hydrology model)		Proprietary data Proprietary data
Validation data (how accurate are the calculations)	VALIDAT01	Proprietary data, permission required on a case by case basis.

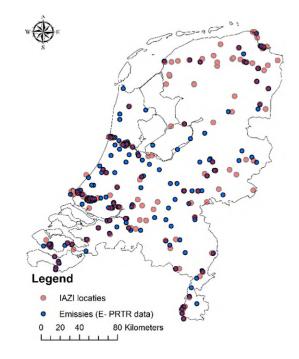
Table 3.3 description of the data sources that are needed in the emission source IWWTPs

3.3.3 Results

The paper of van Wezel et al (2018) provides already an application of the model. In their paper, the spread of modeled substances over Dutch surface waters was very different, depending on which IWWTP emitted the substance, which was in turn dependent on the industry type that used the substance. There was not a very good fit with monitoring data, which could be a result of the one-sided modeling of only IWWTPs

emissions (and not other sources), resulting in an underestimation of concentrations. It could also be that the European data on emissions per industry did not match the situation in Dutch IWWTP substances very well.

The locations between two of the available datasets were compared (figure 3.8). Dataset IWWLOC01 holds the locations of IWWTPs from the ER database, and these were used in the WFD-explorer model to construct the hydrology matrices. Dataset IWWLOC02 holds the locations of IWWTPs as provided by the European E-PRTR. Although many locations coincide, many also do not. This leaves the question what the correct locations of IWWTPs are. This should be clarified in any future analysis.





In our preliminary investigation of information available in Industry permits for emissions, permits did not seem to contain much information on emitted substances. More generic parameters were mentioned such as maximum water allowed, total oxygen consumption, nitrogen, phosphorus, and in one case some metals.

3.3.4 Recommendations

The usefulness and correctness of the different possible data sources for a construction of the emissions from IWWTPs will have to be further evaluated. From that evaluation will follow how many substances can be modeled in the water quality map, based on available data and quality.

3.4 Diffuse sources from agriculture

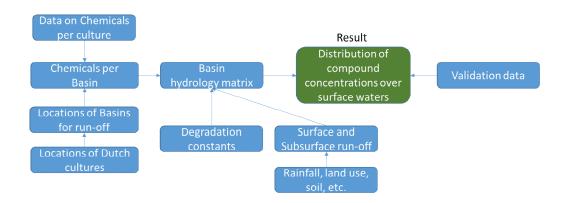
3.4.1 How do these contribute to pollution?

In agriculture, pesticides are used to protect crops against pests and diseases. Depending on the method of application, such as spraying, sprinkling, pouring or dipping, the active substances of pesticides may end up into surface water via drift, atmospheric deposition, run-off or drainage, and into groundwater via leaching or infiltrating surface water. Depending on the properties of the active substance and the prevailing (soil) water environment, active substances can become completely or partially degraded or taken up by plants. Degradation, however, may result in more or less stable transformation products (degradation or reaction products), that can be transported throughout water systems. Both the active substances and the transformation products can individually and as a group (via mixture effects) be toxic.

The fractions or concentrations reaching the ground- or surface water depend on the dosage, land management and application method, crop uptake, soil characteristics, soil moisture, slope, rainfall, adsorption coefficient of the chemical, and other conditions.

3.4.2 Methods

For calculating emission exposure on the Dutch surface waters via runoff entering from diffuse sources, the following data is needed (figure 3.9, table 3.4):



Figuur 3.9 schematic overview of the relation between different required data sources.

Table 3.4 Description of the data sources that are needed in the emission source diffuse emission by agriculture

Description	Dataset Code (see Attachment)	Status
Locations Dutch agricultural culture areas for 2016	CULTLOC01	Public data
Locations Basins 8508 in the WFD-explorer model (Deltares 2018)	BASLOC01	Proprietary data, Deltares
Data on chemicals used (kg/hectare) in different cultures	CULTCOMP01	Public data
	CDC01	

Degradation rate constants of compounds		Public data, model predictions
Basin (subsurface) run-off	Not ready	Envisioned model outcome
Affected surface waters per basin (hydrology model)	HMBAS01 HMBAS02	Proprietary data
Validation data (how accurate are the calculations)	VALIDAT01	Proprietary data, permission required on a case by case basis.

We used datasets in Geodatabase with the following features (shape files CULTLOC01): Data on cultures per agricultural parcel for 2016 contained 296 different cultures in 786572 parcels in the Netherlands.

In ArcGIS the agricultural parcels are overlaid with the boundaries of the 8508 basins from the WFD-explorer and then summarized per culture. The output consist of a table listing the hectares per culture per basin. Each basin contains one (virtual) hydrological element to which diffuse pollution was attributed (figure 3.10).

The cultures were matched with the cultures from the CBS database (CULTCOMP01 in Table 3.4), which held 58 cultures with in total 252 associated chemicals.

Chemicals and cultures are not related to any identifiable standard, which made coupling to other datasets time-consuming. Chemicals were supplemented with CASnumbers partly automatic, partly by hand, and consequently matched with degradation rate constants. The cultures were matched by hand.

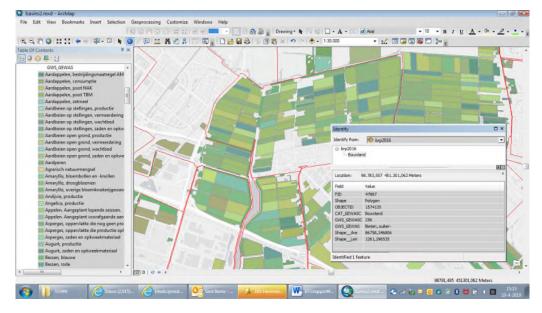


Figure 3.10 BRP cultures (colored) overlayed by WFD Basins (red).

Based on the data that was collected and processed, an investigation could be made towards which surface waters are in principle affected by diffuse sources, for instance at low and high discharge conditions. However, modelling the concentration of the chemical mixture in these surface waters affected by diffuse emission is not possible, as run-off by surface and subsurface needs more detailed modeling. The fraction of chemicals transported by runoff and infiltrated into the soil needs to be determined, which depends on application method and hydrological properties of the soil. Furthermore, the retention of chemicals in the subsurface by sorption and degradation can strongly differ depending on chemical properties and soil type. It is possible to model on a case-by-case basis using for example the GeoPearl model (<u>https://www.pesticidemodels.eu/pearl/pearl-model</u>) (Lahr and van den Berg, 2009) in future projects.

3.4.4 Recommendations

Using the present data, it is possible to make a theoretical, statistically based, assessment of the potential spread of compounds from diffuse emissions across the Netherlands, based on either the maximum amount emitted, or by assuming a fixed emission size.

For a realistic, quantitative estimate of the impact of pesticides on sources for drinking water production, a deterministic approach is required to simulate the relation between pesticide application throughout landscapes and mass transfer towards and through water systems. This can be done by coupling the available codes for simulating the fate of pesticides in different environmental compartments.

GeoPearl can be used to simulate the environmental fate of pesticides on a national level, including the mass transfer of pesticides and metabolites towards surface water and groundwater. The mass transfer towards surface water can be used as input for the hydrological mass transfer matrix to assess the impact at surface water intake points. GeoPearl, however, does not provide code for simulating the fate of pesticides and metabolites during transport towards groundwater wells or groundwater discharge areas (drainage by surface waters). This would require coupling the output of GeoPearl with a second model code for simulating the fate of pesticides in the subsurface. If one is only interested in the water quality at certain points (for instance at abstraction wells), a rather extensive streamline approach is both suitable and effective (Vink et.al., 2011, Stuyfzand, 2019). This approach also provides the opportunity for calibrating uncertain parameters for sorption and decay, and for estimating uncertainty band widths more effectivity than approaches based on a cell-by-cell algorithm. This would require additional information about hydrological connectivity and travel times in different subsurface redox environments in order to simulate decay and sorption using estimates of these chemical properties provided in literature (Stuyfzand, 2019). Regional groundwater models provide the most suitable method for gathering information on groundwater flow, but for national scale purposes the Dutch hydrological model can be used as a rough indication.

