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Prioritization of micropollutants based on removal effort in drinking water purification treatment

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ABSTRACT

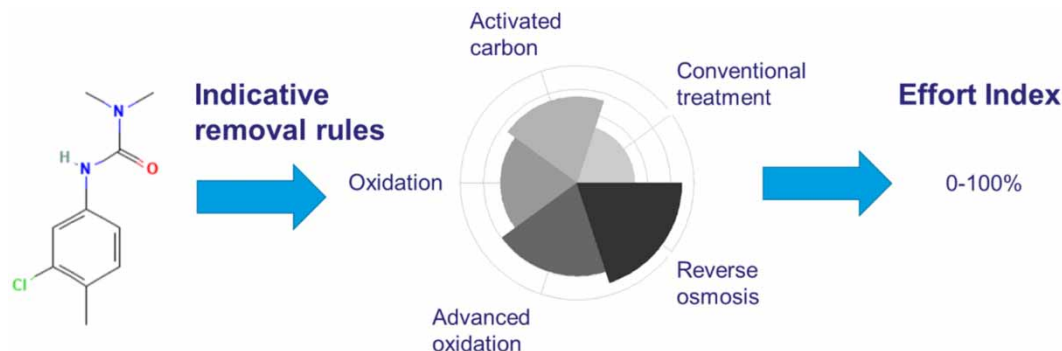
A main focus of water managers with regard to micropollutants is the protection of aquatic ecology. However, micropollutants also have the potential to affect the production of clean drinking water. In this paper, we propose to consider the removal effort when assessing micropollutants with an 'Effort Index' (EI). Assessments using the EI show which micropollutants need more extensive monitoring or abatement because of their difficulty to be removed using low-effort water purification treatment techniques. For water containing mixtures of micropollutants, the averaged EI values can indicate overall water quality. Data on the removal by different purification treatment techniques are not necessarily available for all micropollutants. Therefore, a set of data-driven indicative removal rules is derived to quantify the relation between micropollutant properties and different drinking water treatment techniques. The indicative removal rules provide a rough indication of removability. As an illustration, the water quality of the river Rhine is evaluated between 2000 and 2018. The EI value shows that the Rhine contains increasingly more difficult-to-remove micropollutants. In total, 18 of those are labeled as particularly difficult-to-remove chemicals. These micropollutants are suggested as candidates for abatement to lower the required effort in drinking water production.

Key words: micropollutants, model, prioritization, purification treatment, removal, water quality

HIGHLIGHTS

- The Effort Index (EI) quantifies the removal effort of micropollutants in drinking water treatment.
- The EI can be used to prioritize micropollutants for abatement at the water source.
- The EI is based on the properties of micropollutants.
- Averaged EI values indicate overall water quality for drinking water production.

GRAPHICAL ABSTRACT



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1. INTRODUCTION

The number of micropollutants in the European market is ever-increasing. The 'CL inventory' (ECHA 2021) lists over 200,000 chemical substances that are used in the European Union. An increasing number of these find their way into the freshwater system (van Wezel *et al.* 2017). Many of these micropollutants have the potential to induce adverse effects. Prioritization is a means to focus on the micropollutants that will have the largest effects. For drinking water, Baken *et al.* (2018) prioritized micropollutants in water based on provisional guideline values that are protective for human health. Micropollutant prioritizations with ecotoxicological relevance are widely available. For instance, based on detection frequency, semi-quantitative concentration, bioaccumulation, ecotoxicity, and biodegradability (Liu *et al.* 2022) or based on measured micropollutant concentrations and the predicted no-effects concentrations (Figuère *et al.* 2022).

However, in addition to the effects on the ecological status of the water, micropollutants also pose a potential threat to drinking water production if they cannot be removed easily in purification treatment. So far, it has been difficult to identify which micropollutants will adversely affect the level of purification treatment required. Most often the assessment is based on expert opinions. To facilitate water utilities and water managers in choosing water treatment options, an initial assessment of micropollutants relating to removal efficiency is developed in this paper. This 'Effort Index' (EI) will show which of the many micropollutants need more extensive monitoring or abatement because they will be difficult to remove especially in conventional, low-effort level water treatment installations, requiring less investment in implementation, maintenance, or other costs.

Such a prioritization is useful to water managers since it allows them to take into account the removal effort when determining water quality. According to the European Water Framework Directive (WFD), Article 7.3, all Member States must safeguard water bodies and aim to reduce the level of purification treatment required in the production of drinking water. This means that producing clean drinking water is not only a responsibility for water utilities but also for water managers. A way to assess the removal effort that is needed to produce drinking water will help evaluate whether such goals are met.

A prioritization of micropollutants is also relevant for industry in the *a priori* selection of appropriate chemicals for production and use, especially if these chemicals are expected to be emitted into water. This fits in the 'safe and sustainable by design' framework. In the current production and authorization of chemicals for (industrial) use, the required level of purification treatment for drinking water is not considered. Moreover, in a recent review paper on micropollutants that deserve more global attention, the aspect of the removal effort by drinking water production facilities was overlooked (Yang *et al.* 2022). Schoep & Schriks (2010) concluded earlier, though, that a relation exists between the European Registration, Evaluation, Authorisation, and Restriction of Chemicals (REACH) regulation and the tendency to produce chemicals that are more recalcitrant to purification treatment in the environment, represented by a lower K_{ow} over time. Micropollutants with these characteristics are less likely to bioaccumulate in the environment which was one of the goals of REACH. While these micropollutants are seen as 'harmless', they are of increasing concern for drinking water utilities as they will be more 'mobile' and difficult to remove from the water.

For an evaluation of the overall water quality for drinking water production, the EI values of all individually monitored micropollutants in the water can be averaged. Hence, waters that contain micropollutants that can only be removed using more advanced purification treatment techniques get a less favorable score.

Unfortunately, data on micropollutant removal efficiency by treatment techniques of different effort levels are often not available or are incomplete. Assigning values for removal for all potential micropollutants that occur at water intake points is expensive and not realistic at this moment. For micropollutants with unknown removal values, models provide an approximation. To link the removal effort of individual micropollutants to purification techniques, a relationship between these two needs to be established.

