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Removal requirement and purification treatment effort for Dutch Rhine water from 2000-2018

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1

Introduction

The Water Framework Directive (2000/60/EC) (WFD) has been the most comprehensive instrument of European Union water policy since its introduction in 2000. The main objective of the WFD is to protect and improve freshwater bodies with the aim of achieving good status in European waters. The main instruments for implementing the directive are the 'river basin management plans' (RBMPs) and the 'programs of measures' drawn up in six-year cycles.

In the year 2019, the European Commission is carrying out a fitness check for the WFD and its daughter directives, the Groundwater Directive (2006/118/EC) and the Directive priority substances (2008/105/EC). The purpose of a fitness check is to assess how effective and efficient the legislation is in achieving the set policy objectives. In that view this is a good moment to see what the introduction of the WFD has meant for the quality of water bodies and more specifically in light of improving drinking water sources.

In the WFD, preamble 24 states "Good water quality contributes to securing the drinking water supply of the population." Next, article 7 contains statements related to water used for drinking water:

- WFD article 7.1 requires member states to designate water bodies for the production of drinking water
- WFD article 7.2 states that water quality objectives must be achieved in these water bodies
- WFD Article 7.3 states that "*Member States shall ensure the necessary protection for the bodies of water identified with the aim of avoiding deterioration in their quality in order to reduce the level of purification treatment required in the production of drinking water.*"

Looking back over the years since the introduction of the WFD, the question arises to what extent the WFD was able to prevent deterioration of water quality. In light of the fitness check, Article 7.3 is perhaps one of the most relevant, but lacks a quantitative measure. Therefore, the main goal of this study is to develop a quantitative measure to assess water quality in light of the level of required purification treatment.

As water companies strive for and have to meet strict requirements for drinking water, measures are put in place. Besides extensive monitoring and early warning systems, water companies invest in ways of treatment options, which can be nature-based solutions or advanced technology. The required level of purification depends on the requirements that are set for drinking water and the water quality in the designated drinking water body. The difference between the water quality at the source and the drinking water requirements can be considered as the removal requirement. The effort to do so naturally depends on how difficult it is to remove the chemicals that require removal. The challenge is how water quality in terms of these elements can be expressed and evaluated in a concise way that provides insight into the challenges in evaluating the reduction of the level of purification treatment.

1.1 Water quality indices

'Water quality' is an aspect with many components and a common way to summarize this can be to generate a more simple 'water quality index' (WQI). In the European Water Framework Directive, next to chemical status, the biological quality elements are assessed but not expressed in one index: phytoplankton, aquatic flora, benthic invertebrates, fish (e.g. Birk et al., 2012). In this report, the focus lies on chemical data and its relation to the drinking water in particular. Indices based on chemical data are used since long to communicate water quality in a single aggregated score that is representative of quality impairments (Hurley et al., 2012). In general, any index is composed of the same steps. First, the parameters that are considered to represent water quality are chosen. For each parameter, a sub-index is established. Weights are introduced to give more or less weight to some parameters. Finally, the sub-indices are aggregated according to some function, into one index (Borges Garcia, 2018).

Many indices have been introduced over the years (Borges Garcia, 2018). One of the first was Horton (1965) who introduced an index that consisted of a weighted sum of a maximum of ten subscripts, divided by the sum of the weights. This number was multiplied by two coefficients related to temperature and pollution of a watercourse. Since then, some of the more well-known variations on this are the WQI-NSF (Brown et al., 1970) and variations on that, in which nine (or ten) specific parameters with their specifically determined and assigned weights are aggregated. A WQI by Bascaron (1979) balances the influence of each of the parameters on the final value of the index by normalisation. Also, other parameters can be added in this index. The O-WQI (Cude, 2001) is a variant where the weights are omitted. The WQI-CCME was introduced by the Canadian Council of Ministers of the Environment in 2001 (CCME, 2001). Three elements determine the score: the amount of parameters that at least once do not meet the target value, the percentage of samples that do not meet the target value, the size of the deviation from the target value.

Only more recently, efforts are being directed at developing WQIs specific for assessing treatability of water for the production of drinking water. Hurley et al (2012) describe a variation on the CCME water quality index that relates to the purification treatment effort. A fixed core set of eight parameters (temperature, total organic carbon, turbidity, pH, E. coli, nitrate, total coliforms and iron) reflect the common water concerns. Drinking source target values are established, based on expected treatment efficiency of two treatments: chlorination and chlorination preceded by slow sand filtration. Deviations to this target value (from zero to a maximum of a hundred) were taken as the quality impairment. Parameter unevenness was compensated. Dutra de Oliveira et al. (2019) used the WQI-NSF as a basis to develop the RW-WQIF for raw water quality for purification by conventional treatment process, using parameters representing chemistry (e.g. manganese), biology (e.g. E. coli), and physical state (e.g. apparent colour) and deriving values with a Fuzzy logic algorithm. They could relate the quality as predicted by the index to required coagulant dose and treated water turbidity. Van den Doel et al. (2019) adjusted the WQI-CCME for assessing micro-pollutants removal effort by using a very broad selection of chemical parameters, taking the values from the Dutch Drinking Water Decree (DWB, 2018) as their target value and adding a score for their removability based on Gibbs free energy.

For our purposes, evaluate and communicate the reduction of the level of purification treatment over years, we require a simple, easy to understand measure that on the one hand links source water values to drinking water target values, and in addition takes into account that water quality is not static, referring to the phenomenon that new emerging chemicals can contribute to the purification treatment level.

1.2 Indications for Removal Requirement (RR) and Purification Treatment Effort (PTE)

We define and describe an index to represent removal requirement in this report and apply this measure to evaluate the water quality of the Rhine at drinking water production abstraction points. We name this index the Water Quality Index Removal Requirement (WQI RR). Based on this index we assess whether, since the introduction of the WFD with its Article 7.3 in the year 2000, the quality of the Rhine as a source for our drinking water has indeed not deteriorated and the required removal has decreased. In addition, we develop a water quality index to represent purification treatment effort (WQI PTE). This is intended to evaluate how hard it is to meet the removal of substances. If the parameters that need removal are easy-to-remove substances, the purification treatment effort will be relatively low. If purification treatment effort is low, less advanced techniques or less capacity is needed, resulting in lower investment costs or lower energy and other operational costs. The other way around, if the removal requirement consists of hard-to-remove substances, the purification treatment effort will be high. The WQI PTE will allow an evaluation of the source water quality based on purification treatment effort of the substances. Combined, these two indices can evaluate to what extent the parameters that exceed their target value in the DWB (2018) to a certain level, can be removed and if this extent has improved during the years the WFD has been in use.

2

Methodology

2.1 Calculation of the WQI RR

We evaluate the quality of source water, i.e. water at the abstraction locations for drinking water that is to be treated to a certain degree, with a water quality index representing the removal requirement (WQI RR). The values for each parameter minimally meet the target values for concentrations of parameters in the Dutch Drinking Water Decree (DWB, 2018) (https://wetten.overheid.nl/BWBR0030111/2018-07-01). The removal requirement is defined very simply as the percentage of removal that is needed such that a substance concentration falls below the corresponding DWB value after treatment. Table 1 lists the water quality parameters with their DWB values.

Table 1 Values taken from the Drinking Water Decree (DWB, 2018) that have been tested against. These consist of standards, indicators, and guiding values for parameters from the Drinking Water Decree including operational, organoleptic, aesthetic and signalling parameters. Biological parameters are not included.

Parameter	CAS-number	Maximum value	unit	Parameter Type
Acrylamide	79-06-1	0,1	µg/l	Chemical
Antimony	7440-36-0	5	µg/l	Chemical
Arsenic	7440-38-2	10	µg/l	Chemical
Benzene	71-43-2	1	µg/l	Chemical
Benzo(a)pyrene	50-32-8	0,01	µg/l	Chemical
Boron	7440-42-8	0,5	mg/l	Chemical
Bromate	15541-45-4	1	µg/l	Chemical
Cadmium	7440-43-9	5	µg/l	Chemical
Chrome	7440-47-3	50	µg/l	Chemical
1,2-dichloroethane	107-06-2	3	µg/l	Chemical
Epichlorohydrin	106-89-8	0,1	µg/l	Chemical
Fluoride	16984-48-8	1	mg/l	Chemical
Copper	7440-50-8	2	mg/l	Chemical
Mercury	7439-97-6	1	µg/l	Chemical
Lead	7439-92-1	10	µg/l	Chemical
Nickel	7440-02-0	20	µg/l	Chemical
Nitrate	14797-55-8	50	mg/l	Chemical
Nitrite	14797-65-0	0,1	mg/l	Chemical
N-nitrosodimethylamine (NDMA)	62-75-9	12	ng/l	Chemical
2,4,4'-trichlorobiphenyl	7012-37-5	0,1	µg/l	Chemical
2,2 ', 5,5'-tetrachlorobiphenyl	35693-99-3	0,1	µg/l	Chemical
2,2 ', 4,5,5'-pentachlorobiphenyl	37680-73-2	0,1	µg/l	Chemical
2,3 ', 4,4', 5-pentachlorobiphenyl	31508-00-6	0,1	µg/l	Chemical
2,2 ', 3,4,4', 5'-hexachlorobiphenyl	35065-28-2	0,1	µg/l	Chemical
4,4', 5,5'-hexachlorobiphenyl	35065-27-1	0,1	µg/l	Chemical
2,3,4,5,2 ', 4', 5'-heptachlorobiphenyl	35065-29-3	0,1	µg/l	Chemical
aldrin	309-00-2	0,03	µg/l	Chemical
dieldrin	60-57-1	0,03	µg/l	Chemical
heptachlor	76-44-8	0,03	µg/l	Chemical
heptachlor epoxide	1024-57-3	0,03	µg/l	Chemical
Selenium	7782-49-2	10	µg/l	Chemical

Continuation Table 1

Parameter	CAS-number	Maximum value	unit	Parameter Type
Vinyl chloride	75-01-4	0,1	µg/l	Chemical
Ammonium	14798-03-9	0,2	mg/l	Indicator, Operational
Chloride	16887-00-6	150	mg/l	Indicator, Operational
Aluminium	7429-90-5	200	µg/l	Indicator, Organoleptic/ aesthetic
Iron	7439-89-6	200	µg/l	Indicator, Organoleptic/ aesthetic
Manganese	7439-96-5	50	µg/l	Indicator, Organoleptic/ aesthetic
Sodium	7440-23-5	150	mg/l	Indicator, Organoleptic/ aesthetic
Sulfate	14808-79-8	150	mg/l	Indicator, Organoleptic/ aesthetic
Zinc	7440-66-6	3	mg/l	Indicator, Organoleptic/ aesthetic
Diglyme(s)	111-96-6	1	µg/l	Indicator, Signalling
Ethyl tert-butyl ether (ETBE)	637-92-3	1	µg/l	Indicator, Signalling
Methyl tert-butyl ether (MTBE)	1634-04-4	1	µg/l	Indicator, Signalling
Pyrazole*	288-13-1	3	µg/l	Guiding value
Aromatic amines	Parameters from this group	1	µg/l	Indicator, Signalling
(Chloro) phenols	Parameters from this group	1	µg/l	Indicator, Signalling
Halogenated monocyclic hydrocarbons	Parameters from this group	1	µg/l	Indicator, Signalling
Halogenated aliphatic hydrocarbons	Parameters from this group	1	µg/l	Indicator, Signalling
Monocyclic hydrocarbons / aromatics	Parameters from this group	1	µg/l	Indicator, Signalling
Other anthropogenic substances	Parameters from this group	1	µg/l	Indicator, Signalling
Pesticides and humane relevant metabolites	Parameters from this group	0,1	µg/l	Chemical
Polycyclic aromatic hydrocarbons (PAHs)	Sum parameter	0,1	µg/l	Chemical
Cyanides (total)	Sum parameter	50	µg/l	Chemical
PCB's	Sum parameter	0,5	µg/l	Chemical
Pesticides	Sum parameter	0,5	µg/l	Chemical
Tetra- and trichloroethene	Sum parameter	10	µg/l	Chemical
Trihalomethanes	Sum parameter	25	µg/l	Chemical

** This guiding value for Pyrazole is from the 'Drinkwaterregeling' (https://wetten.overheid.nl/BWBR0030152/2017-10-27)*

As can be seen in Table 1 both organic and inorganic substances have a standard in the DWB (2018). Moreover the DWB (2018) values consist of chemical, organoleptic/aesthetic or signalling indicators. Not all parameters are equally essential to remove for human health. They are all important for acceptable drinking water.

The WQI RR is defined to depend on the maximal removal that is required to meet all (DWB) target values for parameters present in the water. More specifically, we calculate for each parameter the ratio of the maximum concentrations which are measured in a given period and the corresponding DWB values. Then, these ratios will be summated over the number *n* of substances. The summation over a number of substances gives an indication of how manifold the problem is. The WQI RR grows if the maximal measured concentration for any parameter exceeds the DWB values. There is no upper boundary. A high index value would thus indicate that there is a high removal requirement. For parameters for which the maximal measured concentration C_{MAX} exceeds the DWB target value C_{DWB} (so $C_{MAX} > C_{DWB}$), Equation 1 calculates the Removal Requirement (RR) for a single parameter and Equation 2 calculates the total WQI RR.

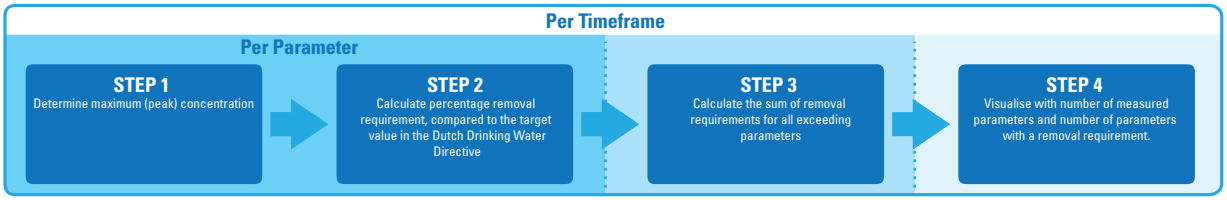
$$RR_i = 100 * \left(1 - \frac{C_{DWB,i}}{C_{MAX,i}}\right) \qquad \text{(Equation 1)}$$

$$WQI\_RR = \sum_{1}^{n} RR_i \qquad \text{(Equation 2)}$$

Here, C_{DWB} is the DWB value (see Table 1) for parameter i, C_{MAX} is the peak concentration in a period for parameter i, and n is the total of parameters. RR is the removal requirement for a single parameter i. Of course, the fewer times a parameter is measured within a period, the more chance there is that C_{MAX} is an underestimation because it cannot be excluded that at some other time the concentration is higher.

In Figure 1 this procedure is shown. The calculation of the WQI RR implies that the more exceedances and the bigger the individual exceedances, the higher the index will be. Every substance that exceeds its target value will maximally add a hundred 'points' to the index. The higher the WQI RR, the more removal is required to meet drinking water regulations in the DWB. The WQI RR can be used to assess water quality in terms of removal requirement, which - by definition - is independent of the type of treatment.

Figure 1. A schematic overview of the calculations of the WQI RR per location.



2.2 Calculation of the WQI PTE

To obtain an indication of how much 'effort' is needed to actually remove contaminants in raw water we formulate a water quality index for purification treatment effort (WQI PTE), in addition to the described WQI RR. For this purpose, we assign a weight factor to each substance that has to be removed, where its value will depend on its estimated difficulty of removal.

The estimated difficulty of removal in this index is based on two substance traits. For a discussion on the used traits, see Appendix 1. The first trait that is chosen to represent removal effort is the (logarithm of) the octanol-water partition coefficient K_{ow}, a measure of hydrophobicity. Log K_{ow} is an indicator for the tendency of a substance to partition between an organic phase (e.g. a soil or adsorbent) and the aqueous phase. The more hydrophobic a substance is (high log K_{ow}), the easier it is to remove from water.

The second substance trait is a biodegradation constant. Biodegradability represents the tendency for substances to be broken down to its constituents. The higher the biodegradability of a substance, the easier the removal by biological processes.

Although many more parameters are involved in removal processes and their efficiency (van Leeuwen et al., 2007; van Wezel et al., 2017; de Munk et al., 2018; Fischer et al., 2019) it was decided to only use log K_{ow} and biodegradation for the removal effort in this report as both adsorption and biodegradation processes occur during drinking water treatment and these will play a role in 'simple' purification treatment. We do not explicitly define purification steps in any particular treatment, as this will differ per treatment installation (Stackelberg et al., 2007; Fischer et al., 2019).

To obtain the traits to an indication for purification treatment effort the values are predicted by the model suite in EPIsuite (US EPA, 2019). For log K_{ow} we use the model Kowwin (Meylan and Howard, 2005). Whenever there was an experimentally obtained value available for log K_{ow} in EPIsuite, this was preferred over the calculated value. For biodegradation we use Biowin3 (Boethling et al., 2004). Biowin3 predicts an order of speed of biodegradation: a value of ›4.75 - 5 is hours, ›4.25 - 4.75 is hours to days, ›3.75 - 4.25 is days, ›3.25 - 3.75 is days to weeks, ›2.75 - 3.25 is weeks, ›2.25 - 2.75 is weeks to months, ›1.75 - 2.25 is months, ‹1.75 is persistent (Boethling et al., 2004).

All possible values of the two traits log K_{ow} and biodegradation are stored in four bins with a weight factor that is scaled per trait (*w1* and *w2*), see Table 2. This approach is based on the approach in Fischer et al. (2011). Biodegradation weights are set on only half of that of log K_{ow}. Biodegradation is an important process for water treatment, and especially in waste water treatment plants (WWTPs). However, in WWTPs treatment is by another type of process than those used in a drinking water treatment process. The water quality (presence of nutrients) is different, and besides the residence time of water in the WWTP is significantly longer (typical 1-3 days) than the contact time in a sand or activated carbon filter (± 40 minutes). Therefore, if not downscaled, the application of biodegradability as measure may overestimate the contribution of biodegradation in drinking water treatment plants.