3.5 Spread of antimicrobial resistance

3.5.1 How do these contribute to pollution

Antimicrobial resistance (AMR) as a concept represents antibiotics and metabolites (AM), antibiotic resistant bacteria (ARB) and antibiotic resistance genes (ARG). AMR has been increasing due to the use, and misuse, of antimicrobials in human and veterinary medicine, and by discharge of waste in the environment during manufacturing (Danner et al, 2019). It is increasingly acknowledged as a serious threat to public health (World Health Organization 2014). The spread of AMR makes antimicrobials less effective and therefore treatment for patients is becoming increasingly problematic, more expensive and in some cases impossible. In the Netherlands AMR spreads in the environment through human sewage and animal manure (Schmitt et al, 2013). Wastewater treatment plants (WWTPs) are not designed to reduce AMR and as a results are an important source for spread of ARB and ARG to the environment. The release of some bacteria, ARB and ARG from SWWTPs was investigated in the Netherlands by the National Institute for Public Health and the Environment (RIVM) (Schmitt et al., 2017). However, the spread of ARB and ARG in the water cycle was not further investigated. Publications point out that the increase of AM, ARB and ARG in the environment have severe ecological and health impact (Ben et al, 2019; Danner et al, 2019). Knowing the fate and loads of AMR related components can be used, for example, to determine the effect of specific intervention measures on the downstream environment.

Relevant Pathways

The three major AMR contamination routes include 1) AMR related to human use, released by WWTPs effluent, 2) veterinary use (to treat livestock against bacterial infections) released as manure on the land, or 3) by waste discharge through AM manufacturers. For the Netherlands this third route is not relevant, as AM are not largely produced in the Netherlands. From the two remaining sources the SWWTPs are best characterized, while manure on land is a diffuse source, and it is difficult to quantify manure that is leaching to water bodies like surface or groundwater (Fatta-Kassinos et al. 2017, Hornstra 2017, Schmitt et al. 2013, ter Laak 2012). Therefore the information used in this study is primarily coming from studies with data from influent and effluent measurements of SWWTP.

The presence of antimicrobial residues in the environment causes selective pressure resulting in the development and increase of antibiotic resistant micro-organisms in the environment. Incorporation of genetic elements harboring ARGs into environmental strains aids to the increase of ARGs in the environment, and increases the risk of subsequent transfer of ARGs from environmental strains to human pathogens. (Schmitt et al., 2013). AMR is spread, through incomplete removal of antimicrobials themselves, the proliferation of AMR bacteria and the spread of mobile AMR genes. SWWTPs in the Netherlands discharge to surrounding surface waters, contributing to the increase of ARB and ARG to the aquatic environment. Human exposure can occur, for example, through recreational use of surface waters which receive SWWTP effluent, the use of surface water for irrigation purposes, or by the fact that surface water is a main source for the production of drinking water (currently under investigation in BTO project ABR implications for drinking water).

3.5.2 Methods

Information of four clinically relevant antibiotic types and their associated ARB and ARG were studied: tetracyclines, sulfonamides, quinolones and beta-lactams. A literature review for the concentration of select ARB and ARG was performed. In total, data from

27 studies were extracted and included in the dataset. Treatment specific removal rates were also extracted from the literature. However, the majority of the studies were from China and therefore the SWWTP processes may not be compatible with treatment processes in use in the Netherlands. From the limited literature review, no studies regarding treatment specific removal rates were found from the Netherlands.

The RIVM performed a survey of 100 of the 341 SWWTPs in the Netherlands and reported concentrations of antibiotic resistant bacteria in raw and treated wastewater (Schmitt et al. 2017). Location specific concentrations were not reported and therefore the concentrations extracted from the report are average concentrations. The concentration of extended spectrum beta-lactamase (ESBL) *Escherichia Coli* and carbapenem-resistant Enterobacteriaceae (CRE) were reported, in addition to the concentration of sulfonamide resistant gene *sul1* and enthromycin resistance gene *ermB* (H. Schmitt et al. 2017). Carbapenemase-producing Enterobacteriaceae (CPE) and vancomycin resistant enterococci were also detected.

How were the data treated for implementation in the model?

To avoid input errors, the data were extracted from articles and input to the dataset without converting the concentration units for the AM, ARB or ARG. For example, some publication report the concentration of ARB as bacteria per ml or log bacteria per 100 ml and ARG as relative abundance (ARG gene copies per copy of the 16S rRNA gene) while others report absolute gene copies separately from the 16S rRNA gene copy number. Therefore, prior to implementation in a model, the concentration data will need to be homogenized. The level of detail in the studies varied – some studies reported the concentration of AMR genes and/or bacteria before and after each treatment process, while others only reported concentrations before or after a single process, or simply the log reduction of a single process. When both the log reduction and the concentration before and after a process were given, the concentration data were given priority.

For ARG concentrations, only studies which either also reported the copies of 16S rRNA gene/sample volume or reported the specific gene as a proportion of 16S rRNA gene (eg. *Sul1* copies/16s rRNA) were recorded. This was to determine if treatment processes were removing specific ARGs preferentially compared with the overall reduction in microbial genes.

In addition to the concentration of ARB and ARG, where possible, the size of the SWWTP (flow and/or population), whether hospital or other care facilities contribute to the flow and to what proportion of the flow was included. Due to time constraints, the data set was not further refined and was not input to the model.

3.5.3 Results

From this data set (table 3.5 and 3.6) the following questions could be answered;

What concentrations of tetracyclines, sulfonamides, quinolones and beta-lactam resistant genes and bacteria can on average be expected in the influent and effluent of SWWTPs, in the Netherlands and internationally?

ARG	Influent (genes copies/16S gene copies)		Efflue (genes copies, copies	Ref. no.	
	Mean SD		Mean	SD	
Beta-Lactams					
bla SHV/TEM	1.3E+00		1.1E+00		18
blaCTX-M	3.7E-01	7.2E-02			6,16,25
blaCTX32	2.6E-04	1.4E-04	6.8E-04	1.1E-03	10
blaOX58	8.0E-03	1.0E-02	3.0E-02	5.2E-02	10
blaSHV34	9.7E-03	1.3E-02	3.0E-02	3.6E-02	10
blaTEM	8.1E-01	1.4E-01	6.3E-01	1.4E-01	6,12,16,22,24
Enthromycin resistance					
ermB	7.8E-01		7.7E-01		21
Quinolones					
qnr	7.3E-01 9.4E-02		7.6E-01	2.5E-01	15
qnrS	8.7E-01	1.5E-01	6.9E-01 1.5E-01		6,12,16,24
Sulfonamides					
sul	1.1E+00	2.3E-01	1.3E+00	6.1E-01	15
sul1	8.1E-01	2.7E-01	7.8E-01 2.9E-01		5,6,21,10,11,12,16,1 7,18,22,23,24,25,26
sul2	7.3E-01 2.1E-01		6.6E-01	2.2E-01	5,10,11,12,16,22,23, 25,26,27,28
Tetracyclines					
tet	8.8E-01	1.3E-01	1.1E+00	4.8E-01	15
tetA	6.1E-01	8.1E-02	5.4E-01	7.8E-02	6,11,22,25,28
tetC	1.6E-03	1.3E-03	1.0E-02	1.7E-02	10,27
tetG	7.3E-01	1.4E-01	5.7E-01	1.6E-01	5,13,23,26,28
tetM	5.9E-01	3.3E-01	4.6E-01	3.1E-01	5,10,11,23
tetO	7.3E-01	1.5E-01	4.9E-01	1.4E-01	5,12,16,17,23,25,26, 28
tetT	4.0E-01	5.7E-02	4.1E-01	1.4E-01	23
tetW	6.9E-01	1.0E-01	4.3E-01	1.7E-01	5,11,12,17,22,23,25, 26

Table 3.5 Concentration of antimicrobial resistant genes in SWWTP influent and effluent. Concentrations given as genes copies/16S gene copies.