The removal efficiency of any micropollutant depends on the type of purification treatment technique and the exact process conditions of the technique (Fischer *et al.* 2019). In addition, the efficiency of a purification treatment technique under specific conditions depends on the (combination of) properties of the micropollutant (Fischer *et al.* 2019). Many different properties that influence removal efficiency are mentioned in scientific literature works, such as molecular charge (e.g. Kovalova *et al.* 2013; Borowska *et al.* 2016), K_{ow} (e.g. Verliefe *et al.* 2008; Kovalova *et al.* 2013), molecular weight (e.g. Bellona *et al.* 2004; Mailler *et al.* 2014), and functional groups (e.g. Sonntag & von Gunten 2012; Mailler *et al.* 2014). Although existing literature links micropollutant properties to a degree of removal in different treatment techniques, this is in many cases either a qualitative link that is not data-driven or a relatively extensive model. An example of qualitative links is classes of removal effort based on biodegradability and K_{ow} (e.g. Fischer *et al.* 2011), and logical assumptions

regarding the relation between micropollutant properties and removal efficiency such as Gibbs free energy and K_{ow} (van den Doel *et al.* 2023). An example of a relatively extensive model is one that also takes specific treatment conditions into account (e.g. de Ridder *et al.* 2010; Vries *et al.* 2013, 2017). In such models, the ranking of micropollutants is dependent on the modeled conditions in a particular treatment, and expected removal is hence calculated for a specific installation with specific settings. A more generic evaluation of expected removal effort is more practical and desired by water managers but can be rather complicated with such models.

Van den Doel *et al.* (2023) derived such a data-driven formula for treatment consisting of clarification, disinfection, and granular activated carbon (GAC) filtration using logistic regression. Here, removability was explained by a combination of biodegradability half-life, melting point, and octanol–air partition coefficient (K_{oa}). To estimate relations between a wider range of different purification techniques and micropollutant properties we derive simple indicative removal rules for the removal efficiency of micropollutants for several treatment techniques representing different levels of effort. These are conventional treatment (coagulation, flocculation), oxidation (with ozone), advanced oxidation (H_2O_2 and ozone or UV), filtration over activated carbon, and reverse osmosis (RO). The data-driven indicative removal rules are derived analogously to quantitative structure–activity relationship (QSAR) models. The rules enable a rough calculated removal efficiency of micropollutants with a simple formula and provide an alternative for estimating the removal efficiency if data on removal with specific water treatment techniques are not available.

2. METHODS

2.1. Calculation of the EI

The EI is a weighted average of the expected removal effort in treatment techniques of different levels of effort (from conventional, to additional, to advanced). The levels of effort are explained in Table 1. The following formula (Equation (1)) expresses the overall removal effort per micropollutant in a single value.

$$EI_s = 100 - \frac{(P_{conv} * 3 + P_{add} * 2 + P_{adv} * 1)}{6} \quad (1)$$

EI_s in Equation (1) stands for EI (substance) and represents the overall purification effort of a micropollutant. P_{conv} is the average (calculated) removal in ‘conventional’ removal techniques, P_{add} is the average (calculated) ‘additional’ removal, P_{adv} is the average (calculated) ‘advanced’ removal. Weights determine how much removal in a treatment level (Table 1) contributes to EI_s . By subtracting the weighted removal from 100, the purification effort is low in case a micropollutant is easily removed and high in case it is difficult to remove. ‘Effort’ (see Table 1) is not defined very precisely. Techniques are classified

Table 1 | Overview of techniques for purification treatment, from top to bottom with increasing effort (‘level of purification treatment’)

Class of effort (‘level of purification treatment’)	Technique	Remark
Low effort: Conventional	Coagulation/Flocculation (sedimentation) with rapid filtration	For surface water, various flocculants can be added here. For groundwater this is not necessary.
Medium effort: Additional (one of these techniques)	Oxidation (Ozone)	This follows conventional. After oxidation, often activated carbon is applied.
	Membrane filtration (ultra or nano) Activated carbon (powder or granular)	This follows conventional. This follows conventional. Micropollutants can be displaced over time by other, better adsorbing micropollutants. With granular carbon, the contact time is important for removal efficiency (Snyder <i>et al.</i> 2007; Mailler <i>et al.</i> 2016). Furthermore, the time to reactivation plays an important role. Dose is important for powdered carbon (e.g. Snyder <i>et al.</i> 2007; Mailler <i>et al.</i> 2014).
High effort: Advanced (one of these techniques)	Reverse osmosis (RO)	This follows conventional instead of ‘additional’.
	Advanced oxidation (AOP) (H_2O_2 and Ozone or UV)	This follows conventional instead of ‘additional’. After oxidation, activated carbon or another filtration process is applied.

from ‘conventional’ to ‘advanced’ based on the required (expected) investment in implementation, maintenance, or expected carbon footprint or energy consumption. Removal by treatment techniques representing the least effort, conventional, is assigned the highest weight value ‘3’, removal by additional techniques will be assigned ‘2’, and removal by advanced techniques is assigned the lowest weight ‘1’. The height of the weights, 1–3, were chosen to weigh up treatment techniques of low effort. Now, if a micropollutant is easily removed by techniques of low effort, this gives a favorable EI_s and the micropollutant will be of lower priority. The fact that treatment techniques of all effort levels are represented in Equation (1) does not imply that it is expected that water production sites implement all these techniques. Rather, the EI represents the theoretical removability of the micropollutant with a wide range of techniques.

Table 1 gives an overview of purification techniques commonly applied in Dutch water utilities and their classification in levels of increasing required effort. To remove micropollutants from water, different purification treatment techniques are most often applied in sequence.

To evaluate overall water quality for drinking water production, all values per micropollutant (EI_s) from Equation (1) are combined into a single measure. Water that contains micropollutants that have a low purification effort (easy removal) will indicate a good quality for drinking water production. Equation (2) gives the calculation for the overall water quality score.

$$EI_w = \frac{\sum^n EI_s}{n} \quad (2)$$

Here, EI_w is the purification EI (water body), ‘ n ’ indicates micropollutants.

2.2. Derivation of the indicative removal rules

Figure 1 shows the steps in deriving indicative removal rules for predicting removal based on micropollutant properties. In the first step, substance properties are collected for (organic) micropollutants with data available on removal efficiency (step 1, Figure 1).

