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Mind the Gap: Persistent and Mobile Organic Compounds—Water Contaminants That Slip Through

Thorsten Reemtsma,[*,†] Urs Berger,[†] Hans Peter H. Arp,[‡] Hervé Gallard,[§] Thomas P. Knepper,[∥] Michael Neumann,[⊥] José Benito Quintana,[#] and Pim de Voogt[∇,○]

[†] Department of Analytical Chemistry, Helmholtz Centre for Environmental Research—UFZ, Permoserstrasse 15, 04318 Leipzig, Germany

[‡] Norwegian Geotechnical Institute (NGI), P.O. Box 3930, Ullevål Stadion, 0806 Oslo, Norway

[§] University of Poitiers, Institute of Chemistry of Materials and Natural Resources, UMR CNRS 7285, ENSIP, 1 rue Marcel Doré, Poitiers Cedex 9, France

[∥] Hochschule Fresenius, University of Applied Sciences, Institute for Analytical Research, Limburger Strasse 2, 65510 Idstein, Germany

[⊥] German Environment Agency (UBA), Section IV 2.3 "Chemicals", Wörlitzer Platz 1, 06844 Dessau-Roßlau, Germany

[#] Department of Analytical Chemistry, Nutrition and Food Sciences, IIAA—Institute for Food Analysis and Research, University of Santiago de Compostela, 15782 Santiago de Compostela, Spain

[∇] Institute for Biodiversity and Ecosystem Dynamics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands

[○] KWR Watercycle Research Institute, Groningenhaven 7, 3430 BB Nieuwegein, The Netherlands

ABSTRACT: The discharge of persistent and mobile organic chemicals (PMOCs) into the aquatic environment is a threat to the quality of our water resources. PMOCs are highly polar (mobile in water) and can pass through wastewater treatment plants, subsurface environments and potentially also drinking water treatment processes. While a few such compounds are known, we infer that their number is actually much larger. This Feature highlights the issue of PMOCs from an environmental perspective and assesses the gaps that appear to exist in terms of analysis, monitoring, water treatment and regulation. On this basis we elaborate strategies on how to narrow these gaps with the intention to better protect our water resources.

INTRODUCTION

Fifty years ago, in 1966, Søren Jensen and co-workers identified polychlorinated biphenyls (PCBs) accumulating in the environment and in food-chains which were causing a threat to top predators like the white-tailed eagle.[1] Particularly alarming was that PCBs were considered to be "perhaps the most stable class of organic compounds in existence",[2] being able to persist in the environment long after their emissions. What causes PCBs to enrich in food-chains, though, was not just their persistence, but also their ability to enrich in animal fats, making them "bioaccumulative". This tendency is related to the molecular structure of PCBs, which are quite "nonpolar", allowing them to be both lipophilic and hydrophobic. Even today, PCBs originating from environmental emissions as far back as the 1960s continue to bioaccumulate in organisms. Since Jensen's pioneering work, PCBs and other persistent, bioaccumulative and toxic (PBT) chemicals have developed into the principle drivers of environmental chemistry, ecotoxicology research and ultimately chemical regulation. Milestones in this regard include the United Nations "Stockholm Convention"[3] and Europe's REACH regulation,[4] both of which intend to prevent emissions of PBT compounds into the environment.

With respect to drinking water quality, however, PBT-based regulations are only marginally effective. Most compounds capable of bioaccumulation are nonpolar and are inherently poorly water-soluble. Therefore, they can be readily removed from water by sorption processes in the environment or during water treatment. In contrast, persistent and mobile organic compounds (PMOCs) are more of a concern for water quality because, like PCBs, they can persist in the environment, but they are not removed from water by sorption processes due to their high polarity and thus excellent water solubility. Therefore, they may end up in drinking water, posing a potential risk to human health. Some examples of known PMOCs found widely in raw waters used for drinking water production include methyl-*tert*butylether (MTBE),[5] ethylenediaminetetraacetic acid (EDTA),[6] short-chain perfluoroalkyl acids (PFAAs),[7] and tris(2-chloroethyl)phosphate (TCEP).[8]