Antimicrobial Resistant	Bacteria	Influent (log CFU/100 ml)		Effluent (log CFU/100 ml)		Ref no.
		Mean	Mean SD		SD	
Carbapenem	Enterobacteriaceae	2.31	0.44	0.31	0.63	21
ESBL	Enterobacteriaceae	4.04	1.53	1.78	0.96	7
ESBL	Escherichia Coli	5.03	0.58	2.45	0.35	3, 4
Sulfonamide	Escherichia Coli	5.20	0.95	3.02	0.70	8
	unspecified					17
Tetracycline	Escherichia Coli	5.05	0.75	3.04	0.39	8
	unspecified	8.85				17

Table 3.6 Concentration of antimicrobial resistant bacteria in SWWTP influent and Effluent. Concentration in log CFU/100 ml

3.5.4 Recommendations

Compared to the chemical data the amount of quantified information of AM, ARB and ARG is very limited specifically for the Netherlands. At this moment, only one study exists that determined AM, ARB and ARG, although not for SWWTP as point sources but as averaged values of many SWWTP. In general it is possible to use averaged AM, AB and ARG data, similar as for chemical data, In this case, the number of datasets (specifically AM, AB and ARG in one study) is very limited, and this provides too little ground to average the numbers. If averaging is appropriate depends in any case on the particular question that is intended to be solved with the model. Therefore, more detailed information about AM, ARB and ARG levels in SWWTP effluent is needed.

- To have a better understanding of the distribution and fate of ARB and ARG in the Netherlands, location specific data are crucial. For example, the raw data from the RIVM study (Schmitt et al. 2017) would be very valuable as input for future modelling, over average values reported.
- Analysis of the data set to determine the removal efficiencies for resistance genes
 of tetracyclines, sulfonamides, quinolones and beta-lactamresistant and bacteria of
 specific SWWTP processes. The data are available for this calculation, but due to
 limited time and budget the analyses could not be performed. Knowing which
 treatment processes are most effective would allow more specific intervention
 scenarios to be modelled. For example, implementing the most effective treatment
 processes at the SWWTPs which feed the most vulnerable surface waters in the
 Netherlands.
- The data set contains studies from twelve countries in Europe, North America, Asia and Africa. From this, future work could examine the concentration of ARB and ARG between countries and to the Netherlands.
- SWWTP performance and inflows can vary between the summer and winter (Zhang et al. 2010) and therefore location specific concentration measurements should be taken at SWWTP in more than one season.
- The concentrations of ARB and ARG from hospitals and other health care center waste streams should be separated from municipal waste streams in order to understand the relative contribution of each of them.

- Combined sewer overflows and misconnections of distribution pipelines are expected to be a significant source of AMR related pollution, and frequency and magnitude of these systems should be evaluated, and could be linked to climate predictions for extreme weather events in the future.
- The contribution of ARB and ARG to the environment should also be quantified, and to correctly model the fate and distribution of ARB and ARG from SWWTP effluents, gene and bacteria specific degradation rate must also be known and implemented in the model.
- A more complete understanding of SWWTP processes in the Netherlands is necessary, specifically what processes are in place at which treatment plants. This is particularly relevant for tertiary (disinfection) processes.
- More studies and measurements from SWWTP processes comparable with SWWTP processes in the Netherlands are required. The dataset collected was dominated by studies from Chinese SWWTP, where the treatment trains, environmental conditions, antibiotic use are not comparable to treatment processes implemented in the Netherlands.
- Finally, overland sources of ARB and ARG have not been included in the data set, but are important. For example, flow from manure on farm fields, from concentrated animal farms, any removal expected from percolation through soil, etc (ter Laak, 2012).

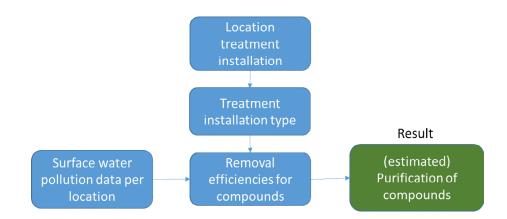
3.6 Abatement by water treatment plants

3.6.1 How do these contribute to water quality

Not only (regional) water authorities, but also drinking water companies are challenged with the question if, where and how to abate compounds of emerging concern in the water cycle (Fischer et al. 2017). Although human health risks seem negligible for current emission loads on surface water and infiltrated groundwater (Houtman et al. 2014), emission outbreaks due to e.g. failing IWWTPs may have severe impact on drinking water production. For example, during the summer of 2015, an emission by an IWWTP of amongst others pyrazole resulted in a long-term stop of surface water intake for drinking water production in the Netherlands (Baken et al., 2016). In 2018, industrial emission of 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoic acid (FRD-903, also known as 'GenX') has led to a debate in the Dutch court because of expected future problems for drinking water production. Also other examples of industrial emissions giving rise to water quality portray the relevance of industrial emissions on surface water quality and drinking water production (see van Wezel et al. (2018) and references therein). Therefore, next to having insight on surface and ground water quality, insight in whether drinking water production sites can meet current water quality criteria is highly relevant for decision makers at drinking water companies.

3.6.2 Methods

The following data and calculations were needed (figure 3.12, table 3.7):



Figuur 3.12 schematic overview of the relation between different required data sources.

Table 3.7 description of the data sources that are needed in the abatement by water treatment plants

Description	Dataset Code (see Attachment)	Status
Locations Drinking water treatment plants (DWTP)	DWTPLOC01	Public data
Treatment types DWTP	DWTT01	Proprietary data, Deltares
Removal efficiencies	DWRM01	Proprietary data, Vitens and Evides
Compounds in surface water	Output water quality map	Output not ready

Data sources

To obtain the removal efficiency of drinking water production sites, measurements of Contaminants of Emerging Concern (CEC) in drinking water and of the water at the inlet of the treatment plant are needed. Data sets from two drinking water companies were considered, i.e. data from measurement campaigns by Evides (D_Evides) and Vitens (D_Vitens). These also contained the treatment types.

To arrive at a concentration in tap water on the basis of compound concentration data in drinking water sources, we have set up a flow chart (figure 3.13).

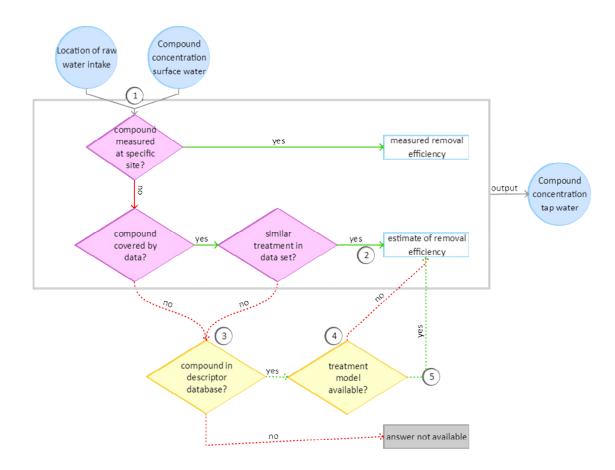


Figure 3.13. Decision flow chart to calculate concentration of compound in drinking water. In this project, we calculate the concentration of a compound in tap water by the procedures in the grey framed box, i.e. using measurement data. The circled numbers relate to different procedures outlined in the main text.

Decisions and flows of procedures that are developed within this study on the basis of available data source are framed grey. The current framework can be extended using a database containing chemical compounds information (see step 3 in Figure 3.13), advanced statistics to calculate similarity between compounds (step 4) and model calculations using both compound information and information about the specific treatment (step 5). If measurements are not available, model calculations could be used to derive removal efficiency estimates (Wols and Vries 2012; Vries, et al., 2013; Lee and von Gunten 2012; van der Hoek et al., 2014) for the case treatment models are available, or a rough estimate of removal efficiency based on similarity of a compound with another (measured) compound.