The micropollutant properties are collected from a variety of sources. Some properties are predicted with the OPERA models that can be collected very easily by downloading these from the online ‘Chemistry Dashboard’ or the OPERA user interface (Mansouri *et al.* 2018). Other properties are added from Open Babel (O’Boyle *et al.* 2011), or calculated by PubChem. These properties can be downloaded via functions in the open-source statistical free software environment ‘R’ with the ChemmineR package (Cao *et al.* 2008). Furthermore, properties predicted by models in EPISuite (US EPA 2012) are easy to collect by downloading the EPISuite software and uploading a list of CAS numbers. In total, 79 (predicted) micropollutant properties are collected for each organic micropollutant (Supplementary material, Supplement II). For each of these properties, it is checked whether the logarithm of the property shows a better normal distribution than its original value. If this is the case, the natural log of the property is taken as input value. If a property is already expressed as \log_{10} , this is maintained.

The second step is to uncover properties associated with measured removal efficiencies using a ‘Random Forest’ regression (Breiman 2001) (step 2, Figure 1). Most often, machine learning techniques use advanced statistical models to ‘learn’ and

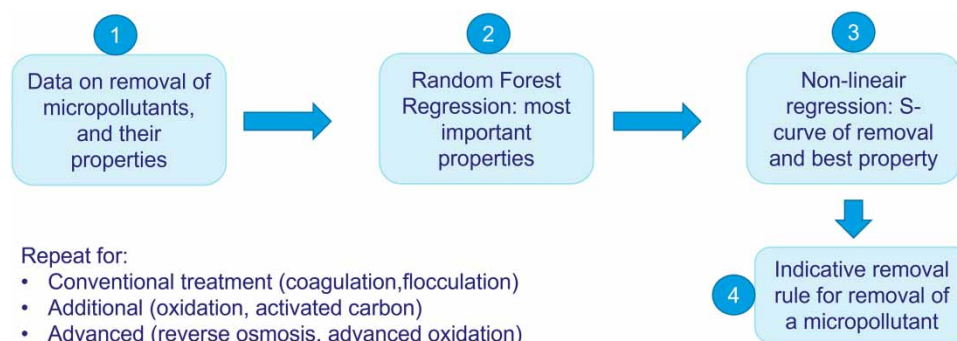


Figure 1 | Steps to arrive at an indicative removal rule based on micropollutant (substance) properties.

make a good prediction model. Random Forest models create random decision trees. The algorithm will select the best explanatory properties from all micropollutants by prioritizing decision trees with properties that best explain removal efficiency. This results in scores for ‘importance’ of the property. The highest correlated properties (correlation > 0.97) are removed beforehand. Properties that have a standard deviation of zero (they do not vary between the different micropollutants) are also removed beforehand. Supplementary material, Tables S1–S6 show examples of properties that were found to be important in relation to removal.

After determining the most important properties, the third step is to establish a relation between the most important property and the removal efficiency (step 3, Figure 1). For an indicative removal rule, an S-shaped relation is a workable solution to achieve a calculated removal between 0 and 100%. A best-fit S-curve through the data was established using nonlinear regression modeling, using the ‘minpack.lm’ package (Elzhov *et al.* 2016) in the ‘R’ free software environment. Equation (3) is the equation for an S-curve.

$$P = \frac{C}{1 + e^{A \cdot (\text{Property value} - B)}} \quad (3)$$

Here, P is purification efficiency (or removal). C is the maximum in a range, B is the center of the S-curve, and A is a measure of steepness. ‘Property value’ is the value of the selected (most important) associated micropollutant property (see step 2).

The nonlinear model will generate statistics to evaluate if the S-curve itself could be established to a significant extent. To examine whether the S-curve also associates with actual removal (step 4 in Figure 1) the fit between calculated values in the indicative removal rule and the measured removal values is calculated, with the coefficient of determination R^2 representing the explained variance.

2.3. Data used for removal of substances

In our case study for deriving indicative removal rules, we use removal efficiency values that are generated by drinking water utilities in the Netherlands, under different circumstances in treatment techniques over the course of 14 years. This makes this suitable data to define generic rules that likely do not only apply to treatments under a single condition. The data are used in recurring studies by the drinking water utilities on ‘robustness’ of treatment techniques in removing diverse, relevant micropollutants. This means that the data are known and have been evaluated in these studies. In Table 2 an overview is presented of the dataset in this study.

For conventional treatment with ferric chloride (FeCl_3) as a coagulant, a large dataset (1,132 measurements) of one drinking water company is available containing data from 2013 onwards. Most compounds were measured relatively frequently, more than eight times. These compounds were selected because multiple measurements give a better average view of removal efficiency related to a compound property. This resulted in a slightly smaller dataset of 988 measurements for 29 micropollutants. The reported pH ranges between 7.9 and 8.4 and the reported coagulant dose ranges between 2.5 and 3.5 mg/L (depending on season; personal communication). Occasionally a negative removal is measured, which may be caused by experimental uncertainties, as concentrations come close to the detection limit, or because metabolites may be transformed back to mother compounds. Micropollutants with occasional negative removal are kept in the dataset. All measurement points that have negative removal are set to zero.

Table 2 | Dataset on removal efficiencies of micropollutants (MPs), measured in different purification treatment processes by drinking water utilities

	Utilities	Conditions	Tested MPs	Measurements	Measurements per MP
Conventional	1	1	29 ^a	988	9–69
Oxidation	1	1	75	128	1–8
Activated carbon	3	8	76	323	1–16
Advanced oxidation	2	3	76	130	1–8
Reverse osmosis	1	2	83	137	1–8

^aOnly compounds with more than eight individual measurements in the dataset, this selection was possible because of the size of the dataset (1,132 measurements).

Data for activated carbon consists of treatments with both GAC and powdered activated carbon (PAC) and data are available from three companies. Varying conditions are exposure time, reactivated carbon, seasonal variations, and stand-alone or after oxidation treatment. Again, several micropollutants have negative removal values. In activated carbon, micropollutants that adhere less well are displaced at a certain point by other compounds that adhere better to activated carbon (both micropollutants and natural organic matter). Initially, these displaced micropollutants will have been well adsorbed. This could provide an additional challenge for determining a rule to reflect removal with activated carbon. Since PAC and GAC have many similarities, it is expected that a single property in an indicative removal rule for activated carbon could apply to both types of applications.