Table 2. The trait values (t1 and t2) for log K_{ow} and Biodegradation are binned to indicate the ease of removal of substances (see Fischer et al. 2011 for a similar approach) with weight values (w1 and w2). A weight value of 1 indicates no removal. A weight value of 0 indicates full removal.

	Normalised bin w1	0	0.33	0.66	1
EPIsuite model Kowwin	Log K_{ow} (t1)	$t1 > 6$	$3 < t1 \leq 6$	$0 < t1 \leq 3$	$t1 < 0$
	Normalised bin w2	**0.5**	**0.67**	**0.83**	**1**
EPIsuite model Biowin3	Biodegradation (t2)	$t2 > 4.75$	$3.25 < t2 \leq 4.75$	$3.25 < t2 \leq 2.25$	$t2 < 2.25$

These scaled weighting factors *w1* and *w2* are used to obtain an indication of the effort to remove the substance in question. The removal is calculated as an additive process (see Equation 3) in which there is an indication w1 for removal from the first trait log K_{ow}. What remains of the substance has an indication w2 for removal from the second trait biodegradability. As a drinking water treatment consists of several subsequent processes, each of which resulting in a certain purification, calculating the purification indication by this method is considered more realistic than a simple averaging of the indications for removal. This indication is referred to as 'Removal Indication' (RI) (see Equation 3).

$$RI_i = w1_i \bullet w2_i \cdot 100 \qquad \text{(Equation 3)}$$

Here RI is the removal indication for parameter *i*, *w1* and *w2* are the weights for a substance for the traits from Table 2. An RI value of zero means that it is expected that the substance will be fully removed. An RI value of one hundred means that the removal of the substance is expected to be negligible. The actual removal in percentages can be calculated for a substance as *100-RI*. Appendix 1 holds these predicted removal percentages, with an expert judgement on how accurate these are predicted by the RI calculation in Equation 3. In 66% of substances, the established RI (Table 2, Equation 3) has a realistic removal by expert judgement for a 'simple' treatment with sorption and biodegradation processes (see Appendix 1).

The WQI PTE is calculated as the average of the removal indication RI (see Equation 3) for all substances *i* that exceed their target value in the DWB (2018) (see Equation 4).

$$WQI\_PTE = \frac{\sum_{1}^{n} RI_i}{n} \qquad \text{(Equation 4)}$$

Here WQI PTE is the water quality index for purification treatment effort over all substances *n* that exceed the target value in the DWB (2018).

The WQI PTE yields a value between zero and a hundred. A WQI PTE of zero means that all substances in the water source can be completely removed. Conversely, a WQI PTE of a hundred means that none of the substances of the water source can be removed.

Note that EPIsuite is not able to calculate the trait constants *t1* and *t2* (see Table 2) for inorganic substances, since these constants fall outside its modelling domain. For the inorganic parameters, the RI was set at a low value to ensure that the removal effort index value corresponds to an easy to remove indicator value (17) as for most of these compounds that this is the case in reality. Exceptions are Chloride, Fluoride, Sulfate, Nitrite, Ammonium, and Nitrate, these are known to be hard to remove. These parameters receive a high RI to indicate their difficulty in removal (83). The RI for such parameters have to be added by hand.

2.3 Data processing

To test our indices in a real-world setting, we used measurement data from an existing database, the RIWA-base (RIWA, 2012). This database contains measurements from existing monitoring programs on substances ('parameters') which are listed in the DWB (2018) and gathers data for locations along the river Rhine in the Netherlands. The data originate from as early as 1972, and vary between locations and years in the amount of and type of parameters measured. We consider five locations for assessing the removal requirement and purification treatment effort indices. The German-Dutch border at Lobith, the Lekkanaal at Nieuwegein, the Amsterdam-Rhine canal at Nieuwersluis, the IJsselmeer at Andijk and the Haringvliet at Stellendam and Middelharnis (Figure 2). The latter four locations are drinking water abstraction points.

The target values for substances in the water in the DWB (2018) are not explicit for all substances. Aside from substance specific target values there are target values for substances that fall within certain groups, and sum parameters for which the combination of substances from a certain group should not exceed. All types are listed in Table 1.

For instance, there is a specific target value for the pesticide Aldrin, which is 0.03 µg/l. For pesticides without a specific target value, the target value for parameters in that group is 0.1 µg/l (group parameter). And, the pesticides altogether should not exceed 0.5 µg/l (sum parameter).

To set the correct target values for parameters in RIWA-base, we used the substance group labels (e.g. 'Pesticides', 'Other anthropogenic') from another Dutch database that collects monitoring data on drinking water related parameters (REWAB). Per compound group, all substances with the compound group label receive that target value, except for substances that already have their own specific target value. For small compound groups that were described in the DWB (2018) itself, such as is the case with PCB's, we used the list in the DWB (2018) to set the target value for the substances in the RIWA-base.

Figure 2.

For sum parameters, the target value is described in the DWB (2018) as: 'Sum of separate – group name – with concentration higher than the detection limit.' All maximum (peak) values within a period above the detection limit with the group name label from REWAB, or described in the DWB (2018), were summed. This was taken as the measurement value of the sum parameter for that period. It should be noted that this method is prone to overestimation, as in reality not all parameters will exceed maximally at the same time. RIWA-Rijn receives several measurement series that are generated by different labs and analytical methods. Per parameter only one time series (the most sensitive and complete) is selected according to RIWA selection criteria (30 jaar RIWA-base, 2012). All measurements under the detection limit received a value of zero. Biological parameters in the DWB (2018) were left out of the calculations. We set the period in which a substance can exceed the target value from the DWB (2018) to one year. We consider the years 2000, which is the start of the WFD, until 2018. We linked the substances in the DWB (2018) (see Table 1) to those in the RIWA-base via their CAS number. To do this, the CAS numbers were added to the substances in the DWB (2018). The reported data from RIWA-Rijn were recalculated for every parameter to a (peak) maximum value per period. If no reported measurements were available for a given period, NA was generated. For every value that was above the target value of the DWB (2018) (Table 1) the percentage to be removed to get the parameter back to the target value was calculated (see Equation 1). All measured maximum values in the period below the target value (Table 1) were omitted as these do not contribute to the removal requirement.

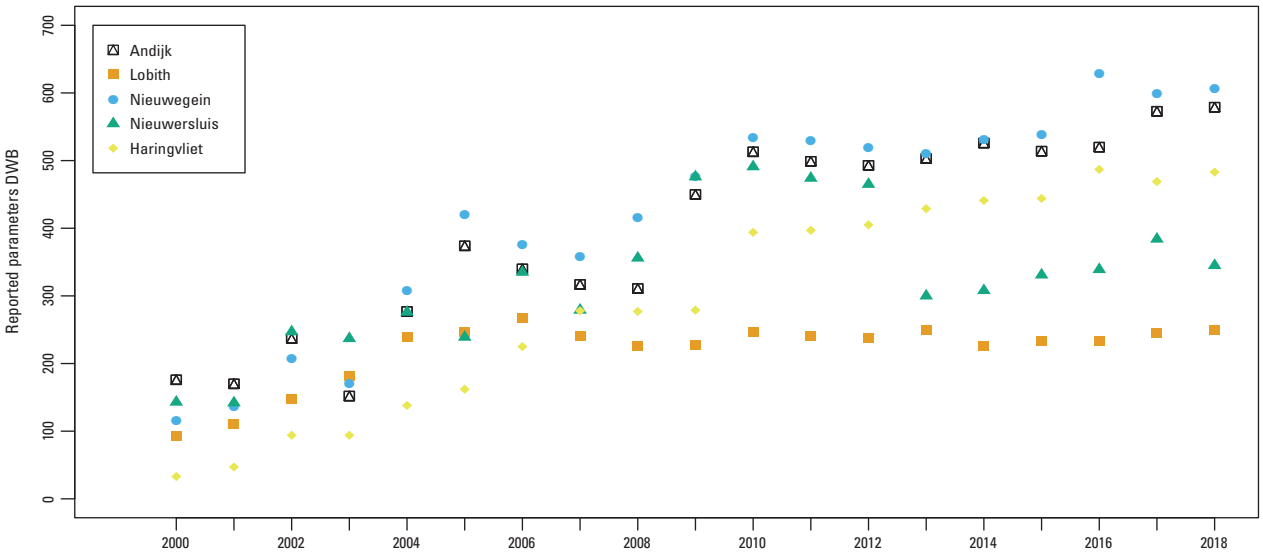
All data processing and visualisations were done in the coding language R. This script, with the accompanying data sources, is available for interested parties for verification of results in this report.

3 Results

3.1 Data characterization

To gain insight into how much was measured in these years at the different Rhine locations and how many of these measured parameters exceed the DWB (2018) value (Table 1), we first visualise these measurement values. Figure 3 shows the number of measured parameters present with a value in the DWB (2018) at the five locations. Figure 4 shows the number of parameters per location that exceed the DWB value. In Figure 3 it can be seen that more parameters are measured in later years than in earlier years (two to sixteen times as many, depending on the location). The number of exceeded substances remains much more constant (Figure 4), and shows only a slight increase (from equal to four times as much, depending on location).

Figure 3. The number of reported parameters per year per location with a value in the DWB (see Table 1)



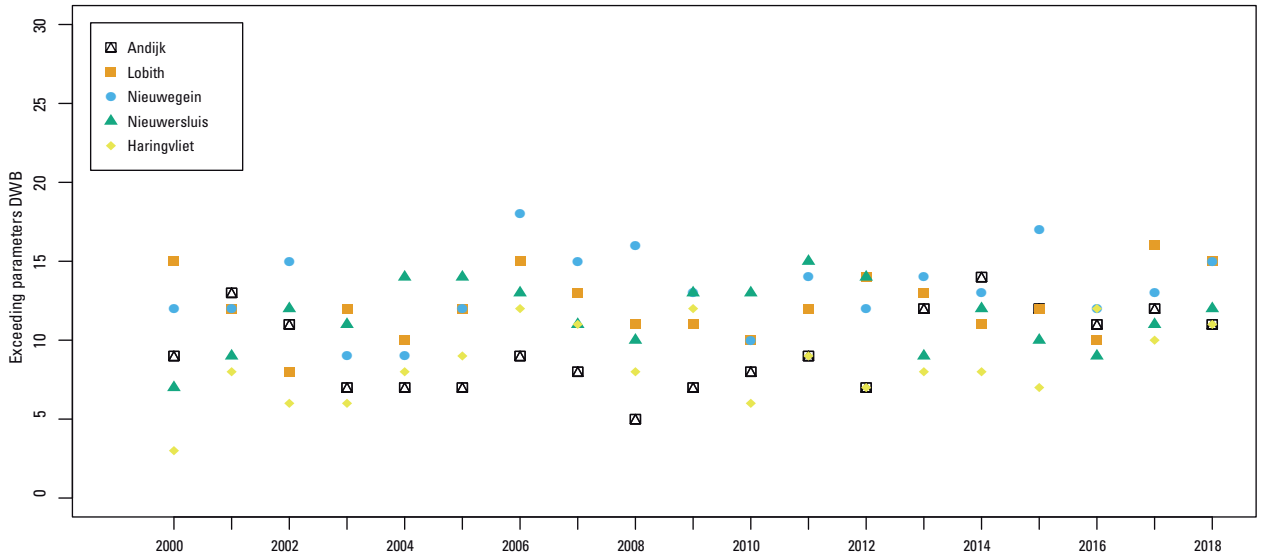
The removal requirement calculation deliberately does not limit itself to substances that have already been measured historically, because this would not capture the problem of emerging substances and changes over time in the use of substances. The monitoring program is constantly adjusted according to the latest insights obtained from scientific literature, reports, or environmental monitoring such as non-target screenings.

In this way the program is always focused on measuring all potentially exceeding substances. New parameters are added with advancing insight or as a precaution. Nowadays, other substances exceed their target value compared to the past. After all, there is a trend of emerging, new substances constantly coming onto the market and also ending up in surface water (van Wezel et al., 2017). For instance, due to legislation, use of certain substances may be restricted and an alternative substance may then be used. It is inherent to the pollution and the associated measures that the problem changes over time.

The vast majority of the parameters that have been newly measured since 2001 at all locations (Figure 3) combined do not immediately exceed upon introduction, so presuming an increasing trend in time, no exceedances in the years before the analysis started have been missed. However, a total of 26 parameters immediately exceeded the target values during the first measurement (between 2001 and 2018), sometimes at multiple locations.

These parameters may have already been exceeding target values before measurements were executed, resulting in a potential underestimation of the removal requirement in the years before the analysis started. An estimation on the likelihood of emission routes existing for these parameters could shed more light on the potential of underestimation, however this is beyond the scope of this report.

Figure 4. The number of parameters per year per location that exceed the DWB value.

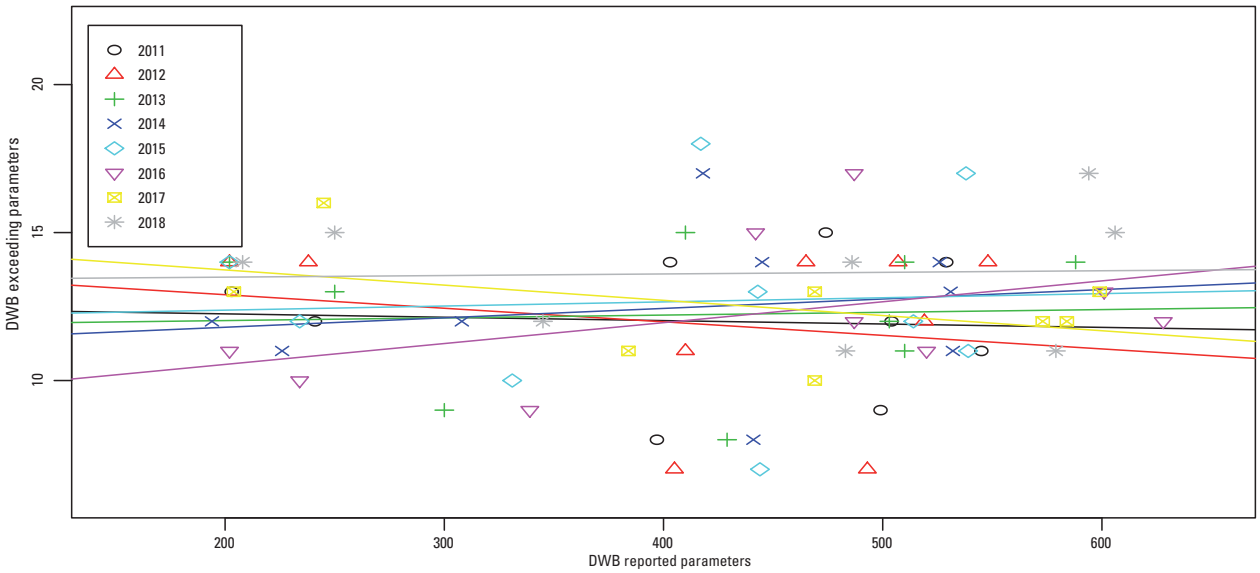


In conclusion, although we cannot exclude that some substances exceed their target value because they are newly measured, it is likely that most measurements are targeted at catching new, emerging substances. Moreover, the hypothesis that more parameters may exceed their target value purely because more are being measured does not hold true when we compare these numbers between locations. We looked at the relationship between measured parameters and parameters that exceed their target value at the locations, within the same year (Figure 5).

We included the measurement data at the three locations in the river Meuse, to be able to base this conclusion more robustly on more measurement points. The relationship between the number of measured parameters and the number of parameters exceeding their target value at the locations is positive in some years and negative in others. None of the relationships is statistically significant. Based on this analysis it cannot be concluded that the more parameters are measured at a location, the more parameters will exceed their target values. This implies that there is no obvious relationship between the number of exceedances and the size of the monitoring program.

We therefore include all parameters in the calculation of the WQI indices. This means all parameters potentially contribute to this purification treatment requirement, also those only recently reported. These new substances also contribute to potential risks and therefore to the need to remove them.

Figure 5. The relationship (lines) per year between reported parameters (x-axis) and parameters that exceed the target values (y-axis) for eight measurement locations along the Rhine and Meuse.