Given the scope of this project and prioritization of activities due to limited resources, calculations using treatment process models are not performed. Hence, only data sources which were readily available have been used.

In this work, removal efficiencies are calculated based on treatment data as supplied by Evides and Vitens (step 1, see again Figure). In order to extract treatment specific information that might be relevant for newly emerging compounds (i.e. compounds that have not been measured yet), the treatment chains are clustered according to unique combinations of treatment steps. These 'treatment clusters' are shown in Table

.8. On the basis of these treatment clusters, removal efficiencies for a set of compounds within the data set can be obtained (step 2). The list of measured removal efficiencies per production site (low uncertainty) and averaged removal efficiencies per treatment cluster (high uncertainty) provide a means to obtain an estimate of the concentration of a compound in tap water when new emission data is available and the configuration of a downstream drinking water production facility is known.

Table	3.8.	Clusters	of	treatment	processes
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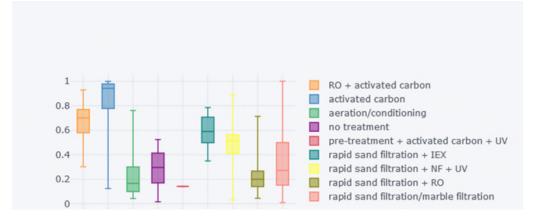
Treatment clusters	Description
	No treatment is applied, i.e.
no treatment	(ground)water is directly distributed
	Reverse osmosis (RO) and activated
RO + activated carbon	carbon filtration (ACF)
activated carbon	ACF
	Aeration and/or conditioning (e.g.
aeration/conditioning	remineralisation or lime softening)
	Pre-treatment (e.g. rapid sand filtration
	and coagulation) is combined with ACF
pre-treatment + activated carbon + UV	and UV disinfection
	rapid sand filtration (RSF) precedes ion
RSF + IEX	exchange
	RSF precedes nanofiltration (NF) and UV
RSF + NF + UV	disinfection
RSF + RO	RSF precedes RO filtration
	RSF or remineralisation by marble
	filtration is the most important
rapid sand filtration/marble filtration	purification step

3.6.3 Results

Removal efficiencies for treatment clusters are calculated based on data as supplied by Evides and Vitens. Average removal efficiencies and the number of compounds within a cluster are shown in Table . Calculated removal efficiencies, including the median, 25 (Q1) and 75 (Q3) percentiles are also depicted as a box-whisker diagram (Figure) with the distance between whiskers defined as the interquartile distance (Q3 – Q1).

Table 3.9.	Removal	efficiencies	per	treatment	cluster
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	Removal efficiency	
	count	average
treatment cluster		
RO + activated carbon	51	0.67
activated carbon	749	0.83
aeration/conditioning	15	0.22
no treatment	27	0.29
pre-treatment + activated carbon + UV	6	0.16
RSF + IEX	89	0.59
RSF + NF + UV	51	0.48
RSF + RO	29	0.22
RSF/marble filtration	1019	0.37





3.6.4 Recommendations

In order to obtain estimates of concentrations of CECs on the basis of emission data, it is recommended to adopt and further develop a workflow that leads to having these estimates. We have proposed such a workflow in Figure . To fulfil this aim, the following is needed:

- Large data sets (monitoring data), comprising multiple measurements of the same compound in influent and drinking water to improve the reliability of removal efficiency estimates based on measurement data alone, and to improve the training of statistical models;
- Calculation of data reliability labels indicating the degree in uncertainty of data of a specific treatment step or compound of emerging concern;
- A database containing data of chemical substances, including structure identifiers and different naming conventions, removal efficiencies, and (literature) source meta data to allow for:
 - o removal efficiency statistics;
 - o advanced statistics to calculate similarity between compounds, and
 - model development (QSPRs, i.e. quantitative structure property relationships) using both compound information and information (properties) about the specific treatment.

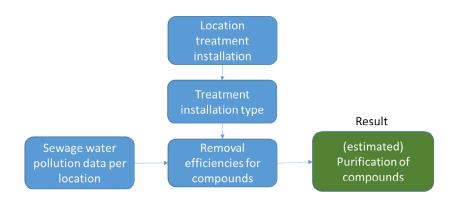
3.7 Abatement by waste water treatment

3.7.1 How do these contribute to purification

Wastewater treatment plants are important for removing pollutants before these enter the water system. Typically, SWWTPs are less advanced than drinking water treatment plants. Humans are less exposed to surface waters than to tap water. Moreover, before pollutants reach sites for drinking water intake, much of the pollutants have been degraded already. Nevertheless, at some occasions pollutants do reach the drinking water intake points (Baken et al. 2016).

3.7.2 Methods

The following data and calculations are needed (figure 3.15, table 3.10):



Figuur 3.15 Schematic overview of the relation between different required data sources.

Table 3.10. Description of the data sources that are needed in the abatement by sewage water treatment plants

Description	Dataset Code (see Attachment)	Status
Locations Sewage water treatment plants (SWTP)	SWWLOC01 SWWLOC02	Public data
Treatment types SWTP	SWWTT01	Public data
Removal efficiencies	SWWCOMP01	Public data
Compounds in waste water	SWWCOMP01	Public data

The Watson database (SWWCOMP01, Table 3.10) contains measurement data on influent and effluent of sewage waste water treatment plants. For industrial waste water plants, there is no public dataset available. Although the Watson database is available and has a lot of data, the measurement data are incomplete (**see 3.2**). Moreover, purification efficiencies may not be accurate. The influent and effluent measurements need to be timed very accurately. If measurements in effluent are hours before or after the influent water body, actually different fragments of the treated water are being compared and removal efficiency estimates are less accurate.

3.7.3 Results

For a total of 519 substances data was available in the Watson database. For 254 of those a removal efficiency was obtained. A total of 40 of those were not removed, but instead increased during the removal treatment. A preliminary analysis showed that on average 75.5% of the concentration of compounds was removed in SWWTPs (not taking into account increasing compounds) (see Figure 3.16).

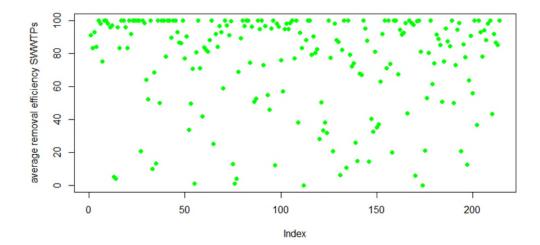


Figure 3.16. The average removal efficiency by SSWTPs for 214 substances in the WATSON database. On the x-axis are the compounds.

3.7.4 Recommendations

As SWWTP effluents potentially reach a large number of surface waters (see **3.2**), it is important to consider their treatment efficiency. Important questions that should be answered are:

- What treatment installation would most effectively remove substances of concern, without forming by-products? What would be the spatial effect of changing treatment installation?
- What SWWTPs have which additional sources aside from grey and black water, for instance small industrial plants?

For the complete picture of purification efficiency of SWWTPs, the treatment train would have to be clear so treatment efficiencies can be extrapolated over SWWTPs where no data is available for particular compounds. This data is available in dataset SWWTT01.

The removal efficiencies of the WATSON database should be explored in more depth, to assess the trustworthiness of the data. For instance, a threshold can be made to prevent the removal efficiency being based on a single measurement.