For oxidation (treatment with O₃, ozone), data are available from one company and no varying conditions are specified. The measurements were done over a period of several years.

Advanced oxidation processes (AOPs) are processes in which hydroxyl radicals are formed, which can react very effectively with a wide range of compounds. Examples of AOP include UV in combination with O₃ or H₂O₂, or the combination O₃/H₂O₂. Data for AOP are available in the dataset for two companies. The conditions that vary are combining O₃, H₂O₂, and UV or only UV and H₂O₂, and seasonal variations. In practice, AOP is not applied without additional filtration with activated carbon or, for example, dune filtration, to remove the excess H₂O₂, transformation products, and formed AOC (assimilable organic carbon). The latter is to maintain/improve the biological stability of the water.

For 'advanced' treatment, RO data are available from one company in different seasons, over several years.

3. RESULTS

3.1. Indicative removal rules

For all purification treatments in the dataset of the Dutch treatment facilities, conventional (coagulation, flocculation), oxidation (with ozone), advanced oxidation (UV and H₂O₂ and/or ozone), activated carbon, and RO, steps 1–4 as given in Figure 1 were executed. Per treatment, the most important micropollutant property was linked to removal efficiency via S-shaped indicative removal rules. For all S-curves, parameters A, B, and C (see Equation (3)) could be significantly established (see Supplementary material, Supplement I). This means the data fit well enough in an S-shaped curve.

$$P_{\text{conv}} = \frac{98}{1 + e^{-1.2 \cdot (\log K_{\text{ow}} - 2.4)}} \quad (4)$$

$$P_{\text{ac}} = \frac{73.4}{1 + e^{-1.99 \cdot (\log K_{\text{oa}} - 3.8)}} \quad (5)$$

$$P_{\text{ox}} = \frac{83.5}{1 + e^{-0.39 \cdot (\text{abonds} - 2.71)}} \quad (6)$$

$$P_{\text{advox}} = \frac{82.6}{1 + e^{-4.4 \cdot (\text{FeatureRingCount3D} - 0.26)}} \quad (7)$$

$$P_{\text{ro}} = \frac{100}{1 + e^{-2.95 \cdot (\ln(\text{Mass}) - 4.63)}} \quad (8)$$

P_{conv} is the purification efficiency (%) for conventional treatment. P_{ac} for activated carbon treatment, P_{ox} for oxidation, P_{advox} for advanced oxidation, P_{ro} for RO (see Table 1). Equations (4)–(8) can be used to calculate EI_s and EI_w in Equations (1) and (2). Most important substance properties for Equations (4)–(8) were, respectively, $\log K_{\text{ow}}$, the logarithm of the n-octanol–water partition coefficient. $\log K_{\text{oa}}$ (the logarithm of the octanol–air partition coefficient, abonds (the number of aromatic bonds of the micropollutant), FeatureRingCount3D (the number of rings), LnMass (the natural logarithm of the average molecule mass). In addition, HBA1 (number of H bond acceptors; determines the number of H-bridges) increased the predictability for P_{advox} (see Supplementary material, Supplement I). To keep the rules as simple as possible, this substance property was not included.

In Table 3, an overview is given of the accuracy of the derived S-shaped indicative removal rules for calculating removal of organic micropollutants. The derivation of these rules is explained in more detail in Supplementary material, Supplement I.

The RF regression gives, in general, a good result with respect to the variation explained, generally better than the S-shaped curves (Table 3). A disadvantage of RF machine learning models is that these are relatively complicated statistical models.

Table 3 | Overview of purification treatment techniques and S-shaped derived indicative removal rules

Treatment effort level	Technique	Random Forest accuracy (variance explained)	Most important substance property	S-curve property vs. measured removal, R^2 (variance explained)
Low effort: Conventional	Coagulation, flocculation	0.78	$\log K_{ow}$	0.56
Medium effort: Additional	Activated carbon	0.12 (with conditions: 0.56)	$\log K_{oa}$	0.17
Medium effort: Additional	Oxidation	0.72	Aromatic bonds	0.41
High effort: Advanced	Advanced oxidation	0.73	Ring count 3D	0.59
High effort: Advanced	Reverse osmosis	0.57	InavMass	0.48

See Table 1 for more explanation on the purification treatment techniques and effort.

Consequently, these models are more challenging to use or implement. Therefore, accuracy is sacrificed for using the simpler S-shaped indicative removal rules as given in Equations (4)–(8).

The spread in actual treatment performance in the dataset is wide (see Supplementary material, Supplement I). The large spread, even within single substances, is likely caused by difficulties in measuring near detection limits or exactly match influent to effluent concentrations in full scale drinking water treatment plants, in combination with variations in other circumstances during water treatment. This doesn't mean, however, that general treatment performance cannot be estimated. Measurements likely vary around 'realistic' treatment performance values. In some cases, the indicative removal rule does not explain the majority of the variation but the model itself is significant. This combination means that the predictor variable still provides information about the response even though the data points fall further from the regression line. The predictions will just not be very precise (in other words, will have a high prediction interval). The aptness of the indicative removal rules to indicate the removability of substances is further validated in the following Sections 3.2 and 3.3.

The R^2 for activated carbon is the lowest of all (Table 3). If the factor 'condition', which reflects the different conditions in the different utilities, is included in the fit (PAC/GAC, contact time, freshness, season, applied after oxidation or not), the explained variance increases to 0.56. This means in this dataset for activated carbon that the conditions determine the removal to a larger extent than micropollutant properties. Therefore, the indicative removal rule for activated carbon can best be rederived in the future in a new attempt with another dataset.

3.2. Accuracy of the indicative removal rules

Although the indicative removal rules represent significant relations between micropollutant properties and removal efficiencies in a treatment technique (see Supplementary material, Supplement I), the R^2 indicates that they are not very accurate. Therefore, these indicative removal rules can only give rough indications. As a verification, Table 4 provides an overview of measured purification efficiencies for several micropollutants as assembled in van der Aa *et al.* (2017). We use broad categories of removal for the comparison because the indicative removal rules are not very precise. A selection was made of micropollutants that were reported to exceed their target or signaling value in the Dutch Drinking Water Decree (2023) at the water abstraction location Nieuwegein in the dataset of RIWA-Rijn. These are compared to the calculated removal efficiencies from the indicative removal rules in Equations (4)–(8).