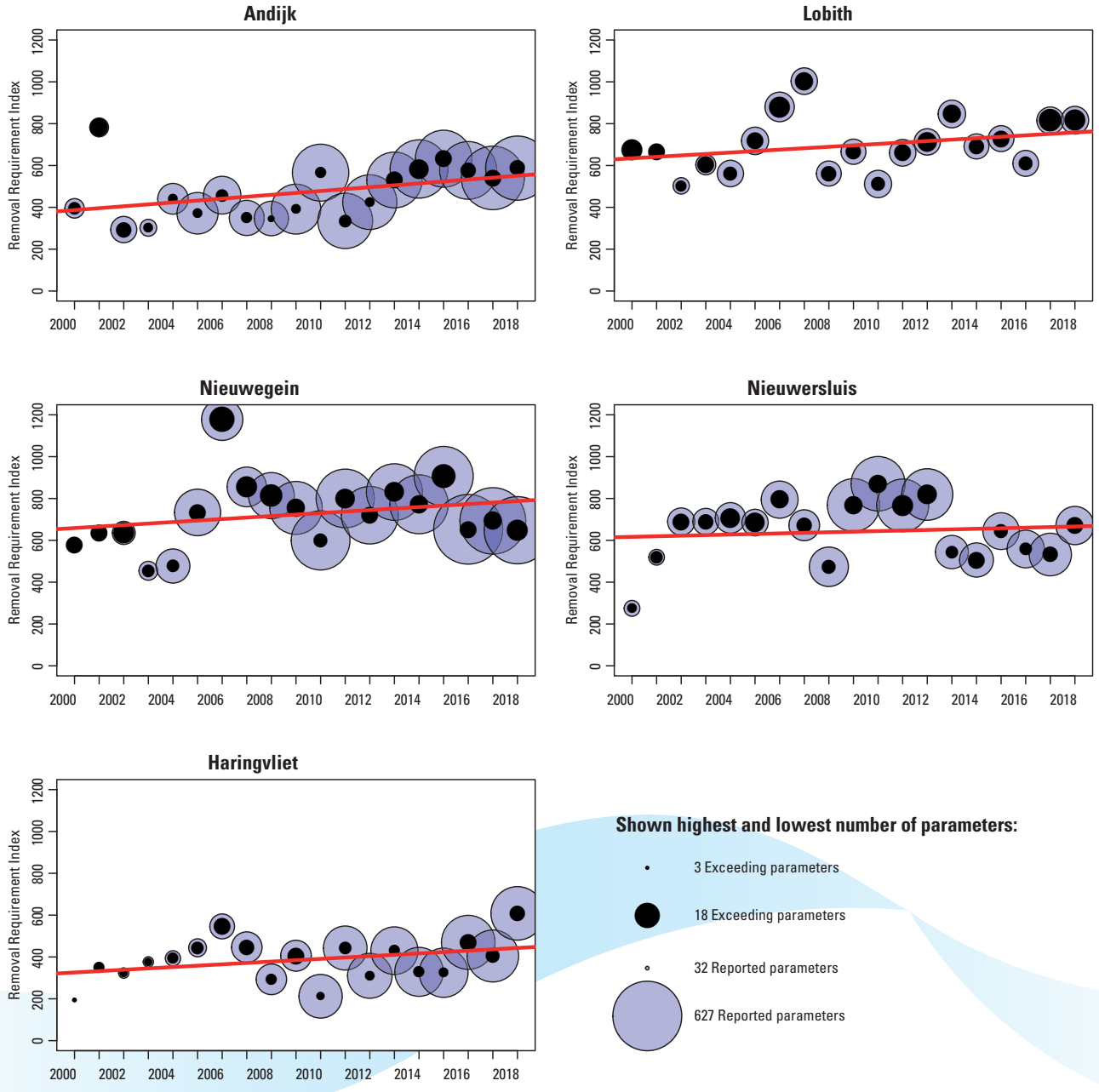


Because a different estimate is made of the (potential) problematic substances for different locations, and therefore the precise measurement packages can vary in what parameters they measure, it is good to view the WQI RR and WQI PTE per location and not for the river Rhine as a whole. Differences in these indices between locations can be evaluated. These results are discussed in the next paragraphs.

3.2 Values and trends in the WQI RR

In Figure 6 the WQI RR for source water at the five locations along the river Rhine in the period 2000 to 2018 is shown. The blue spheres indicate the number of measured substances that are in the DWB (2018) in that year (see also Figure 3). The black cores in it indicate the number of substances exceeding DWB values in that year (see also Figure 4). The height of the blue spheres with black core is the height of the removal requirement. This value is the sum of all removal requirements (RR) for individual substances that exceed the DWB value in that year (see Equation 1).

Figure 6. The calculated WQI RR per year at five measurement locations in the Rhine river, from 2000 to 2018. The solid (red) line is a linear regression through the values of the WQI RR.



In 2018, the order of locations based on their WQI RR from low (better), to high (worse):

Andijk (587) ‹ Haringvliet (608) ‹ Nieuwegein (648) ‹ Nieuwersluis (670) ‹ Lobith (816)

Location Andijk has the lowest WQI RR in 2018, and therefore has the best water quality with regard to this index in 2018. In Table 3 we show the significance values (p-values) of the trends in removal requirement at the locations. Here we show whether the WQI RR is decreasing (green) or increasing (red) and the statistical significance (p-value). Table 3 shows that the WQI RR does not significantly ($p < 0.05$) improve or deteriorate in any of the locations. For Andijk, Nieuwegein, Haringvliet and Lobith there is a trend towards an increase in the WQI RR, which implies a deteriorating trend.

Table 3. Trend in WQI RR between 2000-2018. A green box means that WQI RR shows a downward trend. This means that the quality improved. A red box indicates a deterioration, so the index is increasing. Numbers indicate the significance value (p-value) of the trend. Not coloured are trends with significance level p › 0.5.

	Andijk	Lobith	Nieuwegein	Nieuwersluis	Haringvliet
Trend in WQI RR 2000 -2018	0.10	0.22	0.31	0.66	0.13

Outliers

In Figure 6, a few upward outliers can be seen in the WQI RR. In some cases this is due to a few parameters. We identify parameters within outlier-years with the following calculation. If a parameter in a year exceeds the average of that parameter over 2000-2018 in a year, plus twice the standard deviation 2000-2018 of the parameter, then we see this parameter as noticeably higher than normal. For this calculation we set the RR in years in which a substance does not exceed to zero. This does not take into account whether the substance was actually measured in each year. By taking the average plus twice the standard deviation, we get a picture of the possible incidents that cause the outliers.

Andijk: A high value in 2001 is due to the parameters Dodecane, Decane, Hexadecane, Octadecane, Tetradecane. This might be explained by a diesel spill. The high value in 2010 in Andijk is due to specifically Dibutyl phthalate (DBPH), Aldicarb sulfoxide, and Nitrite (NO_2), of which we do not know the cause.

Lobith shows an outlier in 2007 due to Polycyclic aromatic hydrocarbons (PAHs), Benzo(a)pyrene, Nitrite (NO_2) and Bis(2-ethylhexyl)phthalate (DEHP). In 2013 there is again a high value, not due to some substances that suddenly exceed the DWB value in that year, but rather to increasing concentrations starting in 2011, such as by Hexa(methoxymethyl) melamine (HMMM). This latter is an example of a substance that adds to the index because it is newly measured, at location Lobith this parameter was added to the measurement program in 2011 (in 2007 it was already exceeding at locations Andijk and Nieuwegein).

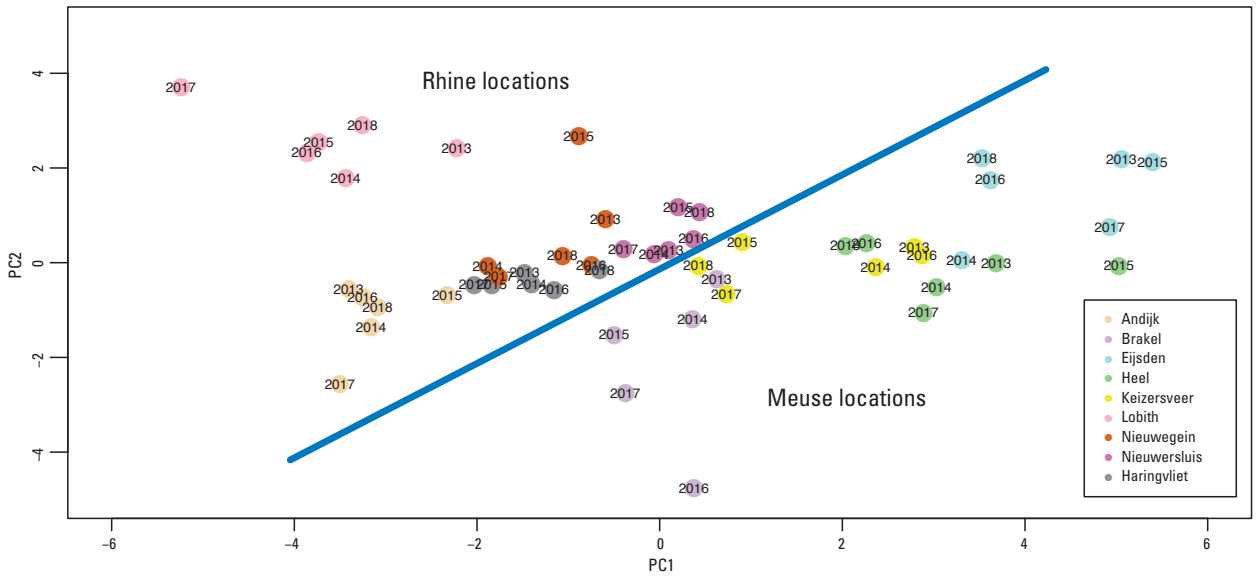
Nieuwegein: The high value of 2006 is due to Dodecane, Decane, Hexadecane, Tetradecane and Diglyme (also exceeding a few years after 2006). This might be related to a diesel spill. Moreover, there was a low water level in the Lekkanaal at the start of 2006 (36 m^3/s) which results in lower dilution of all chemicals entering the water system.

Nieuwersluis shows no clear outlier. The increase in the WQI RR between 2009-2012 is a combination of substance concentrations that are higher in that year, but also incidentally reach that level in other years. These exceedances are mainly pesticides.

Haringvliet shows a peak in 2006 which is caused, among other things, by Polycyclic aromatic hydrocarbons (PAHs). The increase in 2018 is partly due to Trifluoroacetic acid (TFA), Melamine and Guanylureum.

All locations considered have their own (although with large overlap) measurement program. It is interesting to see whether the exceeding parameters that contribute to the WQI RR are the same in all locations. Figure 7 shows the similarity in the locations for different years with respect to the WQI RR. To include more data and see if patterns are specific for the river Rhine, we compared results with data from another river that enters the Netherlands, the river Meuse. For all locations, the similarity within locations is bigger than the similarity within years for the different locations. This means that for most sites, there is a distinctive set of parameters that contributes to the WQI RR. The years of different locations, on the other hand, do not cluster together. In Figure 7, we note that there is a distinction possible between river Meuse (lower right dots) and river Rhine (higher left dots). This indicates that both rivers have partly different parameters that contribute to the WQI RR. No further analyses on river Meuse data is included in this report.

Figure 7. Similarity of the removal requirement between locations and years, visualised by Principal Component analysis. The blue line is hand-fitted and indicates a division between Meuse and Rhine locations.



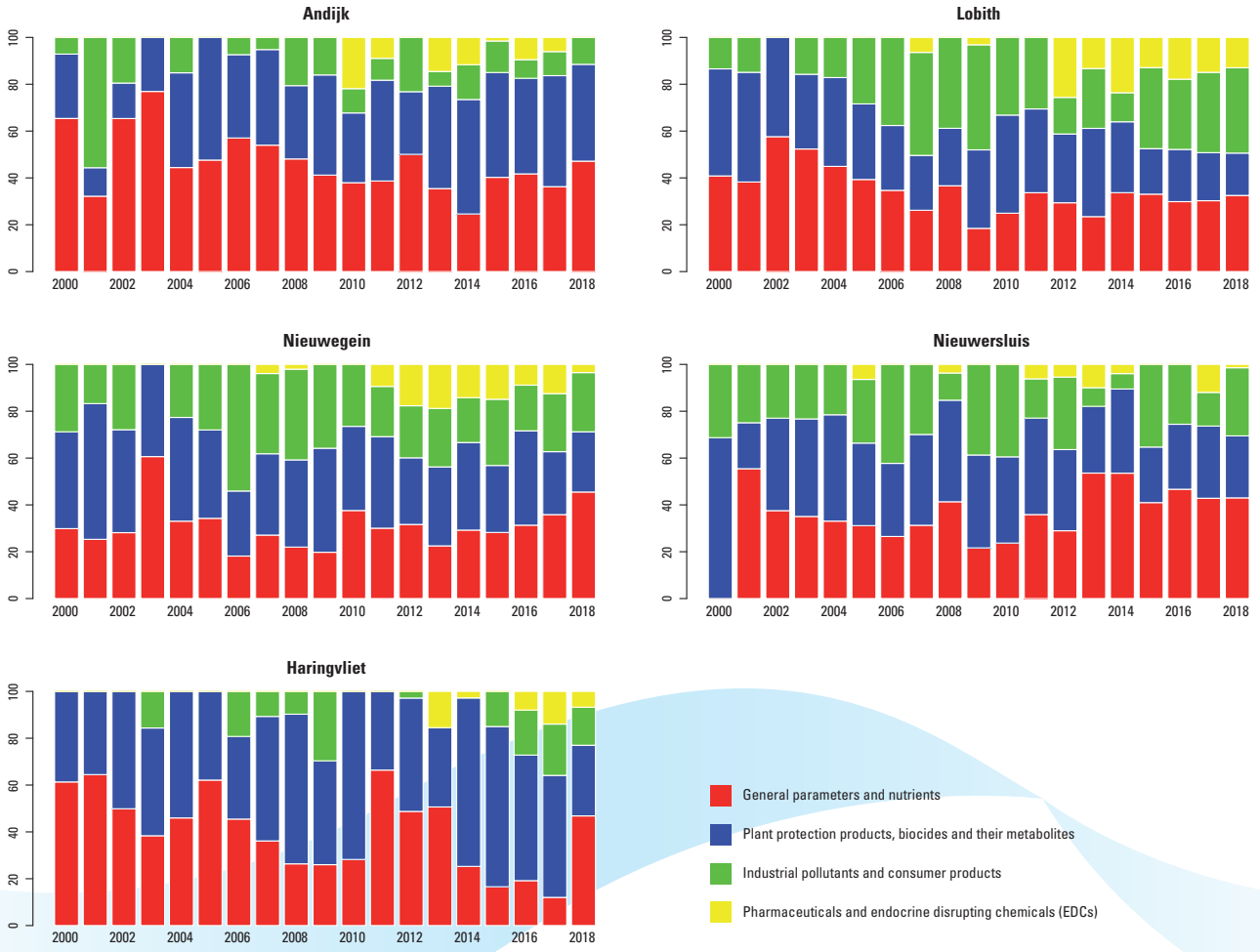
3.3 Contribution of substance categories to the WQI RR

To get an idea what sort of substances contribute to the WQI RR we look at what kind of parameters exceed target values per year per location. We break down all exceeding parameters into four substance categories to clarify the contribution of the different categories.

1. General parameters and nutrients
2. Plant protection products, biocides and their metabolites
3. Industrial pollutants and consumer products
4. Pharmaceuticals and endocrine disrupting chemicals (EDCs)

We calculate the percentage contributions of these categories per location and per year to the WQI RR. These categories are derived from the labels used in the RIWA-base. Because some parameters have more than one label, some parameters are counted in more than one parameter category.

Figure 8. Contributions of parameter categories to the removal requirement (Figure 3). For Nieuwersluis 2000, no parameters were measured in the "General parameters and Nutrients" parameter category.



It is possible that in the future some parameters get another parameter category label. For example, metabolites of pesticides are currently classified as 'Plant protection products, biocides and their metabolites' and no distinction is made between human relevance (with target value 0.1 µg/l) or not (with target value 1.0 µg/l), because for the most part this has not been sorted out in a structured way. As a precaution, these have all been tested against the lower target value. The group "Other anthropogenic substances" from the DWB, which is very broad, is also subject to change and may be adjusted or expanded in the future. For now, we rely on the current parameter group and category labels in the RIWA-base.

Figure 8 shows that parameters that exceed their target value with the labels "General parameters and nutrients" and "Plant protection products, biocides and their metabolites" make a major contribution to the total WQI RR at all locations. At Lobith, Nieuwegein and Nieuwersluis, "Industrial pollutants and consumer products" also make a major contribution, but in recent years their contribution seems to be lower. The "Pharmaceuticals and endocrine disrupting chemicals (EDCs)" only make a small contribution to the WQI RR at all locations, and this contribution is mainly in the most recent years.

Because the percentage contributions in Figure 8 do not show whether parameter categories also increase in an absolute sense, Table 3.3 shows the significance values (p-values) of the trends in removal requirement in the parameter groups over the years 2000-2018, per location.