4 Conclusions and recommendations

4.1 Relevance and potential of the water quality map

Coppens et al., (2015) and van Wezel et al., (2018) focused mainly on relative influence of point sources on receiving waters. The water quality map can be used for more purposes than that. With the project team and based on results in this and former results of Coppens et al., 2015 and van Wezel et al., 2018 possible applications of the water quality map were listed:

- Understand and map all sources for one substance
- Predict overall spatial toxic pressure, and mixture effects (with added toxicological and effect data)
- Retrace pollution to potential sources and quantify contributions
- Prioritize abatement options for pollution per surface water
- Predict spatially explicit what possible compounds can cause problems
- Prioritize and adapt measurements based on prediction of possible compounds
- Risk assessment based on prediction of possible compounds
- Scenario studies on the effectivity of (local) abatement options or effects of population growth, demographics, and climate

4.2 Obstacles and limitations

The biggest limitations for the construction of a water quality map are incomplete (emission) data, and data of unclear origin and accuracy. For the diffuse emissions from agriculture, implementing groundwater modelling techniques required investing in additional resources.

Furthermore, close collaboration with Deltares on hydrology is essential for the success of future work on the water quality map, both for interpretation of the local hydrological results and e.g. modeling of new (point) sources.

Although the hydrology model matrices calculated by the WFD-explorer model are very useful, these have their limitations. Because the hydrology is a quarterly average, detailed dynamics or peaks in emissions cannot be captured. Emissions in reality can happen either continuously, or discontinuously, erratic in size and frequency and period. Deltares is in the process of updating the WFD-explorer to modeling fluxes on a more detailed (daily) timescale. This model could for some of the applications be more accurately reflecting the real situation. This does not mean that the end-state matrices as used in this report are not useful anymore. These could still provide novel insights for the coming years.

4.3 Recommendations

What is possible now?

We have identified possibilities and points of attention for the different emission sources that will contribute to a water quality map, knowledge on the spread of

antimicrobial resistance, and abatement options by purification treatment. With this, we have laid a foundation on which to build in future work.

What is needed to make a similar approach successful in the future?

It is recommended to improve administration and general referral to data sources. It was not easy to recover data sources mentioned in other, related work. It is also recommended to improve reproducibility of research done in general, and have a thorough description of processing of data and results. Although there is centralization of knowledge per source of emission, the knowledge and data of emissions to the water system via the different sources is not centralized. Dedicated 'emission specialists' could keep knowledge updated.

The emissions of industrial waste water plants are for the larger part unknown. There is no obligation for industries to maintain or make public a full list of all known emitted substances.

The availability of datasets in general can be problematic. For proprietary data, often it is not known what processing (if any) was done on the data. Public data is in many cases much better described, and accessible. This will aid in doing reproducible and replicable research.

The constituents of the water quality map can best be put to use in concrete questions that arise. The versatility in questions that can be solved (see 'what questions can we answer' in the results sections in 3.1.3, 3.2.3, 3.3.3, 3.4.3, 3.5.3) exemplify the broad potential usage. Each application could in potential have different requirements to the quality and quantity of the data. For a water quality map of a single compound, for instance, more detailed data can be gathered on emission sources and compound characteristics that will influence the spread of the compound over the different surface waters. The same goes for a water quality map for a single emission point; the specific characteristics of the emission point can be taken into account on a more detailed level than in a general water quality map of all emission sources.

4.4 Conclusion

This report provides an overview of the collected data and recommendations that can be used for future investigations regarding the spatial influence of different emission sources. The water quality map can give good insights into predicted water quality and abatement options. For individual emission routes 'SWWTP' and 'cross-border Rivers' a concept map was constructed, and these are ready for further analysis. Many partial questions can be answered. However, the availability, completeness and quality of data are at present limitations to construct a complete water quality map for the Netherlands. Chapter 3 and Attachment I provide a comprehensive overview of the data currently available and provides a resource for future studies. For similar studies to be successful, it is essential to have good policies for data accessibility and management.

Work on the water quality map will be continued in the 'Kennisimpuls Waterkwaliteit' (KIWK) project 'mengseltoxiciteit' within a collaborative framework with Deltares, WUR and RIVM. Furthermore, the 'Schone Maaswaterketen' develops the 'Prototype Geoportaal Atlas verontreinigingsbronnen' that uses elements from this work. The project 'Bedreigingen van bronnen' will use the matrices to model the spread of two drug-related chemicals from emitting SWWTPs. Lastly, the matrices and insights are currently used in the WiCE-project 'Zuinig met Zoet' where potential of reuse of water in terms of quality and quantity as affected by SWWTPs is evaluated.

Attachment I Data Sources

0. Common datasets

CDC01: Predicted degradation rate constants (half-lives) of 800.000 pollutants ('Opera' model). <u>https://www.epa.gov/chemical-research/distributed-structure-searchable-toxicity-dsstox-database</u> and click DSSToxData. Or go via: <u>ftp://newftp.epa.gov/COMPTOX/Sustainable_Chemistry_Data/Chemistry_Dashboard</u> and click DSSTox_Predicted_NCCT_model.zip. Four files are here, each containing 200.000 parameters.

VALIDAT01: Validation data, RIWA base, measured substances at eight locations along the Rhine and Meuse. Proprietary data, permission granted on a case by case basis by RIWA.

1. Cross-border Rivers

BLRLOC01: Locations of rivers from where they emit to the Dutch surface waters.

BLRFLOW01: The discharge for all surface waters, for a dry and a wet quarter. Obtained from Deltares.

BLRFLOW02: discharge for the Rhine at Lobith and Eijsden in time via <u>https://waterinfo.rws.nl/</u>

VALIDAT01: Validation data. Monitoring data on compounds that enter the Dutch surface waters via the Meuse and the Rhine. Monthly discharge for Lobith and Eijsden in time: RIWA base (not public, permission on a case by case basis)

HMBLR01, HMBLR02: Hydrology matrices for surface waters connected to pollution from cross-border Rivers. Proprietary data KWR, purchased from Deltares.

2. Sewage Waste Water Plants

SWWLOC01, SWWTT01: European Commission urban waste water website. SWWTP locations via http://uwwtd.oieau.fr/Netherlands/download, pick

http://www.uwwtd.oieau.fr/services/ows/?service=WFS&version=1.1.0&request=GetFeat ure&typeName=UWWTD:UWWTD_Netherlands_UrbanWasteWaterTreatmentPlant&CQL_FI LTER=UWWTD:repReportedPerdiod=2014&outputFormat=csv "Urban Waste Water Treatment plants.csv". This provides 415 Dutch SWWTPs with locations and loads (Population equivalents). The full details on model and dictionary are available on EEA website here: http://cdr.eionet.europa.eu/help/UWWTD/UWWTD_613

SWWLOC02: Emissieregistratie. Locations of 324 SWWTPs in the Netherlands. Accessible via <u>http://www.emissieregistratie.nl/erpubliek/misc/Documenten.aspx</u> Algemeen> Exports>Exports Belasting naar Water (Exports Load to Water)> ER1990-2016_krw_2016.zip, tabblad bestemming_individueel_2016, location as lig_x and lig_y in columns. **SWWCOMP01**: Watson database. Substances measured per Sewage Waste Water Plant in the Netherlands http://www.emissieregistratie.nl/erpubliek/erpub/wsn/default.aspx This dataset holds concentrations (µg/l) or loads (mg/day) of in total 918 compounds from RWZI effluent or influent. Also possible per SWWTP.

SWW03: Emissie registratie. Substances emitted via various sources in the Netherlands. Accessible via http://www.emissieregistratie.nl/erpubliek/misc/Documenten.aspx Algemeen> Exports>Exports Belasting naar Water (Exports Load to Water)> ER1990-2016_krw_2016.zip, tabblad belasting_eindb_compleet_2016, emissions in kg with GOF code 'stof' (compound) en EMK-code 'emissieoorzaak' (emission source, also WWPTs) en GAF 'afwateringseenheid' (surface water).

SWWLOAD01: CBS data. Total load per SWWTP in m3/day, given per year. Bron: CBS, 2018. RwziBase 2016, database met microdata van rioolwaterzuiveringsinstallaties. Centraal Bureau voor de Statistiek, Den Haag/Heerlen. Not publicly available. For publicly available loads in terms of inhabitant equivalents, see SWWLOC01.