From Table 4, it can be tentatively concluded that individual indicative removal rules are in the correct range in 68% of the calculations (38 out of 56), nearly correct in 28% of the calculations (16 out of 56), and wrong in 4% of the calculations (2 out of 56). It needs to be emphasized that the literature data collected by van der Aa *et al.* (2017) are Dutch studies, and the indicative removal rules were also trained on data from Dutch water utilities. This means for further generalization, verification with data from other countries is needed.

Two predicted values do not match the literature data. The lack of aromatic rings in glyphosate caused a low predicted removal by advanced oxidation (Table 4). For glyphosate, however, N-bonds are broken during oxidation (Espinoza-Montero *et al.* 2020). Measured removal rates in (advanced) oxidation indeed indicated higher removal. This means the indicative

Table 4 | Purification efficiency ranges from the literature (L*) compared to calculated values with the indicative removal rules (IR, in %)

	Conv		AC		Ox		AdvOx		RO	
	L*	IR	L*	IR	L*	IR	L*	IR	L*	IR
Diglyme	0-40	3	20-60	42	0-40	22	40-60	20	40-60	69
Chlortoluron	0-40	49	60-90	73	80-100	65	80-100	79	80-100	90
AMPA	0-40	0	60-90	73	20-60	22			40-60	56
Benzo[a]pyrene	80-100	97	80-100	73	40-60	83	60-90	82	80-100	93
Glyphosate	0-40	0	40-60	73	40-60	22	80-100	20	80-100	81
MTBE	0-40	15	20-60	7	80-100	22	40-60	20	60-90	39
Atrazine			40-60	73					80-100	90
Diuron	0-40	57	60-90	73	60-90	65	80-100	79	60-90	92
Isoproturon	0-40	57	80-100	73	80-100	65	80-100	79	80-100	89
Metolachlor	0-40	69	60-90	73	80-100	65	80-100	79	80-100	95
Metformine			0-40	8					80-100	66
Mecoprop			40-60	73	60-90	65	60-90	79	80-100	90
2,4-D			60-90	73	60-90	65	60-90	79	80-100	91

'Conv' is conventional treatment, 'AC' is activated carbon treatment, 'Ox' is oxidation treatment, 'AdvOx' is advanced oxidation treatment, 'RO' is reverse osmosis treatment. Green, or medium grey, indicates good concordance, yellow, or light grey, indicates one class difference, red, or dark grey, indicates more than one class difference (within the class range* in the literature ± 1%). *Literature removal ranges are as follows: - indicates removal 0-40%; 0 indicates removal 40-80%; + indicates removal 80-100%; empty indicates no data (van der Aa et al. 2017). '-/0' was interpreted as removal between 20 and 60%, '0/+' was interpreted as removal between 60 and 90%.

removal rule for oxidation misses a relevant property of glyphosate. Methyl-tert-butylether (MTBE) has a high measured removal in RO but a low predicted removal according to the indicative removal rule for RO. This low removal fits such a small, uncharged molecule. Some authors studied RO as a pretreatment for the removal of MTBE, before further oxidation of the compound (Cooper et al. 2002; Li et al. 2008). This indicates that in practice it is difficult to remove MTBE by means of RO and the indicative removal rule can be correct for MTBE.

As an example, a possible visualization of the calculated purification efficiency is given in Figure 2. The figure shows calculated removal efficiencies for two micropollutants and per treatment based on the indicative removal rules (see also Table 4).

3.3. Accuracy of the EI value

The indicative removal rules are meant to be combined to quantify the EI_s (Equation (1)) for micropollutants of interest without (literature) data on removal efficiency. When the removal values as calculated by the indicative removal rules (Equations (4)-(8)) are integrated, less accurate results of Equations (4)-(8) will expectedly be leveled out.

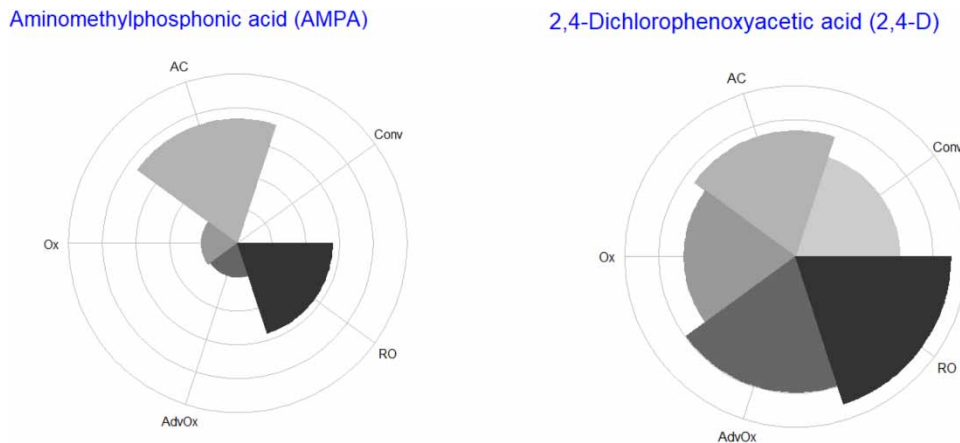


Figure 2 | Example visualizations of the indicative removal rules for calculating the purification efficiency per treatment technique, for two micropollutants. The outer ring is 100% calculated removal, the center is 0% removal. 'Conv' is conventional treatment, 'AC' is activated carbon treatment, 'Ox' is oxidation, 'AdvOx' is advanced oxidation, 'RO' is reverse osmosis (see Table 1).

As a case study, 68 relevant micropollutants were selected from the RIWA-Rijn database, based on their exceedance of target or signaling values in the Dutch Drinking water decree. Supplementary material, Supplement II contains the calculated EI for these micropollutants. Of these 68 micropollutants, 6 are categorized as 'easy to remove' with an EI of 20 or less, while 46 are categorized as 'medium to remove' with an EI between 20 and 60. The 18 remaining micropollutants obtained a value higher than 60 which means that expected removal efficiency is low (see Supplementary material, Supplement II for corresponding removal percentages).