*Table 4. Trends of removal requirement per parameter category of 2000-2018. Green is a decrease, red is an increase in the parameter category at a location. Not coloured are trends with p > 0.5. Significant trends (p < 0.1) are indicated in bold with an *. 'Gew/Bioc/Metab' stands for category 'Plant protection products, biocides and their metabolites'. 'Ind/Cons' stands for category 'Industrial pollutants and consumer products'. 'GenM/EDC' stands for category 'Pharmaceuticals and endocrine disrupting chemicals (EDCs)'. 'Alg/Nutr' stands for category 'General parameters and nutrients'.*

	Andijk	Lobith	Nieuwegein	Nieuwersluis	Haringvliet
Gew/Bioc/Metab	**<0.01***	**0.03***	0.45	0.41	**0.03***
Ind/Cons	0.48	0.100	0.902	0.951	0.100
GenM/EDC	0.08	**<0.01***	**<0.01***	0.09	**<0.01***
Alg/Nutr	0.91	**0.05***	**0.01***	0.065	0.41

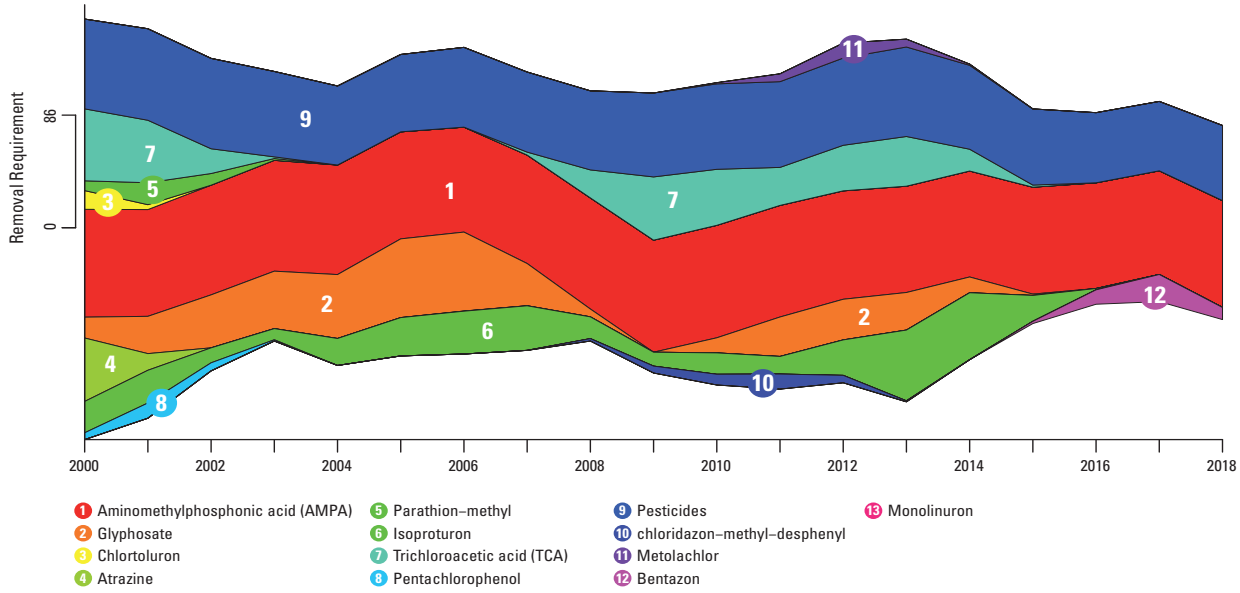
The removal requirement for "Plant protection products, biocides and their metabolites" is increasing significantly at Andijk and Haringvliet. At Lobith, on the other hand, there is a significant decreasing trend from 2000-2018. This means the contribution to this category from the Rhine that comes from abroad has declined. The removal requirement for "Industrial pollutants and consumer products" does not change significantly at the locations. The exceedances are very variable from year to year and there is no clear trend. "Pharmaceuticals and endocrine disrupting chemicals (EDCs)" exceedances are increasing at all locations. These increases are significant at Lobith, Nieuwegein and Haringvliet and near-significant at the others. Removal requirement for "General parameters and nutrients" increases significantly at location Nieuwegein and decreases significantly at location Lobith.

We can deduct from Table 4 that the increase in the WQI RR (see Figure 6 and Table 3) at Andijk and Haringvliet can mainly be attributed to an increase in "Plant protection products, biocides and their metabolites" and "Pharmaceuticals and endocrine disrupting chemicals (EDCs)". For Lobith, this is attributable to "Pharmaceuticals and endocrine disrupting chemicals (EDCs)". For Nieuwegein it is attributable to "Pharmaceuticals and endocrine disrupting chemicals (EDCs)" and "General parameters and nutrients".

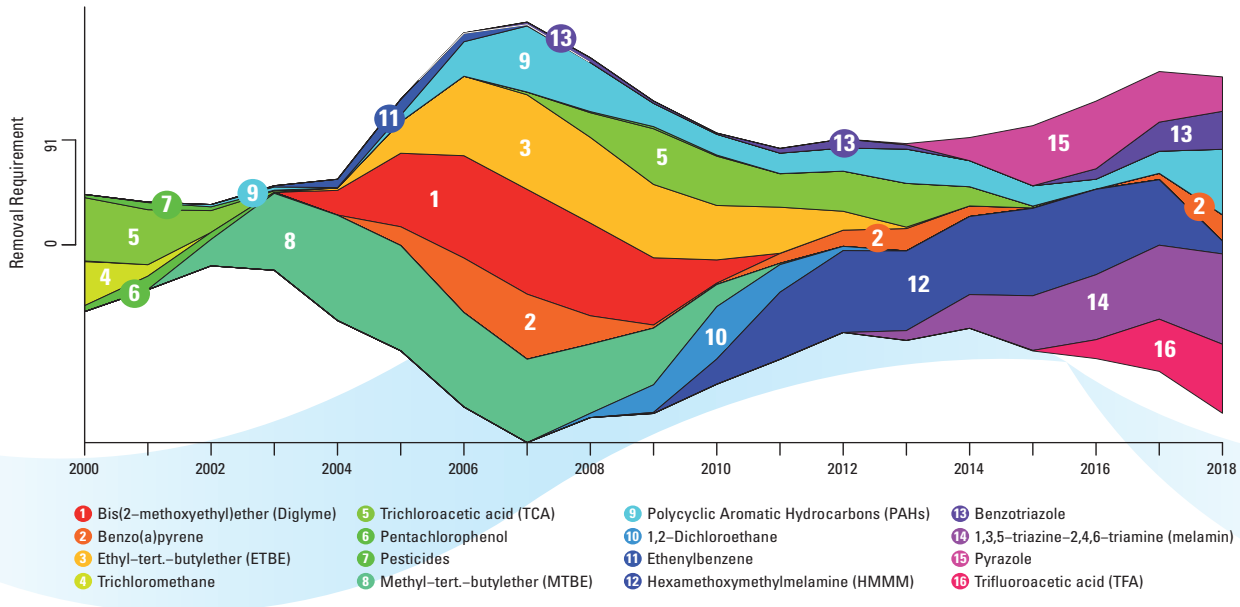
Lobith is of strategic importance because this is the location where the Rhine enters the Netherlands. For this location, we plot individual exceedances of the target value per parameter (to be seen as coloured “ribbons”), per parameter category over the measurement years in Figure 9.

Figure 9. The WQI RR in removal requirement for individual parameters (shown as coloured “ribbons”) per parameter category (in the individual plots) for Lobith. Because of the use of a “smoothing factor” when drawing these figures, the peaks per parameter are slightly spread out over the adjacent years.

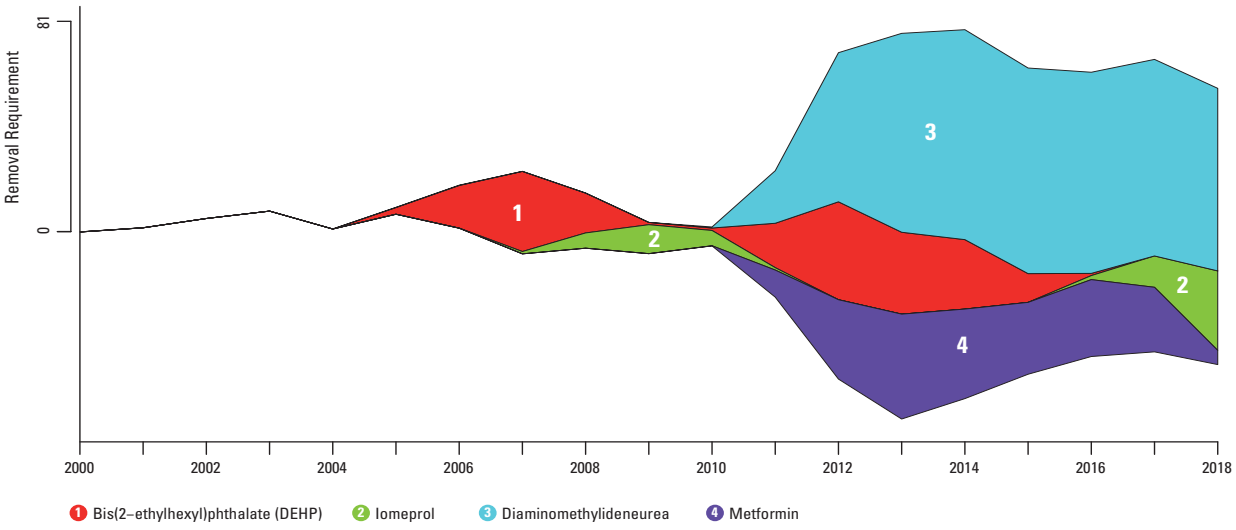
Plant protection products, biocides and their metabolites Lobith



Industrial pollutants and consumer products Lobith



Pharmaceuticals and endocrine disrupting chemicals (EDCs) Lobith



General parameters and nutrients Lobith

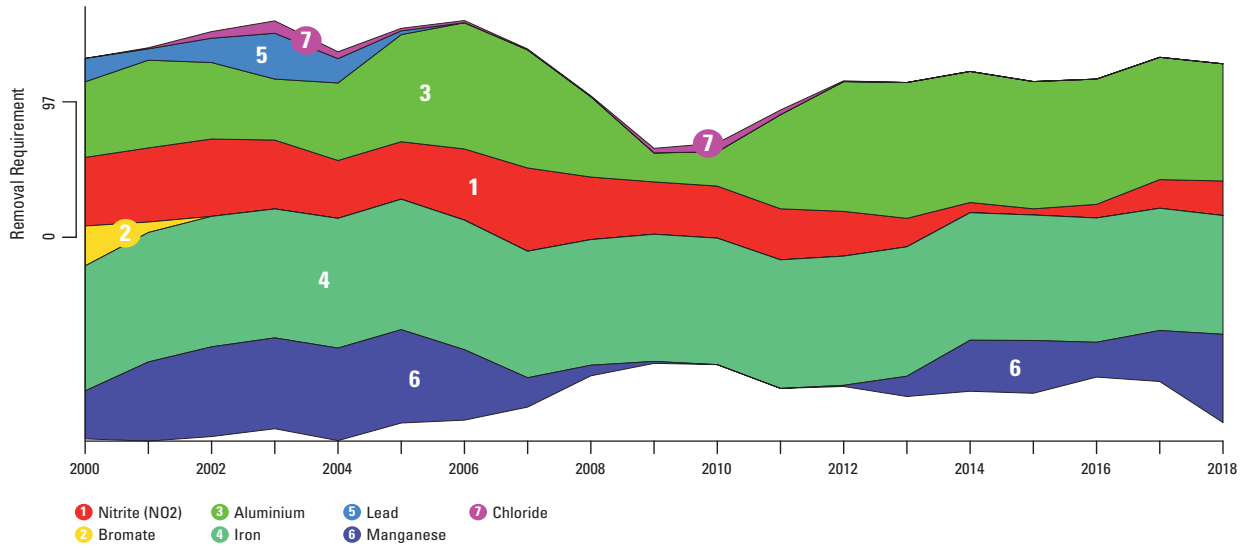


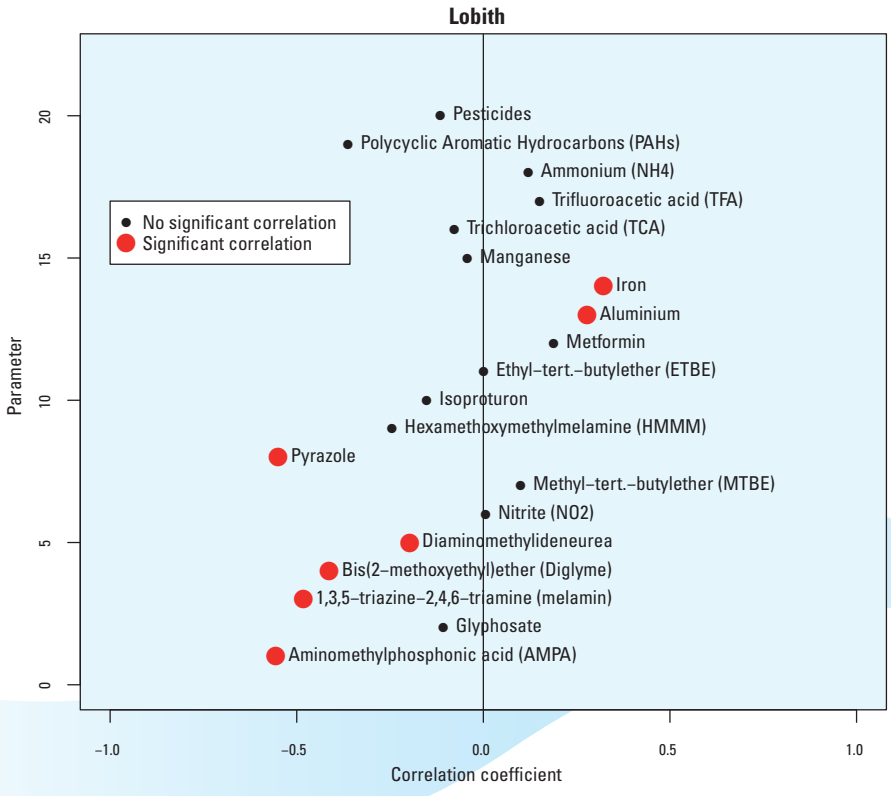
Figure 9 shows that at Lobith, parameters in "General parameters and nutrients" often structurally exceed the DWB value. This includes Iron, Nitrite as NO_2, Aluminium, and Manganese. "Pharmaceuticals and endocrine disrupting chemicals (EDCs)" exceeded from year 2007 onwards, including Iomeprol, Bis(2–ethylhexyl)phthalate (DEHP), Guanylureum, and Metformin. The "Industrial contaminants and consumer products" alternate, and emerge and disappear again. Only the sum parameter PAHs exceeds its target value during a longer period of time. "Plant protection products, biocides and their metabolites" generally exceed for longer periods, occasionally remaining below their target value (Table 1) such as Isoproturon, Glyphosate, Trichloroacetic acid (TCA). Aminomethylphosphonic acid (AMPA) and the pesticide group structurally exceed target values (around 80% and 60-70% respectively).

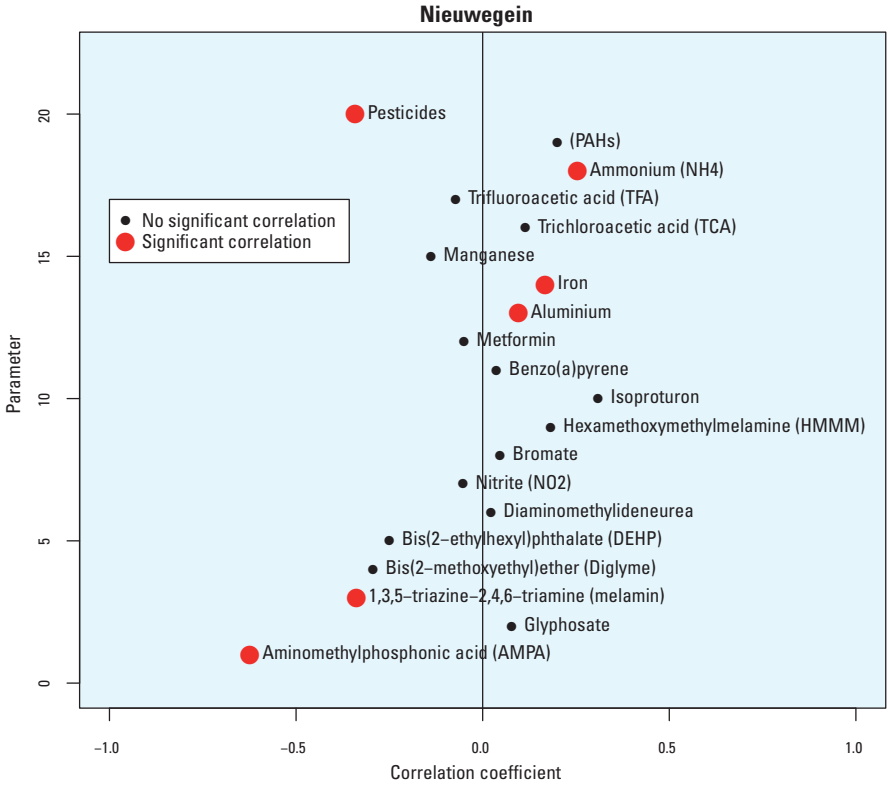
3.4 The influence of drought on the WQI RR

The discharge of water from rivers can influence the removal requirement for parameters. With the expected increase in occurence of extreme drought periods, measured concentrations of parameters in the Rhine water may increase in the coming years, since substances may be less diluted during low water periods. This will make that the removal requirement will increase. On the other hand, during high water periods industries may see an opportunity to discharge more waste water that will stay within legal limits. During the same high-water periods, some other substances may end up in elevated concentrations through the increased run-off from the land with precipitation or reduced removal in wastewater treatment plants due to sewage overflow and suboptimal hydraulic retention times in sewage treatment plants (Brunsch et al, 2018). The removal requirement may increase consequently, but now in a high discharge situation. For that reason, the WQI RR is not expected to be structurally dependent on the water discharge. The dependency rather varies with specific substances that contribute to the WQI RR. This is confirmed in Figure 10. Figure 10 shows the correlations of individual parameters exceeding their target values from the DWB (2018) with water discharge per month. Water discharge data is not available for every location, therefore we can only show this figure for Lobith and Nieuwegein.

The WQI RR as a whole has no significant relationship with water discharge (not shown), this is partly because the individual parameters relate differently to the discharge level. Moreover, water discharge is related to seasons, just like some (but not all) parameters. For example, pesticides are mainly used in the spring and summer, whereas, at the end of the summer the water levels are usually the lowest. Medicines will rather be used throughout the year, however some will also be bound to a specific season. The direct relation between discharge and specific parameters, omitting confounding effects, needs some thorough investigation that lies beyond the scope of this report.

Figure 10. Spearman correlations of removal requirement of parameters with water discharge, both calculated per month.