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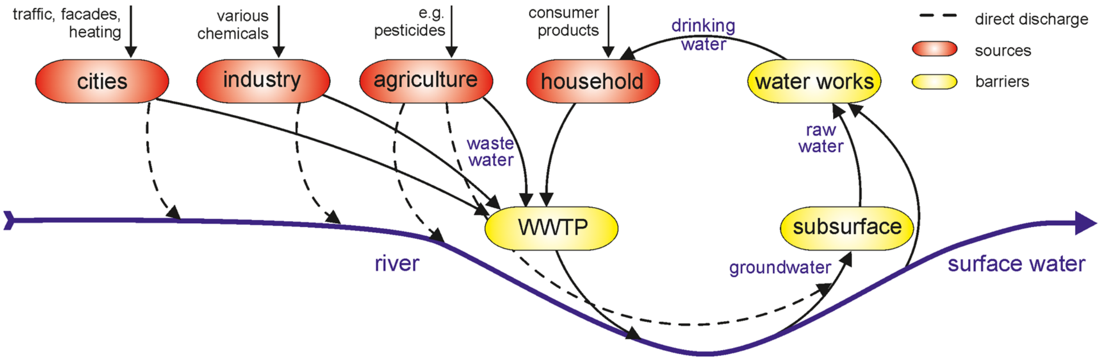

Figure 1. Scheme of a partially closed water cycle with emission sources of chemicals and different barriers (adapted from ref 42).

Unfortunately, unlike PCBs and other PBT chemicals, we currently lack the tools to monitor, treat and regulate PMOCs. This is because profound gaps exist in how to analyze and model these compounds in the environment. Herein we discuss these conceptual gaps, as well as suggest strategies for addressing them in order to protect drinking water resources.

Aquatic Mobility of PMOCs and the Modeling Gap. What makes a compound mobile in the aquatic environment? Mobility can be qualitatively defined as "the potential of the substance (...), if released to the environment, to transport to groundwater or far from the site of release" (Annex II of REACH[4]). However, there is no commonly accepted quantitative definition for a compound's mobility in regulations or elsewhere. Two possible quantifiers of mobility in water are water solubility and sorption tendency. These two properties are governed by the compound's molecular *polarity*, which in turn is dependent on its molecular structure.

Polar compounds contain an asymmetric distribution of negatively charged electrons on their molecular surface, resulting in positive and negative regions known as dipoles. The stronger and more frequent the dipoles are relative to the size of the molecule, the more polar it is and the more water-soluble it is. The extreme form of a dipole is an ionic charge. Ionic charges in molecules can be permanent (e.g., quaternary amines), or in the case of acids and bases, pH dependent. Compounds with multiple ionic groups are particularly water-soluble.

However, even molecules with a high solubility still may not be very mobile in water because of sorption. Solid phases, such as soils and minerals, can also contain dipoles and ionic charges that attract polar contaminants. This especially applies to cationic compounds, because most natural surfaces carry negative charges. Conversely, organic anions are expected to be poorly retained. Generally, the molecules most mobile in water are the ones in which solvation by water is more favorable (energetically) than sorption to environmental solids.

For neutral (uncharged) compounds, the partition coefficient between octanol and water (K_{OW}) can be used as an approximate indicator for a compound's sorption tendency, with a low K_{OW} value indicating high aquatic mobility. It should be emphasized that this is only an approximation, as the mobility of different classes of polar molecules may not necessarily be (logarithmically) proportional to K_{OW}. This is particularly true if these compounds can make additional specific (polar) and/or nonspecific (apolar) interactions with environmental surfaces and bulk phases that they do not make with octanol.[9] For ionic or dissociating compounds, K_{OW} is even more of an insufficient descriptor, as the compound may exist in a variety of complexes or protonation states depending on pH and on the ions present in the (pore)water. For such compounds, the distribution coefficient (D_{OW}) is used to account for the concentrations of all forms of the compound (ionized plus uncharged). As an example, for organic acids, the pH dependency of the D_{OW} can be expressed as

$$D_{ow} = \frac{K_{ow}}{1 + 10^{\mathrm{pH} - \mathrm{p}K_a}}$$

However, the assumption that the D_{OW} value inversely correlates with a compound's aquatic mobility is, certainly, very simplistic. In addition to its reliance on K_{OW}, this assumption disregards ionic interactions with other ions in solution, especially with counterions, and with ionizable or ionic sites on soils and other surfaces.[10] Thus, K_{OW} and D_{OW} can be viewed as the approximate indicators of mobility, which biases toward overestimation due to the lack of accounting for additional interactions between the compound and environmental media or environmental water that do not occur between the compound and octanol or pure water. Despite these shortcomings, we here use modeled log D_{OW} values (at pH 7.4) for illustrative purposes as proxy for polarity and aquatic mobility.