Micropollutants with high EI_s are indicated as hard-to-remove substances and therefore are likely to end up in drinking water ready for delivery. We use data from the Dutch water utilities between 2009 and 2018 (REWAB database, not public, data available upon request) to check if micropollutants with a high EI_s are indeed detected in water after purification treatment more often than micropollutants with low EI_s . REWAB contains yearly maximum concentrations of micropollutants per treatment facility. If this value in REWAB data is recorded above the detection limit of the measurement method, we labeled the micropollutant as 'detected'.

Table 5 shows micropollutants with respectively the highest and lowest EI_s (for all micropollutants, see Supplementary material, Supplement II). Of the 18 micropollutants with high EI_s , 14 (82%) were indeed detected in water after purification treatment, one was not reported. In comparison, only six (35%) micropollutants with relatively low EI_s were detected.

Overall, results in Table 5 indicate that micropollutants with high EI_s are indeed found more often in water after purification treatment than with low EI_s . Nevertheless, there are exceptions where micropollutants with high EI_s are not detected in water after purification treatment. Also, micropollutants with low EI_s are, in some cases, detected.

N-bonds are broken during oxidation and this also applies to oxamyl and butocarboxim. This is not incorporated in the indicative removal rule for oxidation. That can be a reason why these micropollutants were not detected in water after purification treatment although their EI_s were high (Table 5). Another example, Tetrachloromethane, was also not detected but had high EI_s . This compound is rather volatile and could have been removed from water during aeration processes.

For other substances where indicative removal rules do not match the expected detection in treated water, the EI_s do correspond to evidence from the literature. Ethyl-tert-butylether (ETBE) has high EI_s but was nevertheless not detected in drinking water after treatment. ETBE is a small, uncharged, compact (branched) molecule, which will be poorly removed via coagulation/ flocculation/ sedimentation. Many C-C interactions are not possible, so the molecule will also not adsorb very well on activated carbon. Because there are no double bonds, it will not absorb UV radiation. This is consistent with data in the literature (Baus *et al.* 2007) although some removal is possible (Inal *et al.* 2009). Other micropollutants were detected in treated water even if the calculated EI_s was low. It is unclear why these were detected. Mecoprop, naphthalene, paroxetine, tiamulin, and benzo(a)pyrene can be removed by coagulation/flocculation because of their high K_{ow} value, oxidized because of their ring structures, and removed by RO because of their sizes. It could be that starting concentrations before purification treatment were high, these data were not available.

3.4. Water quality score

To showcase the application of the Effort Index for waterbodies (EI_w), Equation (2) is applied to micropollutants exceeding the target or signaling values in the Dutch Drinking Water Decree at locations along the river Rhine in the Netherlands. This data is obtained from RIWA-Rijn (available upon request at RIWA-Rijn). Example programming code to apply Equations (4)–(8) and Equations (1) and (2) to micropollutants in locations of the river Rhine is provided in the data package that is available at <https://doi.org/10.5281/zenodo.5982001>. Figure 3 shows that for locations Nieuwegein, Andijk and Nieuwersluis the average EI_w increased between 2000 and 2018. This is mainly caused by the relative increase of micropollutants with a low $\log K_{ow}$, which are relatively difficult to remove by conventional treatment. For Lobith, the index is decreasing. This is in accordance with previous calculations by Pronk *et al.* 2021. In that paper, a more simple (non-data-driven) index was used, based on biodegradation and $\log K_{ow}$. With respect to the calculations in Pronk *et al.* (2021) the range in the EI_w value is larger, 13–75 vs. 26–69, which means a bigger distinction can be made in water quality with the EI_w .

The EI_w ranges from 13 (at location Nieuwersluis in 2016) to 75 (Lobith, Andijk, several years). The EI_w varies more from year to year if only a few substances are included. This is mainly the case in location Andijk where only a few micropollutants exceed their target or signaling value from the Dutch Drinking Water Decree. The low EI_w in Nieuwersluis in 2016 is also due to a single micropollutant (benzo(a)pyrene) that is easily removed according to its EI_s .

Table 5 | Prioritization of micropollutants detected in the river Rhine, based on their Effort Index (Equation (1))

CAS number	Name	Effort Index	Detected	fraction	%
1634-04-4	Methyl-tert-butylether (MTBE)	82	Yes	88/1627	5.4
76-05-1	Trifluoroacetic acid (TFA)	82	Yes	7/7	100
75-09-2	Dichloromethane	82	Yes	24/1668	1.4
107-06-2	1,2-Dichloroethane	78	Yes	10/1669	0.5
637-92-3	Ethyl-tert-butylether (ETBE)	76	No	0/823	0
111-96-6	Bis(2-methoxyethyl)ether (Diglyme)	75	Yes	64/491	13
67-66-3	Trichloromethane	72	Yes	147/1669	8.8
61-82-5	Amitrole	72	-	-	-
1066-51-9	Aminomethylphosphonic acid (AMPA)	70	Yes	87/755	11.5
75-27-4	Bromodichloromethane	69	Yes	55/1668	3.3
108-78-1	1,3,5-triazine-2,4,6-triamine (melamin)	69	Yes	21/24	87.5
1071-83-6	Glyphosate	68	Yes	12/755	1.6
657-24-9	Metformin	67	Yes	24/71	33.8
288-13-1	Pyrazole	66	Yes	24/29	82.8
23135-22-0	Oxamyl	66	No	0/921	0
76-03-9	Trichloroacetic acid (TCA)	63	Yes	4/70	5.7
56-23-5	Tetrachloromethane	62	Yes	1/1669	0
34681-10-2	Butocarboxim	61	No	0/930	0
626-43-7	3,5-Dichloroaniline	30	No	0/297	0
94-75-7	2,4-Dichlorophenoxyacetic acid (2,4-D)	30	No	0/1178	0
55297-95-5	Tiamulin	29	Yes	22/497	4.4
298-00-0	Parathion-methyl	29	Yes	1/1175	0
18691-97-9	Methabenzthiazuron	28	No	0/1102	0
5611-51-8	Triamcinolonehexacetonide	28	-	-	-
93-65-2	Mecoprop (MCP)	27	Yes	22/1178	1.9
51218-45-2	Metolachlor	27	No	0/1180	0
94-74-6	4-Chloro-2-methylphenoxyacetic acid (MCPA)	27	No	0/1178	0
110488-70-5	Dimethomorph	27	No	0/69	0
91-20-3	Naphthalene	26	Yes	31/1669	1.9
61869-08-7	Paroxetine	21	Yes	9/39	23.1
126833-17-8	Fenhexamid	20	No	0/47	0
126535-15-7	Triflurosulfuron-methyl	19	No	0/62	0
84-74-2	Di-n-butylphthalate (DBPH)	19	No	0/85	0
87-86-5	Pentachlorophenol	18	No	0/242	0
117-81-7	Bis(2-ethylhexyl)phthalate (DEHP)	17	No	0/73	0
50-32-8	Benzo(a)pyrene	13	Yes	3/1667	0.2