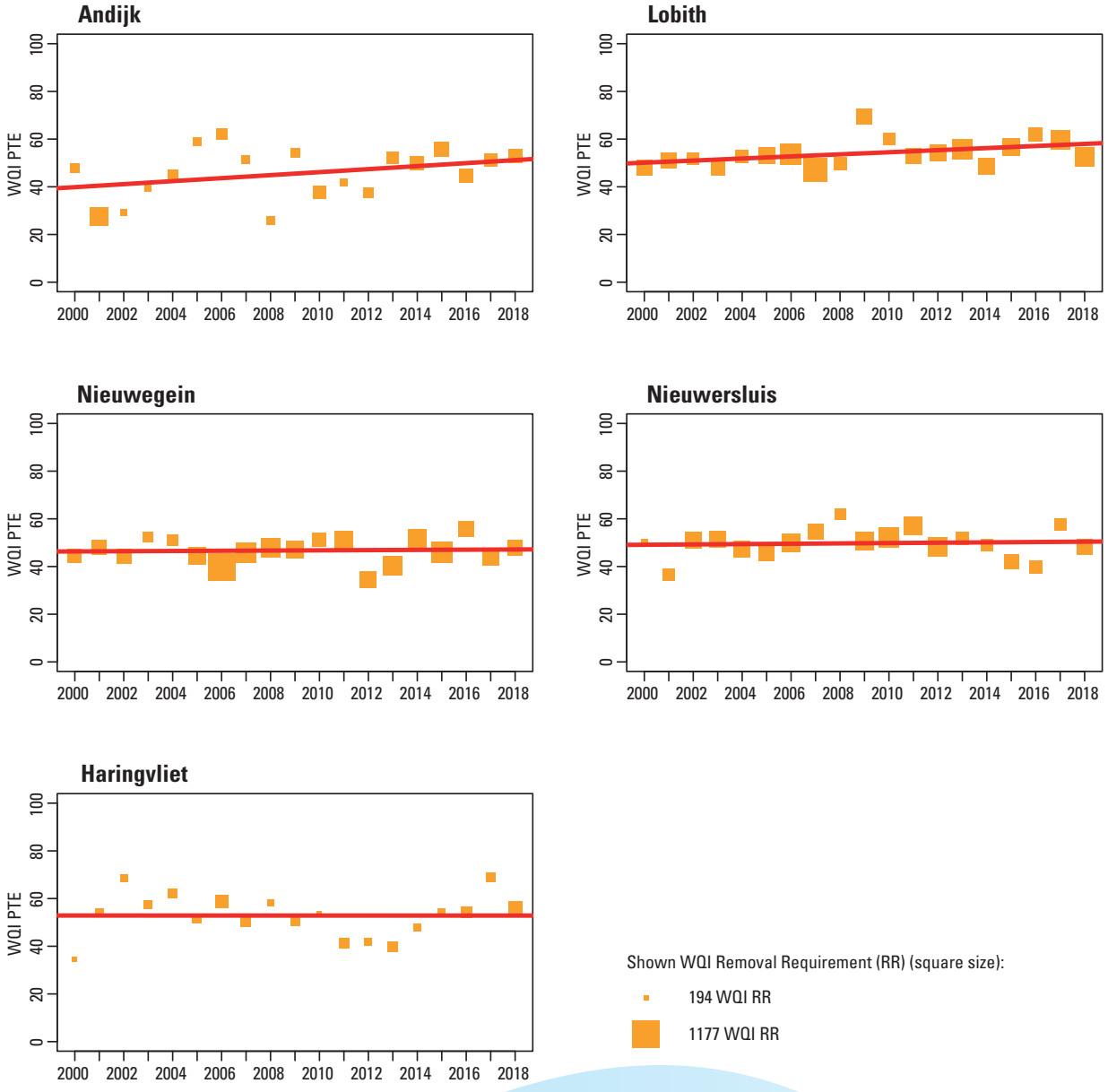
For the negatively correlated parameters (on the left-hand side of each plot in Figure 10) the removal requirement for the parameters is low if the water discharge is high, and vice versa. Dilution effects may play a role here. For the positively correlated parameters (on the right-hand side of each plot), the higher the water discharge, the higher the removal requirement. An increased run-off or increased emissions with higher discharge may play a role there. Roughly, positively correlated are Nutrients and general parameters (Iron, Aluminium, Ammonium). Negatively correlated are pesticides (the sum parameter "Pesticides", and AMPA) and industrial parameters (Melamin, Diglyme, Pyrazole, also AMPA). Also one pharmaceutical was negatively correlated (Diaminomethylideneurea or Guanylurea).

In conclusion, with periods of low discharge, the WQI RR may increase since substances are less diluted. However, it needs to be further investigated to what extent the use and emission of these substances correlate with variation in monthly discharge. Nutrients and general parameters show an opposite relation, and tend to have a high removal requirement at high discharge.

3.5 The purification treatment effort index WQI PTE

The effort of meeting the removal requirement as explained in paragraphs 2.1 and 3.2 to 3.4 can differ depending on the difficulty to remove parameters. We address this aspect by developing a purification treatment effort index WQI PTE (see Equation 4). This index indicates per year if substances contributing to the WQI RR are easy or hard to remove. A high value indicates that the substances in the removal requirement are hard to remove during drinking water treatment. A low value indicates the opposite, parameters are easy to remove. Figure 11 shows the WQI PTE for all five locations over 2000-2018. To indicate a relation with the WQI RR, the size of the squares represents the height of the WQI RR (see Figure 6).

Figure 11. The calculated WQI PTE per year at five measurement locations in the Rhine, from 2000-2018. The solid (red) line is a linear regression through the values of the WQI PTE.



According to Figure 11 the WQI PTE is not improving during the last 20 years for the Rhine river locations. The index indicates that in recent years, substances that have exceeded their DWB value have not become easier to remove. Figure 11 shows the relation between WQI PTE and WQI RR by the size of the squares. For instance Lobith in 2007 has a relatively high WQI RR (large square), however this consists of relatively easy to remove parameters (low WQI PTE value). Lobith in 2009 has a relatively low WQI RR (small square), however the removal is on average more difficult (high WQI PTE value).

Table 5 shows the significance (p-values) of the trends observed in Figure 11. The WQI PTE (see Equation 4) for parameters in Lobith, and to a lesser certainty Andijk, do show an increasing trend. This means that towards later years, more parameters that exceed their DWB value tend to have difficult removal. However, none of the trends are significant. Longer time series are required to unravell whether this increase becomes significant in future. In none of the locations there is a trend towards a lowering of the WQI PTE.

Table 5. Trend in WQI PTE between 2000-2018. A green box means an improvement. A red box indicates a deterioration with regard to water quality. Numbers indicate the significance (p-value) of the trend. Not coloured are trends with p › 0.5.

	Andijk	Lobith	Nieuwegein	Nieuwersluis	Haringvliet
WQI PTE 2000-2018	0.28	0.06	0.99	0.83	0.98

Summarizing, none of the locations show a significant decline in WQI PTE. This means the removal for substances has not become easier in recent years, while this is a goal in the WFD ('...reduce required purification treatment level... '). For two locations there is an indication of deterioration, although the trend is not significant. This is mainly due to the fact that substances that are reported to exceed their target in more recent years are -on average-, more difficult to remove.

3.6 Combining the WQI RR and WQI PTE for a residual removal indication

Required removal (WQI RR) combined with the efficiency of removal (WQI PTE) gives an indication of to what extent the parameters that exceed their target value in the DWB (2018) to a certain level, are removable by purification treatment. If the Removal Indication (RI) (eq. 3) is insufficient to cover the Removal Requirement (RR) for a parameter i, there will be a residual removal requirement (RRR). Equation 5 shows how this combination can be made for each parameter i. If a parameter i has a removal indication that *exceeds* the removal requirement, this is the case when (100-RI) › RR, the residual removal requirement is set to zero. This means negative values are not included in the calculation of the WQI_RR_PTE (see Equation 6), only parameters that have a positive value depending on their RR and RI (see Equation 5), are included.

$$RRR_i = \frac{RR_i - (100 - RI_i)}{100 - (100 - RI_i)} \qquad \text{(Equation 5)}$$

$$WQI\_RR\_PTE = \sum_{1}^{n} RRR_i \qquad \text{(Equation 6)}$$

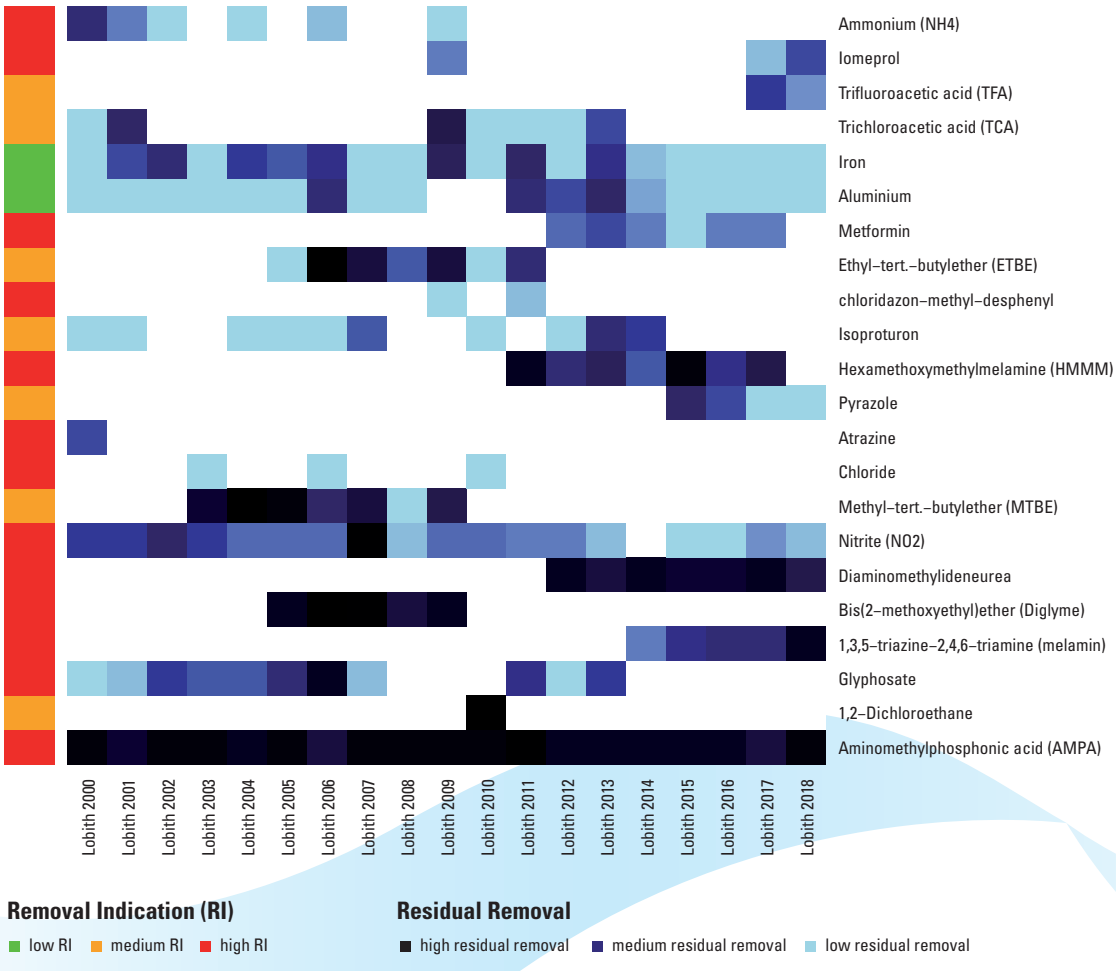
Table 6 shows the significance (p-values) of the trends in WQI RR PTE per location. This index is calculated as the sum of RRR over all parameters i (see Equation 6). This is in accordance with the calculation of the WQI RR in Equation 2. The more manifold the problem, the higher the index. In all but one (Nieuwersluis) location there is an increasing trend in the WQI RR PTE. Two of the trends (Andijk and Nieuwegein) have a near significant trend (p ‹ 0.05). This means that at these locations towards later years, the combination of WQI RR and WQI PTE consists of more parameters that have indications for difficult removal by the size of their exceedance of the DWB (2018) value and/or difficult removal. In none of the locations there is a trend towards a lowering of the WQI RR PTE. Some trends are almost significant towards a higher WQI RR PTE over the years.

Table 6. Trend in WQI RR PTE between 2000-2018. A green box means an improvement. A red box indicates a deterioration with regard to water quality. Numbers indicate the significance (p-value) of the trend. Not coloured are trends with p › 0.5.

	Andijk	Lobith	Nieuwegein	Nieuwersluis	Haringvliet
WQI RR PTE 2000-2018	0.07	0.31	0.05	0.52	0.41

Because Lobith is of strategic value, we show the RRR (see Equation 5) for parameters that exceed their value in the DWB (2018) from 2000 to 2018 for location Lobith as an example in Figure 12. Shown are the parameters for which the Removal Indication, RI (Equation 3), was insufficient indication for complete removal of all exceedances as calculated in the Removal Requirement, RR (Equation 1), resulting in a Residual Removal Requirement, RRR (Equation 5).

Figure 12. The calculated residual removal requirement RRR per parameter for Lobith. On the left, in green to red, the removal indication per parameter (RI). Right is the RRR per parameter from light blue to black. White spots are parameters that did not have a removal requirement (RR) for that year or parameters for which in that year the RI was enough to cover the RR (see Equation 5).



At Lobith, thirteen of the total of twenty-two substances with a residual removal requirement RRR have a high removal indication RI (left bar in Figure 12, red). Logically, substances with high RI have a tendency to have high RRR, and require more extensive purification treatment. Only two substances with high RI from all parameters with a removal requirement RR were completely removed (not shown). This is because these parameters exceeded their DWB value with a low percentage. Most parameters with a low RI (easy removal) do not have any residual removal requirement at any moment because the RI is high enough to remove the RR for that parameter. All years have zero RRR for those substances (not shown).

Iron and Aluminum have a low intrinsic RI (left bar in Figure 12, green) as they are relatively easy to remove, however these parameters exceed the DWB value to such a high extent (RR) that their RI is not enough to indicate complete removal.

We can infer from Figure 12 that the combination of required removal and the ease of removal for parameters, that is a measure for required level of purification treatment, has a tendency to deteriorate in later years for this location, Lobith, but also others (see Table 5). There is a residual removal requirement implied for some parameters by the 'simple' treatment assumed in the WQI PTE for the substances and this has no tendency to decrease in recent years. This is in contrast what the WFD requires: reducing the required level of purification treatment in 2027.

Discussion

Drinking water needs to meet the quality standards of the drinking water directive and should not contain substances that adversely affect human health (DWB, 2018). Preferably, it contains as few chemical substances as possible. This poses a challenge to drinking water companies. They need to apply adequate purification treatment to obtain drinking water of impeccable quality. Higher purification treatment levels are obtained with a more efficient removal process, which implies more effort and higher costs for purification treatment installations. The WFD article 7.3 states that *"Member States shall ensure the necessary protection for the bodies of water identified with the aim of avoiding deterioration in their quality in order to reduce the level of purification treatment required in the production of drinking water."* It is necessary to have a method to evaluate this level of purification treatment required.

We developed two indices for an evaluation of source water quality in relation to level of purification treatment required. First, the WQI RR indicates the need to remove parameters that exceed their target value for good water quality to a level at which these are within the limits again. Second, the WQI PTE indicates how easy it is to remove parameters that exceed their target value. These two indices combined indicate the residual required removal, WQI RR PTE. This is a combination of the removability of the parameters as indicated by the WQI PTE and the removal requirement associated to these parameters as indicated by the WQI RR. Compared over a sequence of years, these indices reveal trends in the required level of purification treatment at different locations in the river Rhine.

The WQI RR is quite factual, and can be calculated for any source water as the sum of percentages of parameters exceeding their target value. As we calculate removal requirement in the WQI RR in percentages of exceedances of the target values in the DWB (2018) the weight of each parameter has a maximum of a hundred and a minimum of zero. If a parameter in a certain year is found to exceed its target value, it contributes to the index, indicating that the purification treatment level should ideally be able to cope with each of these pollutions.

The WQI PTE is more subjective, being calculated by weights that are assigned to traits that we selected to indicate an ease of removal. The removal efficiency is at the moment estimated as a 'simple' process with biodegradation and sorption. Because the assumptions are equally applied to all parameters, a development in WQI PTE can be observed throughout the years as different parameters are temporally exceeding their target value. The weights in the WQI PTE can be replaced, though, by actual removal efficiencies of parameters in a specific drinking water purification treatment installation, or those of a specific removal technique of interest, if needed. The WQI PTE can be used to see if the required effort to remove the substances has declined over the years. The WQI PTE is presently based on two traits, the biodegradability and log K_{ow} (Fischer et al., 2011). More traits affect purification treatment efficiency such as sorption processes based on electrostatic interaction, volatility, chemical reactivity and molecular size (van Leeuwen et al., 2007, van Wezel et al., 2017, de Munk et al., 2018). In general, the compounds that are hydrophilic, small, have low volatility, high solubility, low biodegradability and reactivity and adsorption will be hard to remove. Adding more traits may improve removal efficiency correlation (de Vries et al., 2013). However, the exact treatment steps and constellation of drinking water treatment plants are a big factor in determining the purification treatment efficiency for different micro-pollutants (Stackelberg et al., 2007; Fischer et al., 2019). Therefore it is difficult to determine a generic purification treatment efficiency for any compound. Treatment efficiencies are mostly studied per treatment step (e.g. activated carbon, UV treatment) and the treatment efficiency of compounds varies with each of these steps. Even within treatment steps between different installations this efficiency differs (de Munk et. al., 2018) for instance because of exact doses of purification aids used, or fouling degree of membranes. The WQI PTE should be seen as a very rough indication, and has room for improvement. One addition may be including an uncertainty margin. Also, the bins with values for log K_{ow} (Fischer et al., 2011) at present are quite linear whereas the K_{ow} values are on a log scale, this could be further optimized. Another is to define the simple treatment versus extra treatment, or very advanced treatment and to link what chracteristics of substances are important in each of these levels.

On another note, the predictive model may be improved by using more direct parameters like molecular weight, aromaticity, and e.g. the number of halogens present, instead of indirect parameters as K_{ow} and biodegradability.