HMSWW01, HMSWW02: Hydrology matrices for surface waters connected to pollution from SWWTPs. Proprietary data KWR, purchased from Deltares.

3. Industrial Waste Water Plants

IWWLOC01 / IWWCOMP04: <u>http://ftp.eea.europa.eu/www/eprtr/v16/E-</u> <u>PRTR_database_v16_xls.zip</u> Here are three files: 'waste transfers', 'pollutant releases', 'pollutant transfers' for different countries. Data for 115 Dutch Industrial facilities emitting to water. Emissions per IAZI and industry type per compound (91 total)

IWWLOC02 / IWWCOMP01 / IWWLOAD01: Emissieregistratie. Locations of 212 IWWTPs in the Netherlands. Accessible via

http://www.emissieregistratie.nl/erpubliek/misc/Documenten.aspx Algemeen> Exports>Exports Belasting naar Water (Exports Load to Water)> ER1990-2016_krw_2016.zip, sheet 'bestemming_individueel_2016', location as 'lig_x' and 'lig_y' in columns. Sheet 'belasting_eindb_compleet_2016', emissions in kg with GOF code 'stof' (compound) and EMK-code 'emissieoorzaak' (emission source, also WWPTs) and GAF 'afwateringseenheid' (surface water). Combine this with sheet 'Emissieoorzaak' which holds the Industry code (EMP-code) and industry type. For estimating the load, the TOC emission from the sheet 'Belasting_individueel_compleet_2016' can be used ('gof_code' for TOC is 549).

IWWCOMP02: CBS emission per industry code, alternative from statline for total emission per industry.

https://opendata.cbs.nl/statline/#/CBS/nl/dataset/81353ned/table?ts=154451962813 <u>7.</u>CBS classification to Industrial sector/class per IAZI (NACE code) https://www.cbs.nl/en-gb/our-services/methods/classifications/activiteiten/standardindustrial-classifications--dutch-sbi-2008-nace-and-isic--#id=the-structure-of-sbi-2008version-2018-0 Nace code = 1e 4 nrs SBI code

IWWCOMP03: searched Google with the search phrase: "watervergunning lozing filetype:pdf". Resulting PDFs with texts on substances are public and accessible online.

HMIWW01, HMIWW02: Hydrology matrices for surface waters connected to pollution from IWWTPs. Proprietary data KWR, purchased from Deltares.

4. Diffuse emissions from agriculture

BASLOC01: The locations of the basins from which water enters the Dutch surface waters. Proprietary data, Deltares.

CULTLOC01: Parcels with agriculture in 2016

https://data.overheid.nl/dataset/52172-basisregistratie-gewaspercelen--brp-)

CULTCOMP01: Compounds in kg /ha per culture at: https://opendata.cbs.nl/statline/portal.html?_la=nl&_catalog=CBS&tableId=84010NED& _theme=234

HMBAS01, HMBAS02: Hydrology matrices for surface waters connected to pollution from agriculture diffuse sources. Proprietary data KWR, purchased from Deltares.

5. Abatement by water treatment for drinking water

D_Evides: data set containing concentrations of organic micropollutants in surface intake water and drinking water measured at several production sites (Berenplaat, Braakman, Kralingen) of Evides obtained via a BTO project, via contact: prof. Annemarie van Wezel.

D_Vitens: data set containing concentrations of organic micropollutants in intake water (mostly groundwater) and drinking water of all the production sites of Vitens obtained in a former BTO project, via contact: Rosa Sjerps.

6. Abatement by water treatment for Sewage waste water

SWWCOMP01: Watson database. Substances measured per Sewage Waste Water Plant in the Netherlands http://www.emissieregistratie.nl/erpubliek/erpub/wsn/default.aspx This dataset holds concentrations (µg/l) or loads (mg/day) of compounds from RWZI effluent or influent.

SWWTT01: RWZI treatment trains. European Commission urban waste water website via http://uwwtd.oieau.fr/Netherlands/download, pick

http://www.uwwtd.oieau.fr/services/ows/?service=WFS&version=1.1.0&request=GetFeat ure&typeName=UWWTD:UWWTD_Netherlands_UrbanWasteWaterTreatmentPlant&CQL_FI LTER=UWWTD:repReportedPerdiod=2014&outputFormat=csv "Urban Waste Water Treatment plants.csv". This provides 415 Dutch SWWTPs with locations and loads (Population equivalents). The full details on model and dictionary are available on EEA website here: http://cdr.eionet.europa.eu/help/UWWTD/UWWTD_613 BTO | October 2019

7. Gene and antibiotics

References - literature review as data set

- An, X. L., Su, J. Q., Li, B., Ouyang, W. Y., Zhao, Y., Chen, Q. L., ... Zhu, Y. G. (2018). Tracking antibiotic resistome during wastewater treatment using high throughput quantitative PCR. Environment International, 117(December 2017), 146-153. https://doi.org/10.1016/j.envint.2018.05.011
- 2 Ben, W., Wang, J., Cao, R., Yang, M., Zhang, Y., & Qiang, Z. (2017). Distribution of antibiotic resistance in the effluents of ten municipal wastewater treatment plants in China and the effect of treatment processes. Chemosphere, 172, 392-398. https://doi.org/10.1016/j.chemosphere.2017.01.041
- Blaak, H., De Kruijf, P., Hamidjaja, R. A., Van Hoek, A. H. A. M., De Roda Husman, A. M., & Schets, F. M. (2014). Prevalence and characteristics of ESBL-producing E. coli in Dutch recreational waters influenced by wastewater treatment plants. Veterinary Microbiology, 171(3-4), 448-459. https://doi.org/10.1016/j.vetmic.2014.03.007
- Blaak, H., Lynch, G., Italiaander, R., Hamidjaja, R. A., Schets, F. M., & Husman, A. M. de R. (2015). Multidrug-Resistant and Extended Spectrum Beta-Lactamase-Producing Escherichia coli in Dutch Surface Water and Wastewater. PLoS ONE, 10(6), 1–16. https://doi.org/10.1371/journal.pone.0127752
- 5 Chen, H., & Zhang, M. (2013). Effects of advanced treatment systems on the removal of antibiotic resistance genes in wastewater treatment plants from Hangzhou, China. Environmental Science and Technology, 47(15), 8157-8163. https://doi.org/10.1021/es401091y
- 6 Di Cesare, A., Eckert, E. M., D'Urso, S., Bertoni, R., Gillan, D. C., Wattiez, R., & Corno, G. (2016). Co-occurrence of integrase 1, antibiotic and heavy metal resistance genes in municipal wastewater treatment plants. Water Research, 94, 208-214. https://doi.org/10.1016/j.watres.2016.02.049
- 7 Gómez, M., Díaz, M. T., Araujo, M., Sueiro, R., & Garrido, J. (2010). Waste water treatment plants as redistributors of resistance genes in bacteria. WIT Transactions on Ecology and the Environment, 135(January), 83-94. https://doi.org/10.2495/WP100081
- 8 Harris, S., Morris, C., Morris, D., Cormican, M., & Cummins, E. (2014).
 Antimicrobial resistant Escherichia Coli in the municipal wastewater system: Effect of hospital effluent and environmental fate. Science of the Total Environment, 468-469, 1078-1085. https://doi.org/10.1016/j.scitotenv.2013.09.017
- Korzeniewska, E., Korzeniewska, A., & Harnisz, M. (2013). Antibiotic resistant Escherichia Coli in hospital and municipal sewage and their emission to the environment. Ecotoxicology and Environmental Safety, 91, 96-102. https://doi.org/10.1016/j.ecoenv.2013.01.014
- 10 Laht, M., Karkman, A., Voolaid, V., Ritz, C., Tenson, T., Virta, M., & Kisand, V. (2014). SI - Abundances of tetracycline, sulphonamide and beta-lactam antibiotic resistance genes in conventional wastewater treatment plants (WWTPs) with different waste load. PLoS ONE.
- 11 Lan, L., Kong, X., Sun, H., Li, C., & Liu, D. (2019). High removal efficiency of antibiotic resistance genes in swine wastewater via nanofiltration and reverse osmosis processes. Journal of Environmental Management, 231(August 2018), 439-445. https://doi.org/10.1016/j.jenvman.2018.10.073
- 12 Li, B., Qiu, Y., Li, J., Liang, P., & Huang, X. (2019). Removal of antibiotic resistance genes in four full-scale membrane bioreactors. Science of the Total Environment, 653, 112–119. https://doi.org/10.1016/j.scitotenv.2018.10.305
- 13 Li, N., Sheng, G. P., Lu, Y. Z., Zeng, R. J., & Yu, H. Q. (2017). Removal of antibiotic resistance genes from wastewater treatment plant effluent by coagulation. Water Research, 111, 204–212. https://doi.org/10.1016/j.watres.2017.01.010
- 14 Luczkiewicz, A., Felis, E., Ziembinska, A., Gnida, A., Kotlarska, E., Olanczuk-Neyman, K., & Surmacz-Gorska, J. (2013). Resistance of Escherichia Coli and Enterococcus spp. to selected antimicrobial agents present in municipal