More sophisticated models for very polar and ionic molecules are only just emerging, and only cover a subset of all possible combinations of polar and ionic moieties. A particular challenge is that molecules with multiple functional groups behave differently than expected based on compounds with single functional groups.[11] These challenges in predicting the sorption behavior and thus the aquatic mobility of very polar molecules result in a current modeling gap for PMOCs. Moreover, experimental data, which could be used to calibrate and improve models for a wide range of PMOCs, are scarce.

Partially Closed Water Cycles and the Monitoring Gap. In densely populated areas surface water is an important source for the production of drinking water, either by direct abstraction and treatment or, more often, after subsurface passage such as through bank filtration[12] or after infiltration. Surface waters are, however, the recepients of effluents of wastewater treatment plants (WWTPs) as well as of runoff from impervious urban surfaces and agricultural land (Figure 1). In this way, partially closed water cycles may be (often unintentionally) established on regional scales (Figure 1). In such partially closed cycles, WWTPs and the subsurface environment are the major barriers that prevent contaminants originating from wastewater from reaching raw water used for drinking water production.

In both these barriers essentially the same removal processes are effective: microbial degradation and sorption. Biodegradation in WWTPs occurs mostly aerobically, while in the subsurface anoxic conditions may prevail. Sorption may occur to the microbial biomass (sewage sludge) in WWTPs, to sediments and soils on the way to the subsurface, or to aquifer materials in the subsurface environment. Both wastewater treatment and subsurface barriers are expected to be largely ineffective for the removal of PMOCs, as by definition these are neither biodegraded nor sorbed substantially, and therefore PMOCs may reach the raw waters used for drinking water production. If drinking water treatment is not suitable to remove PMOCs, such partially closed water cycles may turn into chemical cycles for PMOCs.[13] Dilution would then be the only process reducing the concentration of the most persistent PMOCs.

Recently, Sjerps et al.[14] published the outcome of an analytical screening for contaminants in Dutch water samples, covering the journey from WWTP effluent to surface water to groundwater and then to drinking water. They noted that the concentration level of total organic contaminants decreased by about 2 orders of magnitude from the WWTP effluents to the groundwater used for drinking water production. However, using the chromatographic retention time as an indicator for hydrophobicity, the authors concluded that particularly the most polar contaminants in the WWTP effluents remained in the water throughout its passage to groundwater.

The limited number of very polar compounds reported so far to occur in our groundwaters does, by itself, not indicate that only very few such contaminants are present. Rather, this indicates that such compounds have been rarely searched for and that a gap in monitoring exists for PMOCs. As a consequence, we are presently unable to judge how large this monitoring gap may be and how many PMOCs are hidden in raw waters used for drinking water production. But why does this gap in monitoring exist?

The Analytical Gap. The advent of the coupling of liquid chromatography with mass spectrometry (LC-MS) in the 1990s tremendously improved the sensitivity and selectivity of polar contaminant analysis. This was most obvious for pesticides, but also for pharmaceutical residues in the environment.[15] Ever since, the list of emerging polar pollutants, for which LC-MS methods were developed, has grown rapidly. These pollutants include whole classes of contaminants such as surfactants, perfluoroalkyl acids,[16] drugs of abuse,[17] polar industrial chemicals, and polar transformation products of both polar and nonpolar parent compounds.

Indeed, LC-MS technology has been one of the most competitive areas over the last two decades in terms of instrumental development, with advances in the speed and the sensitivity of analysis. State-of-the-art instruments now allow ultratrace multiresidue analysis with hundreds of chemicals being measured in a single run, even after a simple direct injection of an aqueous sample. Another important development is the popularization of high resolution MS (HRMS) analyzers, which has been a huge step for the identification of transformation products of organic pollutants[18] as well as for the screening for unknown substances.[19]