See Supplementary material, Supplement II for more information. The column 'Detected' indicates when the micropollutant is detected in water after purification treatment between 2009 and 2018. Fraction is the number of detections in locations with yearly reports and the total number of locations with yearly reports. Detection is often possible at very low (trace) concentrations well-below health-based guideline values (not shown).

4. DISCUSSION

Micropollutants that are present in natural waterbodies are often considered with regard to their risks to aquatic ecology. However, micropollutants also have the potential to affect the production of clean drinking water. In this paper, we suggest

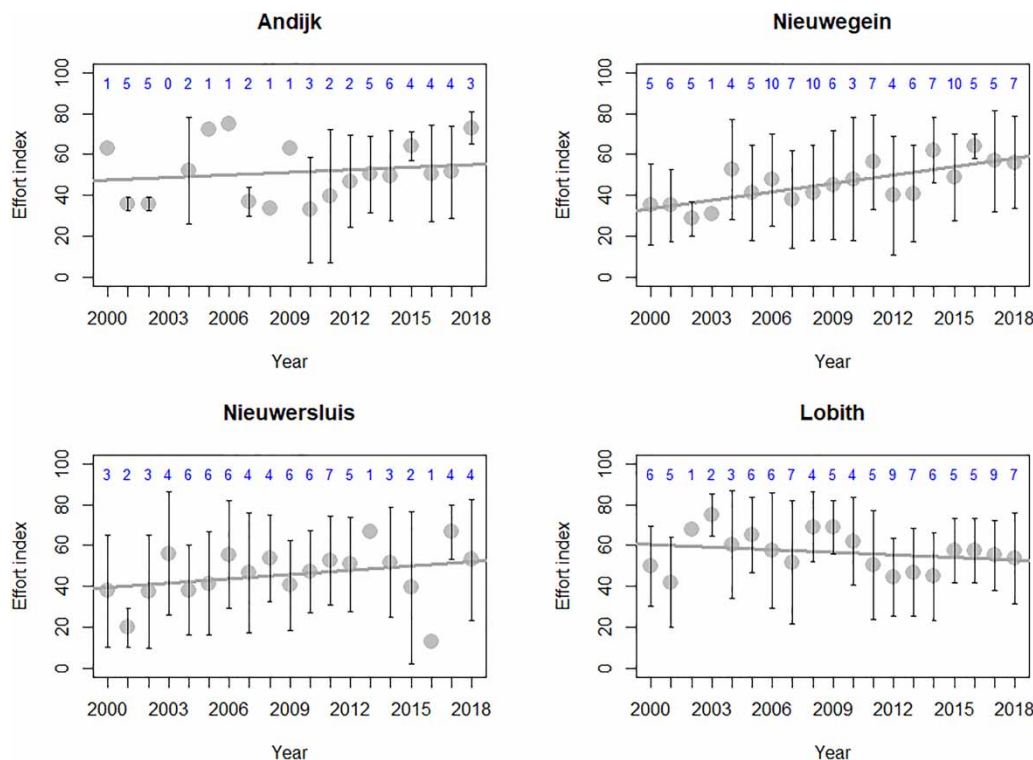


Figure 3 | The EI for locations along the river Rhine, the Netherlands (data source: RIWA-Rijn). Per year, only micropollutants that exceeded their target or signaling value in the Dutch drinking water decree are considered. Their number is indicated above each year.

to take the level of required purification into account when assessing the effects of micropollutants. For this purpose, an effort index (EI_s) per micropollutant is proposed, based on ease of removal. The EI_s can be used to assess which of the many micropollutants may need more extensive monitoring or abatement because of their difficulty to be removed especially in low effort level treatment techniques for drinking water. For this purpose, more weight is assigned to conventional treatment in the formula to calculate EI_s , with a lower weight for additional and lowest for advanced. These weights can be adjusted based on differences in actual required (expected) investment in implementation, maintenance, energy consumptions or carbon footprint for 'conventional' compared to 'additional' and 'advanced' treatment techniques.

The EI_s can be applied by the industry as a first-tier selection of difficult-to-remove micropollutants if these micropollutants are expected to be emitted into water. The EI_s can also assist water quality managers by addressing potential 'problem' micropollutants that are hard to remove especially in low effort level treatment techniques for drinking water. The EI_s per micropollutant can be combined into a single water quality value, EI_w . This value can be used to facilitate decisions to decrease the level of purification treatment required. This helps to achieve the requirement of the WFD to manage surface waters to reduce the level of effort in the production of drinking water.

The weights in the formula that emphasize the importance of removal in low effort level treatment techniques for drinking water are at this moment set quite arbitrarily. This can be adjusted in the future, depending on the application. As an example, costs can be used to assign weights. Costs per m^3 treated water mentioned in De Waal & Hofman-Caris (2021) would indicate that a factor 4 would be appropriate to weigh between 'additional' and 'advanced'. More research is needed to come to support integral weights for the efforts associated with the treatment techniques.

As with all effects of micropollutants, EI_s only implies a risk in case of actual exposure. Hence, in case a micropollutant with a high EI_s is present in concentrations below the drinking water standard, removal will not be necessary. In that case, the micropollutant will have little effect on the actual required purification treatment effort (Pronk *et al.* 2021). For a final prioritization of micropollutants the EI_s can be combined with water quality indices for drinking water production

that use measured concentration and ease of removability as a basis (Pronk *et al.* 2021; van den Doel *et al.* 2023). In addition, for a final prioritization, health-based guideline values should be taken into account (Baken *et al.* 2018).