The WQI RR and WQI PTE in this report are 'open' indexes, to which 'all' micro-pollutants that have a removal requirement contribute. The required purification treatment level is represented in the calculation of the WQI RR and WQI PTE by including all, including the upcoming and newly measured, parameters in the calculation. It is important to do so, as new substances appear constantly. The advantage of the WQI RR and WQI PTE is the independence between timeframes or locations because there is no need for a fixed list of parameters. A measurement program aimed at reporting all relevant parameters is required, though. This approach is suitable for locations where the measurement program has the means and the goal of measuring 'all' known or suspected micro-pollutants. For locations where this is not the case, the comparison of the WQI RR between locations will be hampered by gaps in the reported parameters. It is recommended to stick to a fixed set of parameters, in those cases.

We compare the concentrations of substances found in source water (the highest within a timeframe) to their target values in the drinking water directive, not to target values applying to the source water itself. The WQI RR and WQI PTE and their combination WQI RR PTE are therefore specifically suitable to assess the source water quality in terms of what needs to be improved in the source water by treatment installations before 'good' quality, according to set target values, is reached. The current index is not suitable for indicating environmental or human adverse effects, as the exposure period to the pollution is not incorporated, nor are all water quality standards and used thresholds directly linked to human health, as some parameters might not be an issue from a human health perspective, but are relevant for ecology, aesthetics, or belong to a group of chemicals sharing a threshold of which only a few are actually an ecological or human health issue. It is an index which describes the quality of the source water in relation to the regulatory quality requirements that drinking water in the Netherlands must meet.

Despite all assumptions and potential drawbacks of the indices and calculations, applying such a calculation framework, reveals differences between locations and years in a rather objective and reproducible manner that allows us to study trends of very complex data. This study was designed to answer the question whether the WFD (2000) measures are sufficient to improve water quality for a reduction of required purification treatment level. The calculated WQI RR does not decrease and rather tends to increase at locations on the Rhine, while the aim is to lower it, in accordance with Article 7.3 of the WFD (2000). The calculated WQI PTE does not decrease either and tends to increase at two locations, whereas at the other locations it shows no trend. The combination of WQI RR and WQI PTE via the WQI RR PTE, a net result of removal requirement and removal indication, increases at four locations, of which two have a (near) significant trend.

Zooming in on the specific parameters that cause the deterioration for the WQI RR, we see a dynamic behaviour. In addition to substances appearing incidentally and disappearing, or substances that are constantly present, new substances also appear. This is to be expected because as many as 22468 chemical substances are registered in 2019 to be in use in the European union (https://echa.europa.eu/nl/information-on-chemicals/registered-substances) and this number is increasing every year. An increasing number of chemicals find their way into the freshwater system (van Wezel et al., 2017). All these chemicals are unwelcome in our drinking water.

Parameter groups "Plant protection products, biocides and their metabolites" and "General parameters and nutrients" in particular form a large part of the WQI RR over the years. "Plant protection products, biocides and their metabolites" are an increasing problem in some of the locations. "Industrial contaminants and consumer products" are unpredictable and cause peaks and troughs in the water treatment index over the years. "Pharmaceuticals and endocrine disrupting chemicals (EDCs)" form a small group, but this is increasing at every location without exception and is therefore of increasing concern.

Based on these calculations in these three developed indices it cannot be concluded that the measures taken so far to achieve the objectives of the WFD have led to the improvement of the removal requirement or the purification treatment efficiency, or a combination of those, in the way they are defined as indexes for water quality in this report. The WQI RR has not decreased for the Rhine water locations in the period from 2000-2018 and neither has the WQI PTE and their combination the WQI RR PTE, that represents the level of required purification treatment. At the same time, drinking water production companies have made preparations to do the opposite from reducing purification treatment levels and have increased purification treatment levels to meet drinking water targets. New advanced techniques are being implemented.

Apparently, extra effort is needed in the field of emission reduction, with a focus on new and emerging substances, in order to reduce the purification treatment level required for the preparation of drinking water. Measurement programs must remain attuned to this, whereby it is important to get hold on possible problem substances, so that these are monitored in time. Adequate measures should be taken to limit emissions of these substances.

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Appendix 1

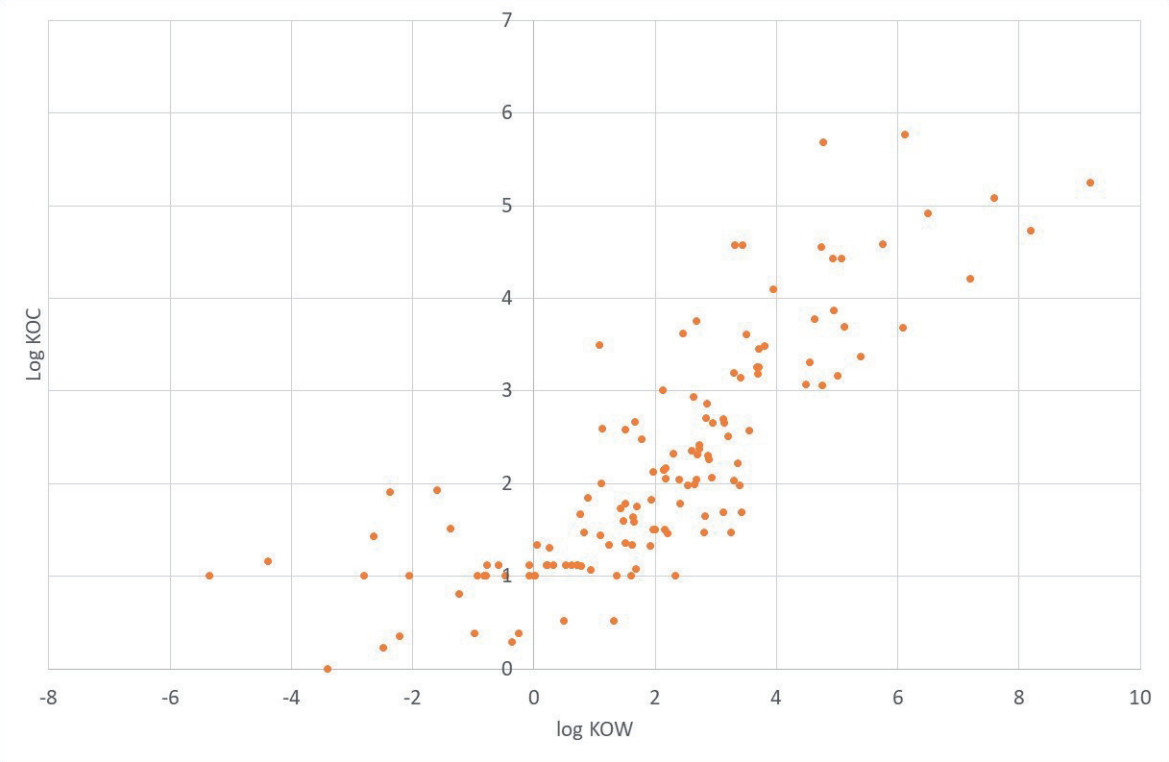
Evaluation of model predictions

For the calculation of the removal indication RI three types of traits were considered:

- Log K_{OW}, showing the distribution of a compound between an organic phase and the water phase. It is a measure of hydrophobicity
- Log K_{OC}, showing the soil adsorption coefficient. This too is a kind of measure of hydrophobicity
- Biodegradability, Biowin 3.

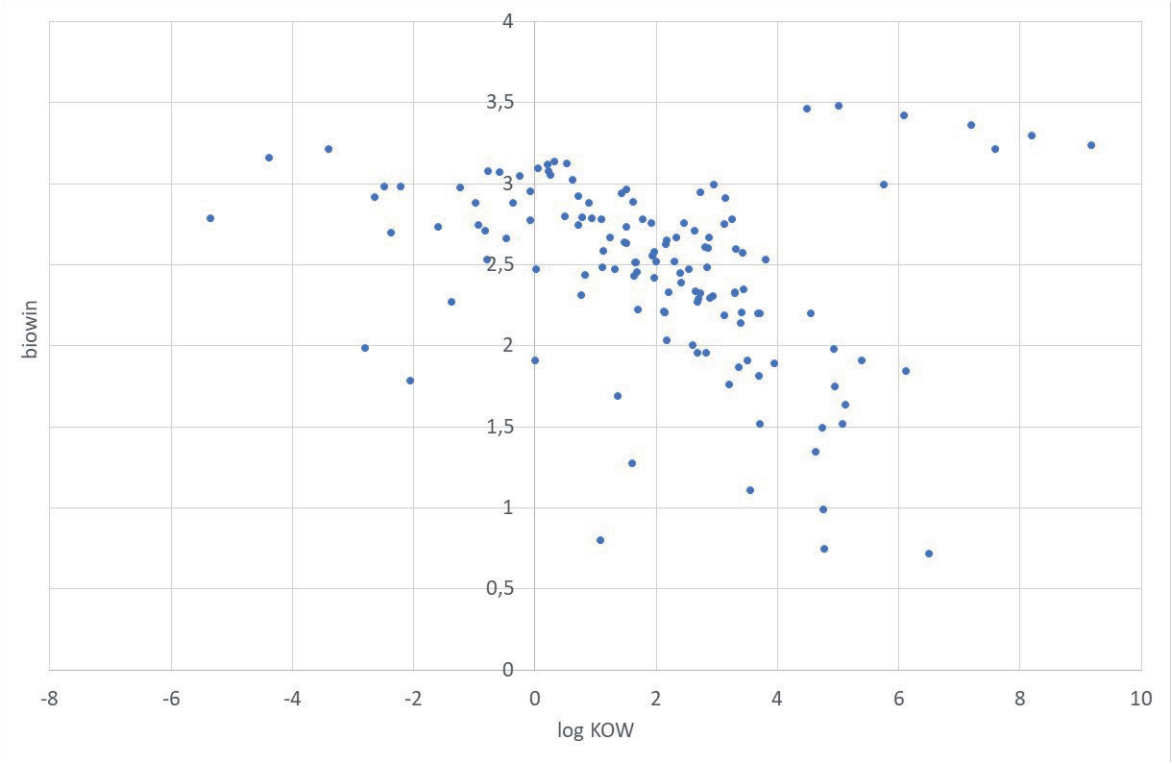
From the above definitions, it can be assumed that there is a strong relationship between log K_{ow} and log K_{OC}. This is confirmed by Figure A.1, which shows this relationship for 135 contaminants found exceeding their target value in the DWB (2018) in this report.

Figure A.1. relationship between log K_{OC} and log K_{ow} for 135 contaminants



Biodegradability depends on both molecular weight and composition. In general it is assumed that low molecular weight compounds are more easily degraded than large compounds. As a result, there also should be a relationship between biodegradability and e.g. log K_{OW}, as hydrophobicity too is strongly determined by the molecular mass of a compound. This relationship is shown in Figure A.2.

Figure A.2. relationship between biodegradability and log K_{ow}



Indeed there is some correlation, but not as strong as between log K_{ow} and log K_{OC}. This is caused by the presence of various atoms in the molecular structure, which makes it better or less biodegradable. Therefore, it was decided to apply both log K_{ow} and the biodegradability for this model prediction.

Both adsorption and biodegradation processes occur during drinking water treatment. Filtration over sand or activated carbon is often applied. Rapid sand filtration is applied in the majority of treatment processes for the removal of flocculates, consisting of iron (or aluminium), natural organic matter, particles, and other metals like manganese. The sand contains micro-organisms, which are able to biologically degrade contaminants. A few years ago it was found that in this way a contaminant like pyrazole could be removed from the water. However, at the same time the vulnerability of this biodegradation process was shown, as it can only function well if the right types of micro-organisms are present. Sand filters which had never been in contact with pyrazole before, or for which the pyrazole concentrations had decreased for some time, were not able to remove this contaminant (anymore). Filtration over activated carbon also is often applied, mainly for the removal of organic micro-pollutants. Here too biomass is present, resulting in the biodegradation of some compounds. Where the adsorption mechanism preferably applies to large organic molecules, biodegradation in general applies to low molecular weight species. Therefore, it was suggested that basing the predictive model on both log K_{ow} and biodegradability, might result in an estimation of the RI value.

De Munk et al. (2018) reported that the log K_{OC} constants correlate (to some extent) with removal efficiency. Log K_{OC} is a soil adsorption coefficient. This too is a kind of measure of hydrophobicity. The parameters log K_{ow} and log K_{OC} are highly correlated between themselves (see Figure A.1) so using both would overestimate the removal. Also vapor pressure relates

to removal efficiency (van Leeuwen et al. 2007). There is however a limited number of parameters in open surface waters for which vapor pressure determines their quick removal, because these parameters are already present for a long time (days).

For a set of 135 contaminants the method was applied, and the results were compared to expert judgement on the removal rates of the contaminants in drinking water processes. The results are shown in Table 1. Expert judgement was based both on information on molecular structure (molecular weight, aromaticity, length of C-chains, presence of charged or polar groups, presence of e.g. halogens) and on experiences with pilot and full scale treatment processes. For the predictions based on multiplied parameters, it was found that in 89 cases (66%) the prediction was in accordance with expert judgement. In 18 cases (13%) a higher removal rate than expected was found, whereas in 28 (20%) cases a lower removal rate was predicted.

Thus, it can be concluded that in about 66% of the cases the removal of a compound is predicted fairly well by the model.

Table A.1. overview of predicted removal rates and expert judgement for parameters that exceeded their target value in at least one location (see Figure 2).

nr.	CAS nr.	Compound	Predicted removal (100-RI)	Validation of new prediction*	Remarks
1	108-78-1	1,3,5-triazine-2,4,6-triamine (melamin)	17	OK	
2	576-24-9	2,3-Dichlorophenol	44	OK	
3	7429-90-5	Aluminium	90	OK	Al in general is added as coagulant, precipitates, and is removed by rapid sand filtration
4	1066-51-9	Aminomethylphosphonic acid (AMPA)	17	OK	
5	61-82-5	Amitrole	17	OK	
6	111-96-6	Bis(2-methoxyethyl)ether (Diglyme)	17	-	
7	16887-00-6	Chloride	17	OK	
8	124-18-5	Decane	78	OK	
9	117-81-7	Bis(2-ethylhexyl)phthalate (DEHP)	100	OK	
10	84-74-2	Di-n-butylphthalate (DBPH)	78	OK	
11	112-40-3	Dodecane	100	OK	
12	1071-83-6	Glyphosate	17	-	
13	141-83-3	Diaminomethylideneurea	17	OK	
14	3089-11-0	Hexamethoxymethylmelamine (HMMM)	33	+	
15	544-76-3	Hexadecane	100	OK	
16	7439-89-6	Iron	90	OK	Precipitates as (hydr)oxide
17	7439-96-5	Manganese	90	OK	Precipitates as (hydr)oxide
18	657-24-9	Metformin	17	OK	
19	14797-65-0	Nitrite (NO2)	17	OK	Only removable by means of biological process, but requires specific circum-stances. Relatively difficult to remove.
20	593-45-3	Octadecane	100	OK	
21	114-26-1	Propoxur	44	OK	
22	288-13-1	Pyrazole	44	OK	Can be removed by biodegradation, but only if specific micro-organisms are present, which is not always the case.

** Validation of new prediction based on multiplied values, by expert judgement. Good = OK. + prediction too high; - prediction too low.*

Continuation table A.1

nr.	CAS nr.	Compound	Predicted removal (100-RI)	Validation of new prediction*	Remarks
23	122-34-9	Simazine	33	+	
24	629-59-4	Tetradecane	100	OK	
25	55297-95-5	Tiamulin	67	OK	
26	5611-51-8	Triamcinolonehexacetonide	67	OK	
27	76-03-9	Trichloroacetic acid (TCA)	44	OK	In general difficult to remove. Possibly by means of biodegradation.
28	76-05-1	Trifluoroacetic acid (TFA)	44	OK	In general difficult to remove. Possibly by means of biodegradation.
29	126535-15-7	Triflusulfuron-methyl	67	OK	
30	94-75-7	2,4-Dichlorophenoxyacetic acid (2,4-D)	44	OK	
31	93-76-5	2,4,5-Trichlorophenoxyacetic acid (2,4,5-T)	72	+	
32	94-74-6	4-Chloro-2-methylphenoxyacetic acid (MCPA)	72	OK	
33	1912-24-9	Atrazine	33	OK	
34	11141-17-6	azadirachtin A	33	-	
35	25057-89-0	Bentazon	44	OK	
36	50-32-8	Benzo(a)pyrene	100	OK	
37	18181-80-1	Bromopropylate	67	OK	
38	34681-10-2	Butocarboxim	44	OK	
39	34681-23-7	Butoxycarboxim	17	OK	
40	10605-21-7	Carbendazim	44	-	
41	15545-48-9	Chlortoluron	44	-	
42	1918-00-9	Dicamba	44	OK	
43	1194-65-6	Dichlobenil	44	OK	
44	60-51-5	Dimethoate	44	OK	
45	110488-70-5	Dimethomorph	33	-	
46	330-54-1	Diuron	44	OK	
47	2593-15-9	Etridiazole	67	+	
48	67564-91-4	Fenpropimorph	67	-	
49	93-65-2	Mecoprop (MCPP)	72	+	
50	57837-19-1	Metalaxyl	44	OK	
51	2179-25-1	Methiocarb-sulfone	44	OK	
52	1634-04-4	Methyl-tert.-butylether (MTBE)	44	OK	
53	150-68-5	Monuron	44	OK	
54	91-20-3	Naphthalene	72		
55	111991-09-4	Nicosulfuron	33	-	
56	82-68-8	Quintocene	67	+	
57	107534-96-3	Tebuconazole	67	OK	
58	5915-41-3	Terbutylazine	67	+	
59	148-79-8	Thiabendazole	44	-	
60	57018-04-9	Tolclofos-methyl	67	OK	
61	107-06-2	1,2-Dichloroethane	44	-	Removal by evaporation during aeration
62	51-28-5	2,4-Dinitrophenol	44	-	
63	95-95-4	2,4,5-Trichlorophenol	67	+	
64	88-06-2	2,4,6-Trichlorophenol	67	+	