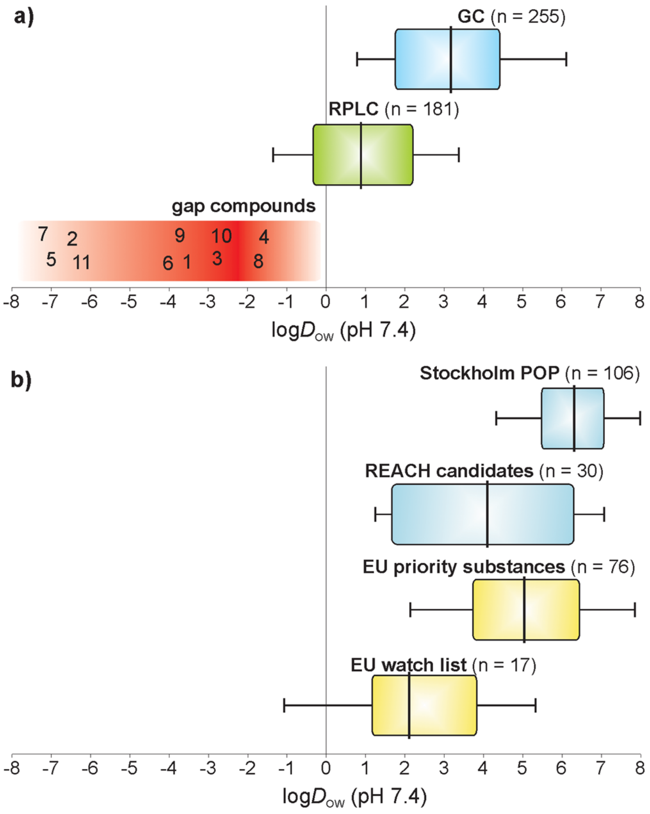

Figure 2. Box and whisker plots of calculated logD_{OW} values at pH 7.4 (ChemAxon) of: (a) contaminants in water analyzed by either GC-MS (EPA methods 8270 D and 8290 A[43,44]) or LC-MS (ref 45 Tables S2 and S3) and of examples of "gap" compounds; (b) contaminants regulated by the Stockholm Convention,[3] candidates of Substances of Very High Concern (SVHCs) according to REACH, Article 57, d–f,[30] the list of priority substances according to the Water Framework Directive (WFD) and the so-called Watch List of the WFD.[29] The whiskers point to the 10th and 90th percentile. Numbers in (a) refer to 1: Aminomethylphosphonic acid (AMPA), 2: Paraquat, 3: Cyanuric acid, 4: *N*,*N*-dimethylsulfamide (DMS), 5: Diquat, 6: 5-Fuorouracil, 7: Glyphosate, 8: Melamine, 9: Metformin, 10: Perfluoroacetic acid, 11: EDTA.

Against this background, it may seem that our instrumental portfolio with gas chromatography (GC)-MS and LC-MS is now complete and principally allows for determining all organic contaminants from water. Indeed, the polarity range (expressed as range in D_{OW} values) that is covered by GC and LC-based methods is impressively wide, covering more than 8 orders of magnitude (Figure 2a). However, LC-MS strongly relies on the use of reversed-phase LC (RPLC), in which retention is based on nonpolar interactions of the analytes with typically a C18 material. Not all contaminants of interest are amenable to either GC or RPLC separation.

In particular, the polarity range characterized by negative logD_{OW} values is hardly amenable to either GC or RPLC separation (Figure 2a). However, it is within this chemical space that most PMOCs are expected to fall, as illustrated in Figure 2a with a few example PMOCs (having a logD_{OW} at pH 7.4 between −1 and −8). The most mobile PMOCs in water are likely also the most poorly retained in RPLC. These would tend to elute together with all the other highly polar matrix constituents of aqueous samples in the so-called "void volume" of the separation system. This prevents their effective ionization in the LC-MS interface. Although novel RPLC phases have been developed, such as columns that work in an expanded pH range or allow for completely aqueous mobile phases (free from organic modifiers), or by polar end-capping, an analytical gap still persists for the most hydrophilic neutral analytes and particularly for permanently charged compounds. Known examples of such chemicals are:

- Highly polar pesticides and their transformation products, for instance glyphosate and aminomethylphosphonic acid (AMPA), with modeled $\log D_{OW}$ values (pH 7.4) of −6.66 and −5.95, respectively. These compounds can only be determined by RPLC after derivatization.[20]
- Short-chain (especially C2 and C3) perfluorinated carboxylates and sulfonates ($\log D_{OW}$ −0.4 to −2.6), which were only recently discovered in surface and drinking water when a mixed-mode LC (MMLC) method was applied.[21]
- Quaternary ammonium compounds, e.g. diquat, paraquat ($\log D_{OW}$ −6.7 to −7.0).
- Complexing agents, such as EDTA, which can be found in high concentrations in drinking water, but again need dedicated methods based on ion chromatography (IC)[22] or ion-pairing.[23]

Although derivatization, ion-pair liquid chromatography (IPLC) and IC can help to fill the analytical gap, these approaches require very specific sample preparation or LC-MS methods. This hampers their application in multiresidue analysis of PMOCs as well as in nontarget LC-HRMS screening for the discovery of new PMOCs.

Besides chromatography, enrichment of very polar chemicals from water samples is also a challenge. Although polar solid-phase extraction (SPE) sorbents, including several mixed-mode sorbents, have been developed, problems with SPE breakthrough or incomplete elution from mixed-mode sorbents still persist for many analytes. Carbonaceous materials have also been tested, but have never become popular due to difficulties in elution.

We conclude that generically applicable enrichment and chromatographic methods for highly polar PMOCs in water samples are currently lacking. Taken together, these obstacles result in a lack of suitable analytical methods for screening or monitoring of PMOCs and, consequently, have led to the gaps in monitoring data and model calibration data described above.

The Treatment Gap. If the environmental barriers in the water cycle are not effective in preventing PMOCs from reaching raw waters used for drinking water production, what about the technical barriers in drinking water treatment? Can we reasonably assume that these remove PMOCs?

Presently, activated carbon (AC) and ozonation are the most widely applied treatments for the removal of organic contaminants in drinking water preparation. But AC also relies on sorption and therefore low efficiency is expected for very polar compounds. Literature data confirm this trend with generally low removal for compounds with $\log D_{OW} < 1$, such as the contrast agent iopromide ($\log D_{OW}$ −0.44), the anti-inflammatory ibuprofen ($\log D_{OW}$ 0.85)[24,25] and short-chain perfluoroalkyl acids.[7] In some cases, however, ionic interactions with the AC surface can potentially support the removal of some more polar compounds, such as with caffeine ($\log D_{OW}$ −0.55).[24,26]

With respect to ozonation, polar compounds with acidic functional groups (e.g., carboxylic and sulfonic acids) usually show low reactivity.[7,27] These and other polar compounds such as small ethers, alkylphosphates and triazines can only be eliminated by reactions with OH radicals. Some compounds like tri(2-chloroethyl) phosphate have a very low reactivity even toward OH radicals and thus may be present in finished drinking water.[27] Taken together, there is a considerable risk that many PMOCs would only be poorly removed in drinking water production, resulting in a treatment gap.

Complicating matters further, ozonation itself can be a source of PMOC transformation products, which are usually smaller and more polar (oxidized) than the parent compounds. For example, Schmidt and Brauch[28] recognized that the carcinogenic *N*-nitroso-dimethylamine (NDMA) is formed from the ozonation of drinking water containing *N*,*N*-dimethylsulfamide (DMS). Because of the analytical gap for very polar compounds, one must assume that many ozonation products may not have been recognized thus far. Fortunately, there is some indication that ozonation products are generally less toxic and less persistent than their parent molecules, and are more likely to be biodegraded during biological filtration following ozonation, as was the case for NDMA.[28] However, almost certainly, there are also exceptions to this rule.

To conclude, PMOCs may not be removed and may even be formed during drinking water treatment. However, judging the extent of this problem remains speculative because of the analytical, monitoring and modeling gaps.

The Regulatory Gap. Raw water resources for the production of drinking water merit a high level of protection. Under the Water Framework Directive (2000/60/EC)[29] in the European Union, priority substances and priority hazardous substances have been identified for which emissions have to be either reduced or eliminated. Such decisions are, *inter alia*, dependent on monitoring data, which, in turn, require established analytical procedures. It comes thus as no surprise that the overwhelming majority of contaminants or classes of contaminants defined as "priority substances" or included in the so-called "watch list" in the EU are nonpolar or only moderately polar (Figure 2b). Only three (clothianidine, imidaclopride, azithromycin) of the approximately hundred compounds on these two lists exhibit a $\log D_{OW}$ value below 1.