Calculation of the EI_s relies on data of removal efficiencies of micropollutants by treatment techniques of different levels of effort, from conventional to advanced. However, these data are not always available. Modeling is an alternative way to link removability to individual micropollutants that lack measurement data. In this paper, a framework is proposed to derive indicative removal rules to describe the relation between a micropollutant property and removal efficiency in a particular purification treatment. We used removal data of micropollutants that were analyzed by different Dutch drinking water companies under different conditions and over a timespan of several years. Because of the large timespan and conditions, there is a large range in removal efficiencies for each specific micropollutant. This means that, by definition, the indicative removal rules will show considerable noise. This proved especially true for purification treatment with activated carbon. The different conditions used in this treatment technique explained most of the variance in this study, instead of a micropollutant property.

The advantage of linking a micropollutant property to a generic value for removal with the indicative removal rules is that there is no need to consider (implicit) settings of treatment installations. The disadvantage is that verification of calculated removal values is difficult. Actual removal data are variable because specific settings in treatment facilities can change removal efficiency for a micropollutant. Verification showed that within a broad range, the calculated EI_s fit measured removal values correct (68%) or nearly correct (28%). Another verification with data on the detection of micropollutants in treated water showed that the EI_s , calculated with the indicative removal rules could prioritize hard-to-remove substances. Of the highest prioritized, difficult-to-remove micropollutants, 82% were detected at some point in water after purification treatment. Of the lowest prioritized micropollutants, only 32% were detected at some point. This means that micropollutants that are prioritized with the EI are indeed often encountered in drinking water after treatment.

Several substances, however, deviated from what was expected from their EI_s . This is because the simplicity of the rules inherently holds some caveats. The link between micropollutants and removal efficiency based on one property will not apply equally well to every micropollutant. The deviating substances pointed to improvements in the indicative removal rules. In a future refinement of the indicative removal rule for (advanced) oxidation, the presence of N-bonds should be added. This will be valid for micropollutants that have N-bonds that can be oxidized (Espinoza-Montero *et al.* 2020). No rule was derived specifically for aeration processes, whereas this can be a cause for easy removal of substances. This could be an added rule in the future. Also, PFAS (Per- and Polyfluorinated Substances) do not adhere to the indicative removal rule for conventional treatment (Xiao *et al.* 2013) possibly because their K_{ow} is hard to predict by models (Hodges *et al.* 2019). In general, when a QSAR model is developed for a varied dataset, prediction accuracy can be improved by classifying the micropollutants in this dataset into specific groups that contain similar micropollutants (de Ridder *et al.* 2010).

Other micropollutants were detected in treated water even if the calculated EI_s was low and could not be disputed. It is unclear why these were detected. This may simply indicate that the actual removal of micropollutants is a process that can have a variable success. This is also shown by the high variability in measured removal within substances in the dataset (see Figure S1 in Supplementary material, Supplement I).

If the relationship between a micropollutant property and its behavior in a purification treatment is known from the literature, there is more reason to have confidence in the indicative removal rule. A high K_{ow} makes chemicals readily adsorb to particles and is therefore indeed expected as a defining property in predictor efficiency for pretreatment by coagulation/ flocculation. Aromatic compounds are a known predictor of oxidation with ozone (Sonntag & von Gunten 2012). The number of rings is a logical micropollutant property to indicate removal in advanced oxidation (e.g. Jenkin *et al.* 2020; Jin *et al.* 2020). Mean mass is a well-known and logical micropollutant property to predict removal for RO (Verliefde *et al.* 2008; Yangali-Quintanilla *et al.* 2009). For activated carbon, the substance property octanol-air partition coefficient (K_{oa}) emerged and this has not previously been associated with removal efficiencies. Its accuracy was also very low. A recent paper, however, also found K_{oa} as one of the properties explaining removability for treatment consisting of clarification, disinfection and GAC filtration (van den Doel *et al.* 2023). This indicative removal rule will have to be further confirmed or rederived in the future.

It should be noted that indicative removal rules do not predict removal rates for any specific treatment installation. Each individual drinking water treatment plant will have optimized treatment technology for the water matrix composition and type of pollution they encounter. Hence, it can be argued that the removal of 'known' micropollutants in general in any specific drinking water treatment plant is better than calculated by the indicative removal rule. The indicative removal rule indicates an average removal in such techniques or under different circumstances, based on micropollutant properties.

For a precise prediction of removability for specific treatment plants, more extensive models such as the tool 'AquaPriori' (Vries *et al.* 2013, 2017) can be used. This model also considers mutual interactions of micropollutants and interactions of the purification treatment with natural organic matter, which is not explicit in the indicative removal rules. AquaPriori is currently being further developed for multiple techniques and many micropollutants.

Processes in wastewater treatment facilities differ from those in drinking water treatment facilities. Some water types will even require very different treatment techniques than typical wastewater, like desalination brine treatment systems (Panagopoulos & Giannika 2022). Currently, for wastewater, the model 'SimpleTreat' (Struijs 2014) can be used to predict removal. Like with 'AquaPriori' (Vries *et al.* 2013, 2017) settings on the specifics of the treatment facility are required in this model. The same techniques as presented in this study can be used to derive indicative removal rules based on substance properties relevant to wastewater treatment systems. That could for instance be instrumental in a permit evaluation for industrial wastewater emissions.

To conclude, the performance of the individual indicative removal rules matches for the larger part the measured removal by treatment techniques in the Netherlands. Moreover, traces of micropollutants with a high EI_s based on the indicative removal rules are indeed found more often in drinking water. In total, 18 micropollutants that exceed their drinking water target or signaling value in the Rhine are labeled as particularly difficult-to-remove chemicals. The prioritization of micropollutants helps to direct policymakers to take measures to decrease emissions or otherwise remediate micropollutants to reduce the level of purification treatment required for the production of drinking water. Further validation of the indicative removal rules will set their shape more reliably and will make these rules applicable in a wider context. Indicative removal rules for other treatment techniques can be added or developed for distinct types of micropollutants. Moreover, the weight of treatment techniques from smaller to larger 'effort' can be established more objectively by adding a quantifiable measure for an effort such as increased energy expenditure, carbon footprint, and costs and basing weights on that measure. Until then, the EI_s can be used as the best available first-tier prioritization for low effort-removal efficiency of micropollutants.

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DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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