wastewater. Journal of Water and Health, 11(4), 600-612. https://doi.org/10.2166/wh.2013.130

- Mao, D., Yu, S., Rysz, M., Luo, Y., Yang, F., Li, F., ... Alvarez, P. J. J. (2015). Prevalence and proliferation of antibiotic resistance genes in two municipal wastewater treatment plants. Water Research, 85, 458-466. https://doi.org/10.1016/j.watres.2015.09.010
- 16 McConnell, M. M., Truelstrup Hansen, L., Jamieson, R. C., Neudorf, K. D., Yost, C. K., & Tong, A. (2018). Removal of antibiotic resistance genes in two tertiary level municipal wastewater treatment plants. Science of the Total Environment, 643, 292–300. https://doi.org/10.1016/j.scitotenv.2018.06.212
- 17 Munir, M., Wong, K., & Xagoraraki, I. (2011). Release of antibiotic resistant bacteria and genes in the effluent and biosolids of five wastewater utilities in Michigan. Water Research, 45(2), 681-693. https://doi.org/10.1016/j.watres.2010.08.033
- 18 Quach-Cu, J., Herrera-Lynch, B., Marciniak, C., Adams, S., Simmerman, A., & Reinke, R. A. (2018). The effect of primary, secondary, and tertiary wastewater treatment processes on antibiotic resistance gene (ARG) concentrations in solid and dissolved wastewater fractions. Water (Switzerland), 10(1), 13–18. https://doi.org/10.3390/w10010037
- 19 Rijksinstituut voor Volksgezondheid en Milieu. (2017). Watson Database. Retrieved March 14, 2019, from http://www.emissieregistratie.nl/erpubliek/erpub/wsn/default.aspx
- Schijven, J. F., Blaak, H., Schets, F. M., & De Roda Husman, A. M. (2015). Fate of Extended-Spectrum β-Lactamase-Producing Escherichia Coli from Faecal Sources in Surface Water and Probability of Human Exposure through Swimming. Environmental Science and Technology, 49(19), 11825–11833. https://doi.org/10.1021/acs.est.5b01888
- 21 Schmitt, H., Blaak, H., Kemper, M., van Passel, M., Hierink, F., van Leuken, J., ... Zuidema, T. (2017). Bronnen va antibioticaresistentie in het milieu en mogelijke maatregelen. Bilthoven, Netherlands. https://doi.org/10.21945/RIVM-2017-0058
- Tao, C. W., Hsu, B. M., Ji, W. T., Hsu, T. K., Kao, P. M., Hsu, C. P., ... Huang, Y. L. (2014). Evaluation of five antibiotic resistance genes in wastewater treatment systems of swine farms by real-time PCR. Science of the Total Environment, 496, 116-121. https://doi.org/10.1016/j.scitotenv.2014.07.024
- 23 Wang, J., Mao, D., Mu, Q., & Luo, Y. (2015). Fate and proliferation of typical antibiotic resistance genes in five full-scale pharmaceutical wastewater treatment plants. Science of the Total Environment, 526, 366-373. https://doi.org/10.1016/j.scitotenv.2015.05.046
- 24 Wang, M., Shen, W., Yan, L., Wang, X. H., & Xu, H. (2017). Stepwise impact of urban wastewater treatment on the bacterial community structure, antibiotic contents, and prevalence of antimicrobial resistance. Environmental Pollution, 231, 1578-1585. https://doi.org/10.1016/j.envpol.2017.09.055
- 25 Wen, Q., Yang, L., Duan, R., & Chen, Z. (2016). Monitoring and evaluation of antibiotic resistance genes in four municipal wastewater treatment plants in Harbin, Northeast China. Environmental Pollution, 212, 34-40. https://doi.org/10.1016/j.envpol.2016.01.043
- 26 Yuan, L., Li, Z. H., Zhang, M. Q., Shao, W., Fan, Y. Y., & Sheng, G. P. (2019). Mercury/silver resistance genes and their association with antibiotic resistance genes and microbial community in a municipal wastewater treatment plant. Science of the Total Environment, 657, 1014–1022. https://doi.org/10.1016/j.scitotenv.2018.12.088
- Zhang, Y., Li, A., Dai, T., Li, F., Xie, H., Chen, L., & Wen, D. (2018). Cell-free DNA: A Neglected Source for Antibiotic Resistance Genes Spreading from WWTPs. Environmental Science and Technology, 52(1), 248-257. https://doi.org/10.1021/acs.est.7b04283
- Zheng, W., Wen, X., Zhang, B., & Qiu, Y. (2019). Selective effect and elimination of antibiotics in membrane bioreactor of urban wastewater treatment plant.
 Science of the Total Environment, 646, 1293-1303. https://doi.org/10.1016/j.scitotenv.2018.07.400

Attachment II References

- Baken, K., Kolkman, A., Van Diepenbeek, P., Ketelaars, H., and Van Wezel, A. 2016.
 'Signallling "Other Antropogenic Substances", and Then? The Pyrazole Case'. *H2O online*, September 2016.
 https://www.h2owaternetwerk.nl/index.php/vakartikelen/signalering-vanoverige-antropogene-stoffen-en-dan-de-pyrazool-casus.
- van Batenburg-Eddes, T., van den Berg-Jeths, A., van der Veen, A.A., Verheij, R.A. and de Neeling, A. (2002) Consumption in the Netherlands, regional variations in consumption of pharmaceuticals. "Slikken in Nederland. Regionale variaties in geneesmiddelengebruik", p. 76, RIVM, Bilthoven.
- Bertelkamp, C., et al. (2019). DPWE Robuustheid uitvoering doseerproeven 2017/2018 -Resultaten van doelstofanalyses, non-target screening en bioassays. Nieuwegein, KWR.
- Coppens, L.J.C., van Gils, J.A.G., ter Laak, T.L., Raterman, B.W., van Wezel, A.P. 2015. Towards spatially smart abatement of human pharmaceuticals in surface waters: defining impact of sewage treatment plants on susceptible functions. Water Res., 81, pp. 356-365
- Danner, M.C., Robertson, A., Behrends V., and Reiss, J. 2019. Antibiotic pollution in surface fresh waters: Occurrence and effects. Science of the Total Environment 664 (2019) 793-804
- Dutch Drinking water directive (2018) https://wetten.overheid.nl/BWBR0030111/2018-07-01
- Fatta-Kassinos, D., Michael-Kordatou, I., Ioannou-Ttofa, L., Toumazi, T., Medema, G.J., Paulus, G.K. and Hornstra, L.M. (2017) Antibiotics and mobile resistance elements in wastewater reuse applications - Answer H2020-MSCA-ITN-2015/675530, p. 7, European Union Horizon 2020.
- Fischer, Astrid, Thomas ter Laak, Jan Bronders, Nele Desmet, Ekkehard Christoffels, Annemarie van Wezel, and Jan Peter van der Hoek. 2017. 'Decision Support for Water Quality Management of Contaminants of Emerging Concern'. *Journal of Environmental Management* 193 (May): 360-72. https://doi.org/10/gfwgsv.
- Hoek, J. P. van der, C. Bertelkamp, A. R. D. Verliefde, and N. Singhal. 2014. 'Drinking Water Treatment Technologies in Europe: State of the Art - Challenges -Research Needs'. *Journal of Water Supply: Research and Technology-Aqua* 63 (2): 124-30. https://doi.org/10/f5wn2n.
- Hofs, B., et al. (2014). Robuustheid zuiveringen DPW 2012-2103; zomer en winter. Nieuwegein, KWR: 139.