** Validation of new prediction based on multiplied values, by expert judgement. Good = OK. + prediction too high; - prediction too low.*

Continuation table A.1

nr.	CAS nr.	Compound	Predicted removal (100-RI)	Validation of new prediction*	Remarks
65	7440-43-9	Cadmium	90	OK	
66	58-08-2	Caffein	17	OK	
67	75-01-4	Chloroethene	44	OK	
68	2921-88-2	Chlorpyriphos-ethyl	67	+	
69	1698-60-8	Chloridazon	44	-	
70	120-36-5	2,4-Dichlorprop (2,4-DP)	72	+	
71	108-20-3	Di-iso-propylether	44	OK	
72	163515-14-8	Dimethenamid-p	33	-	
73	67-64-1	Acetone	17	OK	
74	16984-48-8	Fluoride	17	OK	
75	34123-59-6	Isoproturon	44	-	
76	7439-92-1	Lead	90	OK	
77	67129-08-2	Metazachlor	33	+	
78	51218-45-2	Metolachlor	67	+	
79	19937-59-8	Metoxuron	44	OK	
80	7440-02-0	Nickel	90	OK	
81	25154-52-3	Nonylphenol	72	-	
82	298-00-0	Parathion-methyl	44	OK	
83	23103-98-2	Pirimicarb	33	OK	
84	14808-79-8	Sulfate	17	OK	
85	10543-57-4	Tetraacetylethylenediamine (TAED)	17	OK	
86	127-18-4	Tetrachloroethene	67	-	Removal by evaporation during aeration
87	79-01-6	Trichloroethene	44	_	Removal by evaporation during aeration
88	88-75-5	2-Nitrophenol	44	OK	
89	2008-58-4	2,6-Dichlorobenzamide (BAM)	44	OK	
90	7440-36-0	Antimony	90	OK	
91	101-27-9	Barban	67	+	
92	95-14-7	Benzotriazole	44	-	
93	80-05-7	Bisphenol A	72	OK	
94	15541-45-4	Bromate	90	OK	
95	333-41-5	Diazinon	72	+	
96	134-62-3	Diethyltoluamide (DEET)	44	-	
97	77182-82-2	Glufosinate-ammonium	17	OK	
98	3567-62-2	1-(3,4-Dichlorophenyl)-3-methylurea (DCPMU)	44	OK	
99	1646-87-3	Aldicarb-sulfoxide	17	OK	
100	309-00-2	Aldrin	100	+	
101	62-53-3	Aniline	44	OK	
102	75-27-4	Bromodichloromethane	44	OK	
103	124-48-1	Dibromochloromethane	44	OK	
104	87674-68-8	Dimethenamid	33	OK	
105	26225-79-6	Ethofumesat	44	OK	
106	72-43-5	Methoxychlor	67	OK	
107	108-88-3	Methylbenzene	44	OK	

** Validation of new prediction based on multiplied values, by expert judgement. Good = OK. + prediction too high; - prediction too low.*

Continuation table A.1

nr.	CAS nr.	Compound	Predicted removal (100-RI)	Validation of new prediction*	Remarks
108	311-45-5	Paraoxon-ethyl	44	OK	
109	24579-73-5	Propamocarb	44	OK	
110	39184-27-5	Thiofanox-sulfoxide	44	OK	
111	67-66-3	Trichloromethane	44	-	Removal by evaporation during aeration
112	100-42-5	Ethenylbenzene	44	OK	
113	637-92-3	Ethyl-tert.-butylether (ETBE)	44	OK	
114	78649-41-9	Iomeprol	0	-	
115	1746-81-2	Monolinuron	44	OK	
116	87-86-5	Pentachlorophenol	67	+	
117	2327-02-8	1-(3,4-Dichlorophenyl)urea (DCPU)	44	OK	
118	626-43-7	3,5-Dichloroaniline	44	OK	
119	319-85-7	beta-Hexachlorocyclohexane (beta-HCH)	67	+	
120	126833-17-8	Fenhexamid	67	-	
121	18691-97-9	Methabenzthiazuron	44	OK	
122	23135-22-0	Oxamyl	17	OK	
123	117-96-4	Diatrizoic acid (Amidotrizoic acid)	33	OK	
124	75-09-2	Dichloromethane	44	-	Removal by evaporation during aeration
125	100-41-4	Ethylbenzene	72	OK	
126	73334-07-3	Iopromide	0	-	
127	61869-08-7	paroxetine	67	-	
128	56-23-5	Tetrachloromethane	33	-	Removal by evaporation during aeration
129	171118-09-5	metolachloro-S-metabolite	44	OK	
130	161326-34-7	Fenamidone	72	OK	
131	34681-24-8	Butocarboxim-sulfoxide	17	OK	
132	658066-35-4	Fluopyram	67	OK	
133	6339-19-1	Chloridazon-desphenyl	17	OK	
134	92075-50-8	Ammonium (N)	17	-	
135	152019-73-3	metolachloro-C-metabolite	44	OK	

** Validation of new prediction based on multiplied values, by expert judgement. Good = OK. + prediction too high; - prediction too low.*

Some remarks on Table A.1

Most heavy metals are removed during coagulation/flocculation/sedimentation, followed by rapid sand filtration.

Anions like chloride, fluoride and sulfate are very difficult to remove. In general concentrations are so low that it's no problem to meet drinking water standards, but as a result of salinization and dry hot summers in some places the chloride concentrations in the influent water may become (too) high.

Nitrite, nitrate and ammonium can be biologically removed, but only under special conditions. These are not always applied in drinking water treatment plants. A common way to deal with too high nitrate concentrations is to mix the water with water with a much lower nitrate concentration.

Halogen containing alkanes (chloro methane, ethane) and alkenes in general are removed during aeration, as they are very volatile. Their removal is not or only partly related to biodegradation. Adsorption and filtration in general are not very effective, as the molecules are very small.

100% removal is predicted for 1,2-Benzenedicarboxylic acid, bis(2-ethylhexyl) ester, whereas "only" 78% is predicted for 1,2-Benzenedicarboxylic acid, dibutyl ester (compounds 9 and 10). This difference is not expected in practical installations. Here, the model seems to indicate a higher accuracy than is realistic. The same applies to compounds 8, 11 and 15 (Decane, Dodecane, and Hexadecane). All three compounds will be removed to a high degree, but there is no reason to assume that the removal of dodecane will be significantly better (100 % predicted) than that of decane (78% predicted).

For compounds 21 and 22 (Phenol, 2-(1-methylethoxy)-, methylcarbamate and pyrazole) both 44% removal is predicted. However, it is expected that compound 21 can be removed more easily than compound 22, as it is larger and thus will adsorb better onto e.g. activated carbon. Furthermore, as mentioned above, pyrazole can only be removed by biodegradation if the right kinds of micro-organisms are present in the sand filter. If the sand filter has never before been into contact with pyrazole, or if the pyrazole concentration in the water has been significantly lower for a while, biodegradation will be limited or even prevented.

Compounds 27 and 28 (TCA and TFA) are predicted to be removed to a degree of 44%. However, this is considered very high, as these small compounds cannot or hardly be removed by means of filtration and adsorption (or oxidation) processes. Biodegration may occur, but in a drinking water process contact times will probably be too low to obtain a high degradation rate.

For compound 31 (Acetic acid, (2,4,5-trichlorophenoxy)-) 72% removal is predicted, whereas for compound 30 (Acetic acid, (2,4-dichlorophenoxy)-) only 44% removal is predicted. However, compound 31 contains some more chlorine atoms, so it is unlikely that this compound will show a higher removal rate. For both a low removal rate is expected.

For compound 38 (Butocarboxim) 44% removal was predicted, whereas for compound 39 (Butoxycarboxim) only 17% removal was predicted. However, it is to be expected that both are difficult to remove, and in fact that compound 38 would even be harder to remove than compound 39, as the latter contains a SO2-group.

Compounds 63 (2,3,5-trichloro phenol) and 64 (2,4,6-trichloro phenol) show a predicted removal rate of 67%, whereas for compound 62 (2,5-dinitro phenol) only 44% is predicted. However, as the first two compounds contain much more polar Cl-groups, and compound 62 only nitro-groups, it would be expected the other way around.

For compounds 77 and 78 (metazachlor and metolachlor, respectively) a large difference in removal rate is not expected. From real drinking water treatment plants it is known, that both compounds are very hard to remove.

Compound 86 (tetrachloro ethene) a removal of 67% is predicted, whereas for compound 87 (trichloro ethane) only 44% is predicted. Both compounds have a very similar structure, and such a difference is not very likely. Besides, both will be difficult to remove by means of filtration or adsorption processes. Evaporation is a more likely removal process, as they are relatively volatile. Sub-soil biodegradation of these compounds is known, but with long residence times.

Summarizing, it can be seen that in about 66% of the cases the model gives a fair prediction of its removal rate. The model sometimes results in a different prediction for similar compounds. This may be attributed to the fact that log K_{ow} and biodegradability to a certain extend are related parameters, which both are calculated from (partly the same) molecular properties. The predictive model may be improved by using more direct parameters like molecular weight, aromaticity, and e.g. the number of halogens present, instead of indirect parameters as K_{ow} and biodegradability.

Appendix 2

Tables with Removal Requirements per year, per location

Andijk	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	CAS nr.
Aminomethylphosphonic acid (AMPA)	58.3	63	41.2	60	96	81.5	88	79.2	70.6	65.5	66.7	79.2	66.7	69.7	72.2	73.7	56.5	67.7	67.7	1066-51-9
Glyphosate	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	9.1	0	0	28.6	1071-83-6
1,3,5-triazine-2,4,6-triamine (melamin)	0	0	0	0	0	0	0	0	0	0	0	0	0	16.7	9.1	56.5	37.5	47.4	23.1	108-78-1
Bis(2-methoxyethyl)ether (Diglyme)	0	0	0	0	0	0	21.3	0	0	0	0	0	0	0	0	0	0	0	0	111-96-6
Perfluorododecanoic acid (PFDoA)	0	75	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	112-40-3
Propoxur	0	0	0	0	0	0	0	9.3	0	0	0	0	0	0	0	0	0	0	0	114-26-1
Bis(2-ethylhexyl)phthalate (DEHP)	0	0	0	0	0	0	0	0	0	0	54.5	32	0	0	19.4	0	14.5	0	0	117-81-7
Simazine	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	122-34-9
henicosafluoroundecanoic acid (PFUnA)	0	95.5	13	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	124-18-5
Triflusulfuron-methyl	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	28.6	0	126535-15-7
Diaminomethylideneurea	0	0	0	0	0	0	0	0	0	0	0	0	0	37.5	50	9.1	41.2	33.3	0	141-83-3
Nitrite (NO2)	27.5	27.5	25.7	0	53.2	21.9	60.9	15.3	0	0	84.2	0	0	0	0	0	0	0	0	14797-65-0
metolachloro-C-metabolite	0	0	0	0	0	0	0	0	0	0	0	0	0	41.2	41.2	52.4	50	33.3	28.6	152019-73-3
Chloride	0	0	0	16.7	3.2	7.4	2.6	0	0	2.6	0	17.1	0	0	18.9	0	0	24.6	43.4	16887-00-6
metolachloro-S-metabolite	0	0	0	0	0	0	0	0	0	0	0	0	0	61.5	64.3	67.7	63	61.5	58.3	171118-09-5
Pyrazole	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	23.1	0	0	0	288-13-1
Hexamethoxymethylmelamine (HMMM)	0	0	0	0	0	0	0	1.5	0	0	0	0	0	16.7	0	0	0	0	0	3089-11-0
Hexadecane	0	80	13	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	544-76-3
Tiamulin	0	0	0	0	0	0	0	0	0	0	0	0	0	16.7	0	0	0	0	0	55297-95-5
Triamcinolonehexacetonide	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0.3	0	0	0	0	5611-51-8
2,3-Dichlorophenol	0	0	0	0	0	0	0	0	0	0	0	0	85.7	0	0	0	0	0	0	576-24-9
Octadecane	0	75	19.4	0	66.7	0	0	0	71.4	0	0	0	0	0	0	0	0	0	0	593-45-3
Amitrole	0	0	0	0	0	54.5	0	0	0	0	0	0	0	0	0	0	0	0	0	61-82-5
heptacosafluorotetradecanoic acid (PFTDA)	0	83.3	9.1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	629-59-4
Metformin	0	0	0	0	0	0	0	0	0	0	0	0	0	23.1	9.1	0	0	0	0	657-24-9
Aluminium	66.5	32.9	0	57	0	0	0	0	0	0	0	30.3	68.3	64.1	46.1	70	85.3	51.3	83.1	7429-90-5
Iron	88.3	93.9	89.7	84	89.5	83.3	83.3	97.2	80	91.7	86.7	65	83.1	76.1	76.3	91.7	97.5	69.7	89	7439-89-6
Manganese	79.7	82.1	70.6	74.9	50	64.3	88.1	76.2	86.8	84.8	68.8	25.8	69.7	48	24.6	76.2	61.5	53.7	61.5	7439-96-5
Trichloroacetic acid (TCA)	28.6	0	0	0	0	0	0	0	0	69.7	65.5	33.3	16.7	0	81.8	0	9.1	9.1	0	76-03-9
Trifluoroacetic acid (TFA)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	44.4	76-05-1
Di-n-butylphthalate (DBPH)	0	0	0	0	0	0	0	0	0	0	83.6	0	0	0	0	0	0	0	0	84-74-2
Ammonium (NH4)	23.1	47.5	9.1	0	0	0	46.3	0	0	26.4	0	11.2	0	0	0	41.5	0	0	0	92075-50-8
Polycyclische aromatische koolwaterstoffen (PAK's)	0	0	0	0	0	0	9.1	16.7	0	0	0	0	0	0	0	0	0	0	0	
Pesticiden	23.2	26.5	2	9.7	82.4	58.7	58.6	54.2	38	50.8	56	41.4	34.8	59.7	70.2	61.6	60.5	60.4	59.7	

Lobith	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	CAS nr.
Ethenylbenzene	0	0	0	0	0	28.6	0	0	0	0	0	0	0	0	0	0	0	0	0	100-42-5
Aminomethylphosphonic acid (AMPA)	83.9	77.8	86.3	84.8	83.1	84.4	75	85.9	84.8	85.1	85.5	87.1	81.5	81.5	79.6	83.1	82.8	75.2	83.2	1066-51-9
1,2-Dichloroethane	0	0	0	0	0	0	0	0	0	0	94.1	0	0	0	0	0	0	0	0	107-06-2
Glyphosate	16.7	23.1	50	41.2	41.2	56.5	83.1	23.1	0	0	0	54.5	9.1	50	0	0	0	0	0	1071-83-6
1,3,5-triazine-2,4,6-triamine (melamin)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	33.3	52.4	56.5	58.3	81.1	108-78-1
Bis(2-methoxyethyl)ether (Diglyme)	0	0	0	0	0	82	91.7	88.1	73.4	81.1	0	0	0	0	0	0	0	0	0	111-96-6
Bis(2-ethylhexyl)phthalate (DEHP)	0	0	0	0	0	0	0	64.3	0	0	0	0	74.5	0	49.2	0	0	0	0	117-81-7
Diaminomethylideneurea	0	0	0	0	0	0	0	0	0	0	0	0	79.2	73.7	80.8	77.3	75.6	79.2	68.8	141-83-3
Nitrite (NO2)	49.3	49.3	61.9	49.3	39.1	39.1	39.1	89.9	23.9	39.1	39.1	35.2	35.2	23.9	0	7.7	2.7	30	21.3	14797-65-0
Bromate	33.3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	15541-45-4
Chlortoluron	16.7	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	15545-48-9
Methyl-tert.-butylether (MTBE)	0	0	0	85.9	95.5	89.4	75.1	82	39.4	80.5	0	0	0	0	0	0	0	0	0	1634-04-4
Chloride	0	0	0	18.5	0	0	4.3	0	0	0	12.9	0	0	0	0	0	0	0	0	16887-00-6
Monolinuron	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1746-81-2
Atrazine	56.5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1912-24-9
Bentazon	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	41.2	0	25057-89-0
Pyrazole	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	73.7	63.4	33.3	31.8	288-13-1
Parathion-methyl	0	33.3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	298-00-0
Hexamethoxymethylmelamine (HMMM)	0	0	0	0	0	0	0	0	0	0	0	84.6	66.7	71.4	52.4	88.2	63	76.7	0	3089-11-0
Isoproturon	16.7	44.4	0	0	28.6	33.3	16.7	61.5	0	0	41.2	0	9.1	73	67.7	0	0	0	0	34123-59-6
Benzo(a)pyrene	0	0	0	0	0	0	50	90.9	0	0	0	17.4	0	38.7	0	0	0	0	26.5	50-32-8
Metolachlor	0	0	0	0	0	0	0	0	0	0	0	0	23.7	0	0	0	0	0	0	51218-45-2
chloridazon-methyl-desphenyl	0	0	0	0	0	0	0	0	0	9.1	0	23.1	0	0	0	0	0	0	0	6339-19-1
Ethyl-tert.-butylether (ETBE)	0	0	0	0	0	3.9	96.5	82.7	61.3	81.5	18	72.2	0	0	0	0	0	0	0	637-92-3
Metformin	0	0	0	0	0	0	0	0	0	0	0	0	37.5	47.4	33.3	16.7	33.3	33.3	0	657-24-9
Trichloromethane	44.4	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	67-66-3
Aluminium	47.6	73.8	61.6	24.8	58.9	76.5	95.1	83.3	79.2	0	0	95.1	93.7	95.5	91.2	90.4	90.2	88.3	83.3	7429-90-5
Iron	88.6	93.6	95	89.6	93.8	93.1	94.4	89.5	87.5	95.7	85.7	95.3	90.4	94.4	90.7	89	89.4	88	84.6	7439-89-6
Lead	23.1	0	0	69.7	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	7439-92-1
Manganese	28.6	66.7	61.5	64.3	58.3	73.7	64.3	0	15.3	0	0	0	0	0	51	53.7	0	40.3	66.7	7439-96-5
Trichloroacetic acid (TCA)	44.4	75	0	0	0	0	0	0	0	80.8	41.2	9.1	33.3	64.3	0	0	0	0	0	76-03-9
Trifluoroacetic acid (TFA)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	66.7	52.4	76-05-1
Iomeprol	0	0	0	0	0	0	0	0	0	23.1	0	0	0	0	0	0	0	9.1	33.3	78649-41-9
Pentachlorophenol	0	23.1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	87-86-5
Ammonium (NH4)	57.4	33.3	16.7	0	4.8	0	23.1	0	0	20	0	0	0	0	0	0	0	0	0	92075-50-8
Benzotriazole	0	0	0	0	0	0	0	0	9.1	0	0	0	16.7	0	0	0	0	37.5	28.6	95-14-7
PAK's (6 van Borneff)	67.9	70.3	69.8	66.6	58.4	57.4	62.7	64.6	52.9	69.8	63.8	66	62.4	74.5	62	58.5	53.4	50.7	59.1	
Polycyclische aromatische koolwaterstoffen (PAK's)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	29.5	
Pesticiden	0	0	0	9.1	0	0	9.1	97.5	34.9	0	30.1	21.4	0	58.3	0	37.8	0	7	66.4	