High production volume industrial chemicals have to be registered in Europe as part of the REACH Regulation (1907/2006 EG).[4] Registrants are responsible for the chemical risk assessment and are asked to ensure a high level of protection of human health and the environment. Chemicals identified as substances of very high concern (SVHCs) by the Member State Committee in accordance with Article 59 of the REACH Regulation[4] are added to the so-called Candidate List.[30] SVHCs may be included in the Authorisation List and then become subject to authorization. To date, all chemicals that are on the Candidate List are there because their intrinsic properties caused them to be classified as either PBT or very persistent and very bioaccumulative (vPvB), and, therefore, hazardous to the environment. These compounds cover a broader polarity range than the legacy persistent organic pollutants (POP) regulated under the Stockholm Convention[3] (Figure 2b). But still, no really polar, mobile chemical has been identified as SVHC. This reflects that the protection of raw water resources is not yet in the focus of the REACH process.

Thus, neither in the field of surface water and groundwater nor of chemical regulation have PMOCs been considered specifically. We suggest that there is a regulatory gap for PMOCs, though the lack of analytical methods, monitoring, and modeling data, again, hampers an evaluation of the magnitude and severeness of this gap.

Steps toward Closing the Gaps. With so many gaps in our knowledge regarding PMOCs, we cannot assess their importance for water quality or human health, nor can we develop strategies to prevent future risks from emerging PMOCs. However, the existence of such knowledge gaps is in itself a risk. Therefore, these gaps need to be closed. How can this be accomplished?

Analysis. Obviously, the analytical gap is the basic problem. Ineffective chromatography due to poor retention on RPLC columns is the major culprit. Presently, hydrophilic interaction LC (HILIC), MMLC, and supercritical fluid chromatography (SFC) with normal phases appear promising for the chromatographic separation of very polar organic contaminants prior to their detection by mass spectrometry, besides the already established IPLC and IC. For example, very recently, perfluoromethanesulfonic acid and other halogenated methanesulfonic acids were discovered as novel PMOCs in wastewater, surface water and raw waters used for drinking water production by using HILIC-MS.[31]

MMLC typically combines RPLC (or HILIC) properties with ion exchange mechanisms. Currently, different manufacturers provide mixed-mode columns with either anionic or cationic exchange capabilities, and, more interestingly, state-of-the-art columns with both types of ion exchange mechanisms.[32,33] Yet, to date, most applications of MMLC focus on bioanalysis or characterization of pharmaceutical products, since both the drug and the counterion can be quantified in a single run. However, we expect that mixed-mode columns will considerably expand the polarity range of chemicals in LC-HRMS screening and LC-MS multiresidue analysis.

SFC with normal phase columns is another chromatographic technique that has recently reached maturity. Its application range can be extended toward highly polar analytes by using polar cosolvents in the supercritical CO_2 mobile phase.[34] However, some fundamentals, such as the selectivity of the separation process toward the polarity of analytes, are still poorly understood.[34] It remains to be proven if SFC can help to fill the analytical gap for PMOCs.

Figure 3 shows how these three novel separation techniques can improve chromatographic retention in comparison to the conventional RPLC approach for a selection of PMOC "gap" compounds. At this stage, unfortunately, none of the three approaches will likely be suitable for the retention and chromatographic resolution of all PMOCs. But they appear as valuable complements to the more established methods like IPLC and IC.

Monitoring. With the help of these novel analytical approaches we will be able to monitor for more PMOCs, and close, at least partially, the monitoring gap. However, a second prerequisite for monitoring is the knowledge of PMOCs that are expected to occur in the aquatic environment. Two strategies may help us to identify and prioritize potential analytical targets: (a) nontarget screening for PMOCs with these novel chromatographic methods combined with full-scan HRMS and (b) the search in compound databases for candidates that are likely to be PMOCs based on their measured or modeled physicochemical properties.