- Hornstra, L. (2017) Antibioticaresistentie-genen in oppervlakte-water en in drinkwater zuiveringsprocessen, p. 47, KWR, Nieuwegein.
- Houtman, Corine J., Jan Kroesbergen, Karin Lekkerkerker-Teunissen, and Jan Peter van der Hoek. 2014. 'Human Health Risk Assessment of the Mixture of Pharmaceuticals in Dutch Drinking Water and Its Sources Based on Frequent Monitoring Data'. *Science of The Total Environment* 496 (October): 54-62. https://doi.org/10/f6h97q.
- Howard, P.H. (1991) Handbook of Environmental Fate and Exposure Data for Organic Chemicals, Pesticides, vol (III. ed.), Lewis Publishers, Chelsea, MI
- ter Laak, T. (2012) Mobility of antibiotic resistance genes in the environment and potential threats for drinking water, p. 18, KWR, Nieuwegein.
- ter Laak, T.L., Raterman, B. and Meijers, E. (2016) Ruimtelijke modellering van geneesmiddelen in het stroomgebied van de Dommel, p. 38, KWR, Nieuwegein.
- ter Laak, T.L, Kooij, P.J.F., Tolkamp, H., Hofman, J.A.M.H. (2014) Different compositions of pharmaceuticals in Dutch and Belgian rivers explained by consumption patterns and treatment efficiency – Environmental Science and Pollution Research 21-22 p.12843-12855)
- Lahr, J. and F. van den Berg (2009). Uitspoelconcentraties en persistentie van antibiotica in de bodem berekend met het GeoPEARL 3.3.3 model. Wageningen, the netherlands, Alterra: 29.
- Kroesbergen, J., ter Laak, T.L. and Kors, L.J. (2018) Stofspecifieke Risicoschatting innamestops, p. 25, KWR, Nieuwegein.
- Lee, Yunho, and Urs von Gunten. 2012. 'Quantitative Structure-Activity Relationships (QSARs) for the Transformation of Organic Micropollutants during Oxidative Water Treatment'. *Water Research* 46 (19): 6177-95. https://doi.org/10/gfwgwx.
- Liefting, E., de Man, H. 2017. Achtergrondrapport bij de in 2017 geactualiseerde factsheet 'Effluenten RWZI's, regenwaterriolen, niet aangesloten riolen, overstorten en IBA's'. Deltares, Rijksinstituut voor Volksgezondheid en Milieu.
- Mansouri K, Grulke CM, Judson RS, Williams AJ. 2018. OPERA models for predicting physicochemical properties and environmental fate endpoints. Journal of Cheminformatics 10(1):10 10.1186/s13321-018-0263-1
- Munthe, J., Brorström-Lunden, E., et al. (2017) An expanded conceptual framework for solution-focused management of chemical pollution in European waters. Environmental Sciences Europe 29:1, art. no. 13
- Pieters, B. (2011) Verbetering schatting effluentvrachten RWZI's, p. 102, Grontmij, Amsterdam, the Netherlands.
- Schmitt, H., H. Blaak, M. Kemper, M. van Passel, F. Hierink, J. van Leuken, A. M. De Roda Husman, et al. 2017. "Bronnen va Antibioticaresistentie in Het Milieu En

Mogelijke Maatregelen." Bilthoven, Netherlands. https://doi.org/10.21945/RIVM-2017-0058.

- Schmitt, Heike, Thomas ter Laak, and Karen Duis. 2013. "Development and Dissemination of Antibiotic Resistance in the Environment under Environmentally Relevant Concentrations of Antibiotics and Its Risk Assessment."
- Sjerps, R., et al. (2016). Effect van klimaatverandering en vergrijzing op waterkwaliteit en drinkwaterfunctie van Maas en Rijn. H2O-Online.
- Smeets, P. W. M. H., et al. (2010). "Practical applications of quantitative microbial risk assessment (QMRA) for water safety plans." Water Science and Technology 61(6): 1561-1568.
- Stuyfzand, P. J. (2019). Predicting organic micropollutant behavior for standardized public supply well field types, with TRANSATOMIC Lite. Nieuwegein, KWR: 39.
- Vergouwen, A.A., Pieters, B.J. and Kools, S. (2011) Inventarisatie van emissie van geneesmiddelen uit zorginstellingen, STOWA, Amersfoort, The Netherlands.
- Vink, C., Bonte, M., Puijker, L., Crijns, J.W.A.M., 2011. Pesticidenonderzoek Zuid Limburg: Pesto 1. Fase 1b en c: validatie, ruwwaterprognose en maatregelen. KWR 2011.098.
- Vries, Dirk, Bas A. Wols, and Pim de Voogt. 2013. 'Removal Efficiency Calculated Beforehand: QSAR Enabled Predictions for Nanofiltration and Advanced Oxidation'. Water Science and Technology: Water Supply 13 (6): 1425-36. https://doi.org/10/gdcdhs.
- Water Framework Directive (2000) http://ec.europa.eu/environment/water/waterframework/index_en.html
- van Wezel, Annemarie P. van, Floris van den Hurk, Rosa M. A. Sjerps, Erwin M. Meijers, Erwin W. M. Roex, and Thomas L. ter Laak. 2018a. 'Impact of Industrial Waste Water Treatment Plants on Dutch Surface Waters and Drinking Water Sources'. *Science of The Total Environment* 640-641 (November): 1489-99. https://doi.org/10/gfwhm4.
- van Wezel, Annemarie P. van, Floris van den Hurk, Rosa M. A. Sjerps, Erwin M. Meijers, Erwin W. M. Roex. 2018b. 'Impact of Industrial Waste Water Treatment Plants on Dutch Surface Waters and Drinking Water Sources'. KWR rapport - KWR 2018.006

WFD-explorer: https://publicwiki.deltares.nl/display/KRWV/KRW-Verkenner

- Wols, Bas A., and Dirk Vries. 2012. 'On a QSAR Approach for the Prediction of Priority Compound Degradation by Water Treatment Processes'. Water Science and Technology 66 (7): 1446–53. https://doi.org/10/gdcdhr.
- World Health Organization. 2014. "Antimicrobial Resistance Global Report on Surveillance." <u>https://doi.org/10.17226/6121</u>.

- Yujie Ben, Caixia Fua, Min Hua, Lei Liua, Ming Hung Wong, Chunmiao Zheng 2019. Human health risk assessment of antibiotic resistance associated with antibiotic residues in the environment: A review. Environmental Research 169 (2019) 483-493
- Zhang, Han, Miaomiao Du, Hongyou Jiang, Dandan Zhang, Lifeng Lin, Hong Ye, and Xian Zhang. 2010. "Occurrence, Seasonal Variation and Removal Efficiency of Antibiotics and Their Metabolites in Wastewater Treatment Plants, Jiulongjiang River Basin, South China." Environmental Science: Processes & Impacts 3 (2): 225-34. https://doi.org/10.1039/b916098a.