Nieuwegein	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	CAS nr.
Aminomethylphosphonic acid (AMPA)	76.2	75.6	0	84.6	90	90	88.5	88.6	89.1	88.9	83.1	87.7	85.7	84.4	81.1	87.7	83.9	84.8	81.5	1066-51-9
Glyphosate	23.1	16.7	0	0	0	72.2	76.7	33.3	0	9.1	28.6	33.3	0	0	16.7	23.1	78.3	0	9.1	1071-83-6
1,3,5-triazine-2,4,6-triamine (melamin)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	54.5	69.7	64.3	63	67.7	108-78-1
Methylbenzene	0	0	0	0	0	0	0	0	23.1	0	0	0	0	0	0	0	0	0	0	108-88-3
Bis(2-methoxyethyl)ether (Diglyme)	0	0	0	0	0	0	58.7	61.3	41.2	9.1	0	0	0	0	0	0	0	0	0	111-96-6
Perfluorododecanoic acid (PFDoA)	0	0	0	0	0	0	91.7	0	0	0	0	0	0	0	0	0	0	0	0	112-40-3
Bis(2-ethylhexyl)phthalate (DEHP)	0	0	0	0	0	0	0	33.3	16.7	0	0	70.6	70.6	68.8	0	0	0	35.5	0	117-81-7
henicosafluoroundecanoic acid (PFUnA)	0	0	0	0	0	0	70.6	0	0	0	0	0	0	0	0	0	0	0	0	124-18-5
Fenhexamid	0	0	0	0	0	0	0	0	0	97.8	0	0	0	0	0	0	0	0	0	126833-17-8
Diaminomethylideneurea	0	0	0	0	0	0	0	0	0	0	0	0	0	54.5	74.4	63	61.5	56.5	23.1	141-83-3
Nitrite (NO2)	25.7	17.7	29.2	61.5	35.2	57.1	27.5	4.8	34.2	0	61.4	18	25.9	2	0	0	2	6.5	33.8	14797-65-0
Bromate	0	0	0	0	0	0	33.3	0	0	0	0	0	37.5	9.1	23.1	37.5	0	9.1	41.2	15541-45-4
Chlortoluron	9.1	61.5	75	0	9.1	0	0	0	33.8	0	0	0	0	0	0	0	0	0	0	15545-48-9
Methyl-tert.-butylether (MTBE)	0	0	0	0	76.2	0	54.5	0	83.3	0	0	88.9	0	0	9.9	0	0	0	0	1634-04-4
Chloride	0	0	0	0.3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	16887-00-6
Monolinuron	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1746-81-2
Methabenzthiazuron	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	18691-97-9
Atrazine	0	23.1	23.1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1912-24-9
Oxamyl	0	0	0	0	0	0	0	0	0	0	0	58.3	0	0	0	0	0	0	0	23135-22-0
1-(3,4-Dichlorophenyl)urea (DCPU)	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	2327-02-8
Bentazon	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	16.7	0	0	0	25057-89-0
Pyrazole	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	28.6	21.1	0	0	288-13-1
Hexamethoxymethylmelamine (HMMM)	0	0	0	0	0	0	0	68.1	28.6	37.5	0	0	0	0	0	0	0	0	0	3089-11-0
beta-Hexachlorocyclohexane (beta-HCH)	0	37.5	16.7	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	319-85-7
Diuron	9.1	0	0	23.1	0	16.7	28.6	41.2	0	0	0	0	0	0	0	0	0	0	0	330-54-1
Isoproturon	64.3	79.6	80.4	0	16.7	0	0	58.3	39.4	0	0	28.6	0	66.7	70.6	0	0	0	0	34123-59-6
Butocarboxim-sulfoxide	0	0	0	0	0	0	0	0	16.7	0	0	0	0	0	0	0	0	0	0	34681-24-8
Benzo(a)pyrene	80	50	80	0	0	90.9	75	87.5	66.7	87.5	50	0	41.9	67.1	0	78.8	0	0	7.4	50-32-8
Metolachlor	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	47.4	0	0	0	51218-45-2
Hexadecane	0	0	0	0	0	0	76.7	0	0	0	0	0	0	0	0	0	0	0	0	544-76-3
Triamcinolonehexacetonide	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	41.2	0	0	0	5611-51-8
3,5-Dichloroaniline	0	0	0	0	0	9.1	0	0	0	0	0	0	0	0	0	0	0	0	0	626-43-7
heptacosafluorotetradecanoic acid (PFTDA)	0	0	0	0	0	0	89.5	0	0	0	0	0	0	0	0	0	0	0	0	629-59-4
Metformin	0	0	0	0	0	0	0	0	0	0	0	9.1	68.8	47.4	50	33.3	0	0	0	657-24-9
Aluminium	0	0	0	0	0	42.9	0	85.7	0	0	0	90.6	92.3	92.4	85.1	80	82.1	83.7	87.9	7429-90-5
Iron	92.7	90	90.9	97.9	77.8	93.5	91.7	91.3	90	92	94.6	91.6	90.3	95	89.4	84.6	83.3	91.9	90	7439-89-6
Lead	9.1	0	0	43.5	0	0	0	0	0	3.8	0	0	0	5.7	0	0	0	0	0	7439-92-1
Manganese	45.1	53.3	50	58.3	58.3	68.7	70.6	50	54.5	64.3	88.4	54.5	3.7	0	58.3	58.3	50	73.1	46.2	7439-96-5
Trichloroacetic acid (TCA)	0	0	0	0	41.2	23.1	68.7	0	0	71.4	54.5	47.4	68.8	76.2	83.9	16.7	41.2	44.4	9.1	76-03-9
Trifluoroacetic acid (TFA)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	60	47.4	76-05-1
Ammonium (NH4)	0	0	31	23.1	0	0	16.9	0	0	16.9	4.6	0	0	0	0	0	0	0	0	92075-50-8
Mecoprop (MCPP)	0	0	0	0	0	0	0	0	47.4	0	0	0	0	0	0	0	0	0	0	93-65-2
2,4-Dichlorophenoxyacetic acid (2,4-D)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	94-75-7
Benzotriazole	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	9.1	95-14-7
Polycyclische aromatische koolwaterstoffen (PAK's)	86.5	56.5	88.6	0	0	91.2	79.2	76.2	72.2	84.4	67.3	45.5	64.8	83.1	0	67.2	8.8	15.3	25.5	
Pesticiden	57.3	74.9	70.6	62.1	72.7	77.9	78.8	74.6	76.8	92.3	67.4	76.2	70	78.5	75.3	72.7	76.2	68.9	69.3	

Nieuwersluis	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	CAS nr.
1,3,5-Trimethylbenzene	0	0	0	0	0	0	0	0	0	0	66.7	0	0	0	0	0	0	0	0	100-41-4
Carbendazim	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	10605-21-7
Aminomethylphosphonic acid (AMPA)	75	0	73.7	88.1	89.4	88.1	88.8	89.1	88	87	85.3	89	85.7	84.8	86.7	85.5	85.1	87.3	87.2	1066-51-9
Glyphosate	33.3	0	60	50	33.3	28.6	28.6	52.4	9.1	9.1	33.3	0	16.7	0	23.1	0	0	9.1	16.7	1071-83-6
Methylbenzene	0	0	0	0	0	0	0	0	0	28.6	60	0	0	0	0	0	0	0	0	108-88-3
Dimethomorph	0	0	0	0	0	0	0	0	0	47.4	0	0	0	0	0	0	0	0	0	110488-70-5
Bis(2-methoxyethyl)ether (Diglyme)	0	0	0	0	0	0	32.9	33.8	52.4	0	0	0	0	0	0	0	0	0	0	111-96-6
Nicosulfuron	0	0	0	0	0	0	0	0	0	0	0	83.3	50	0	0	0	0	0	0	111991-09-4
Bis(2-ethylhexyl)phthalate (DEHP)	0	0	0	0	0	44.4	0	0	0	0	0	0	0	0	0	0	0	0	0	117-81-7
Diatrizoic acid (Amidotrizoic acid)	0	0	0	0	0	0	0	0	16.7	0	0	0	0	0	0	0	0	0	0	117-96-4
Nitrite (NO2)	0	76.6	68.9	57.7	64.2	49.3	52.4	39	36.7	40.5	53.5	40.1	18	37.9	18	10.7	25.9	37.5	45.1	14797-65-0
Bromate	0	0	0	0	0	0	0	0	0	0	0	0	33.3	0	28.6	0	0	0	0	15541-45-4
Methyl-tert.-butylether (MTBE)	0	0	0	51.3	0	0	0	0	0	0	0	9.1	0	0	0	0	0	50.7	0	1634-04-4
Pyrazole	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	31.8	0	0	0	288-13-1
beta-Hexachlorocyclohexane (beta-HCH)	15.3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	319-85-7
Diuron	0	0	0	0	9.1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	330-54-1
Isoproturon	0	0	50	0	0	0	0	31.5	0	0	0	0	0	0	0	0	0	0	0	34123-59-6
Butocarboxim	0	0	0	54.5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	34681-10-2
Butocarboxim-sulfoxide	0	0	0	0	0	0	0	0	0	0	66.7	0	0	0	0	0	0	0	0	34681-24-8
Benzo(a)pyrene	37.5	50	66.7	50	50	50	50	75	0	88.9	75	7.4	74.9	0	0	88.3	52.2	0	19.4	50-32-8
Metolachlor	0	58.3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	51218-45-2
Tetrachloromethane	0	0	0	0	0	23.1	0	0	0	0	0	0	0	0	0	0	0	0	0	56-23-5
Octadecane	0	0	0	0	0	0	69.7	0	0	0	0	0	0	0	0	0	0	0	0	593-45-3
paroxetine	0	0	0	0	0	0	0	0	0	0	0	0	0	0	9.1	0	0	0	0	61869-08-7
Metformin	0	0	0	0	0	0	0	0	0	0	0	16.7	50	50	9.1	0	0	54.5	0	657-24-9
Iopromide	0	0	0	0	0	0	0	0	0	0	0	9.1	0	0	0	0	0	0	0	73334-07-3
Aluminium	0	0	0	0	0	0	0	0	0	0	0	80.4	83.6	74.2	62.6	80.4	67.9	62.5	79.5	7429-90-5
Iron	0	90.9	90.9	91.3	81.8	91.7	85.7	85.7	85.7	84.6	81.8	84.4	85.4	84	79.8	86.7	85.7	73.3	84	7439-89-6
Manganese	0	80.8	73.7	70.6	73.7	75	73.7	75	64.3	61.5	70.6	79.2	47.5	73.1	58.3	72.2	68.8	54.5	65.5	7439-96-5
Dichloromethane	0	0	0	0	0	0	60	0	0	0	0	0	0	0	0	0	0	0	92.9	75-09-2
Trichloroacetic acid (TCA)	0	0	0	0	28.6	28.6	58.3	0	0	64.3	58.3	72.2	87.8	0	0	0	0	0	0	76-03-9
Iomeprol	0	0	0	0	0	0	0	0	0	0	0	23.1	0	0	0	0	0	9.1	9.1	78649-41-9
1-Chloronaphthalene	0	0	0	0	0	28.6	0	0	0	56.5	0	0	0	0	0	0	0	0	0	91-20-3
Ammonium (NH4)	0	71.4	67.5	63.6	71.5	48.7	54	34	22	34	59.8	47.4	26.4	41.5	44	34	26.4	0	0	92075-50-8
Mecoprop (MCPP)	0	0	0	0	28.6	0	0	0	0	0	0	0	0	0	0	0	0	0	0	93-65-2
4-Chloro-2-methylphenoxyacetic acid (MCPA)	0	0	0	0	33.3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	94-74-6
2,4-Dichlorophenoxyacetic acid (2,4-D)	0	0	0	0	0	0	0	0	28.6	0	0	0	0	0	0	0	0	0	0	94-75-7
PAK's (6 van Borneff)	65.2	29.6	61.8	68.2	79.3	70.6	73.8	74.4	70.2	78	76.5	81.7	80.4	58.3	56.2	59.2	62.6	67.6	64.8	
Polycyclische aromatische koolwaterstoffen (PAK's)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	33.4	
Pesticiden	48.5	61.5	76.2	44.4	64.3	58.3	66.7	82.1	0	88.2	83	44.1	79.4	40	30.4	95.2	83.8	25.8	73.2	

Haringvliet	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	CAS nr.
Aminomethylphosphonic acid (AMPA)	75	81.1	63	84.1	87.2	87.7	86.8	88.4	86.7	87.7	88.2	86.5	86.5	85.3	83.9	84.4	83.6	87.3	86.8	1066-51-9
Glyphosate	0	0	73.7	0	65.5	28.6	28.6	0	28.6	0	0	0	0	0	0	0	19.4	0	0	1071-83-6
1,3,5-triazine-2,4,6-triamine (melamin)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	73.7	56.5	63	108-78-1
Bis(2-methoxyethyl)ether (Diglyme)	0	0	0	0	0	0	28.6	6.5	28.6	0	0	0	0	0	0	0	0	0	0	111-96-6
Bis(2-ethylhexyl)phthalate (DEHP)	0	0	0	0	0	0	0	0	0	0	0	0	0	33.3	0	0	0	0	0	117-81-7
Diaminomethylideneurea	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	37.5	56.5	41.2	141-83-3
Nitrite (NO2)	0	69.6	64.3	52.4	62.9	49.3	58.3	13	14.5	13.8	19.4	21.3	15.3	0	0	0	11.5	9.1	44.4	14797-65-0
Bromate	0	0	0	0	16.7	37.5	37.5	9.1	0	37.5	16.7	37.5	33.3	28.6	37.5	33.3	9.1	33.3	0	15541-45-4
Chlortoluron	0	0	0	0	0	0	0	9.1	0	0	0	0	0	0	0	0	0	0	0	15545-48-9
Chloride	0	0	0	44.6	19.4	58.8	48.3	0	0	25	3.8	48.5	0	32.4	0	11.8	0	6.2	0	16887-00-6
Chloridazon	0	0	0	0	0	0	0	28.6	0	0	0	0	0	0	0	0	0	0	0	1698-60-8
Pyrazole	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	48.7	16.7	9.1	0	288-13-1
Isoproturon	0	0	0	0	0	0	0	37.5	9.1	9.1	0	0	0	0	33.3	0	0	0	0	34123-59-6
chloridazon-methyl-desphenyl	0	0	0	0	0	0	0	0	0	0	0	0	0	0	56.5	73	78.3	58.3	33.3	6339-19-1
Metformin	0	0	0	0	0	0	0	0	0	0	0	0	0	33.3	9.1	0	0	0	0	657-24-9
Trichloromethane	0	0	0	0	0	0	0	41.2	0	78.3	0	0	0	0	0	0	0	0	0	67-66-3
Aluminium	0	0	0	0	0	0	0	79.2	0	0	0	78	49.9	73.1	12.7	0	22.5	0	87.2	7429-90-5
Iron	81.5	81.8	94.6	74	77.8	71.8	56.5	60	28.6	13	20	84.8	57.1	83.7	32.9	8.7	41.3	0	90.1	7439-89-6
Manganese	37.5	54.5	0	0	0	33.3	25.4	0	34.2	12.3	0	17.6	0	0	0	0	5.7	0	63.2	7439-96-5
Bromodichloromethane	0	0	0	0	0	0	0	0	0	28.6	0	0	0	0	0	0	0	0	0	75-27-4
Trifluoroacetic acid (TFA)	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	23.1	23.1	76-05-1
2-Nitrophenol	0	0	0	0	0	0	0	0	0	0	0	0	9.1	0	0	0	0	0	0	88-75-5
1-Chloronaphthalene	0	0	0	69.7	0	0	0	0	0	9.1	0	0	0	0	0	0	0	0	0	91-20-3
Ammonium (NH4)	0	30.4	4.6	0	9.1	0	35.5	0	0	24.2	0	0	0	0	0	0	0	0	0	92075-50-8
Polycyclische aromatische koolwaterstoffen (PAK's)	0	0	0	0	0	0	69.7	0	0	0	0	0	0	0	0	0	0	0	12.8	
Pesticiden	0	32.4	23.1	51.5	55	51.9	64.8	73.9	63	67.4	64.1	62.3	58.9	60.4	63	64.9	70.7	65.2	63.3	

Colophon

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