Well-designed monitoring studies will reveal the number and identity of PMOCs present in the water cycle as well as their typical concentration levels. This will allow us to study their sources, formation processes and transport pathways. Monitoring along the water cycle, from surface water to groundwater and to raw waters used for drinking water production, will show to which extent the existing barriers in partially closed water cycles are effective in removing certain PMOCs, and which PMOCs occur in raw waters used for drinking water production. For these PMOCs, generation of toxicity data (if not already existing in, e.g., REACH registration dossiers) is a further required step toward regulation. It will have to be proven whether existing models to predict toxicity are adequate for the domain of PMOCs.

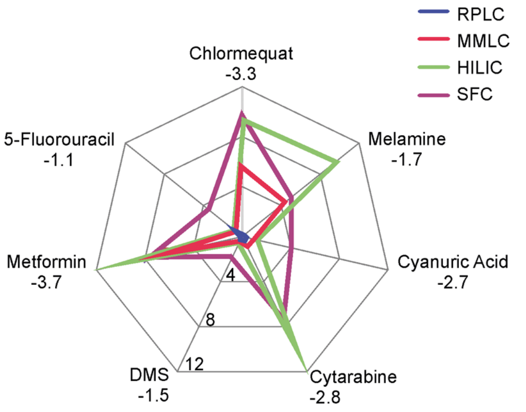

Figure 3. Comparison of capacity factors (defined as$(t_r-t_0)/t_0$, where t_0 is the retention time of the void volume and t_r the retention time of the analyte), presented as distance from the center, obtained by RPLC, MMLC, HILIC, and SFC for 7 highly polar chemicals (unpublished data). RPLC: Waters C18 column, gradient from 2 to 100% methanol with ammonium acetate. MMLC: Thermo Trinity P1 column; dual gradient from 2 to 80% acetonitrile and from 10 to 40 mM ammonium acetate at pH 5.5. HILIC: MN Nucleodur HILIC column; gradient from 95 to 40% acetonitrile containing 5 mM ammonium formate (pH 3). SFC: Waters BEH 2P column, gradient from 10 to 50% cosolvent ($MeOH/H_2O$ 95/5 with 0.1% NH_4OH) in CO_2. The logD_{OW} values for pH 7.4 were obtained from ChemAxon and are indicated below the compound names.

Modeling. As the analytical gap narrows, it will also become easier to directly measure critical parameters like logD_{OW} or even more importantly, sorption coefficients to environmentally relevant media like soils, minerals and water treatment filters, covering a wide range of aquatic parameters (pH, salt concentration) and ion exchange and sorption sites. Additionally, experimental data regarding their persistence in biodegradation or hydrolysis assays can be obtained. This data will help to test and calibrate sorption and persistency models, and ultimately help to close the modeling gap for PMOCs. The generation of monitoring data will assist in prioritizing compound classes of PMOCs for which models need to be developed as well as in validating the models.

Water Treatment. Provided that PMOCs are, indeed, present in our drinking water resources, two treatment options appear most promising: advanced oxidation processes and high pressure membrane processes. The peroxone process (i.e., O_3/H_2O_2) generates higher concentrations of OH radicals than ozone alone and also minimizes bromate formation. The UV/H_2O_2 advanced oxidation system has usually been implemented after microfiltration and reverse osmosis (RO) for potable reuse applications.[35] More recently, the Cl_2/UV process was also proposed as an economic solution for the control of emerging contaminants,[36] for example, for the removal of NDMA and trichloroethylene from groundwater.[37] While these techniques seem promising for treatment of PMOCs, their efficacy still needs to be proven and the risk of generating unwanted byproducts needs to be evaluated. All these oxidative processes would likely require polishing by a biologically active filter.

High pressure membrane filtration processes, either as nanofiltration (NF) or RO, have a great advantage over oxidation processes: they are able to remove polar organic compounds from water without producing unknown and unwanted transformation products. However, as these processes do not destroy the contaminants, they produce a considerable volume of brine (approximately 25% of the treated water) that has to be treated separately. NF was first implemented in full scale drinking water treatment in 1999 at Méry sur Oise near Paris (France). The use of NF on a wider scale is still hindered by high costs and fouling phenomena. Furthermore, neutral molecules with molecular weights <275 Da may be removed only partially.[38] RO generally provides better removal of contaminants than NF, but in practice RO membranes have also been found to only moderately remove low molecular weight organics and other small uncharged compounds.[39] Nevertheless, plans are on the way in different European countries to establish full scale RO facilities for the production of drinking water.[40] Once these systems are implemented, we will have the chance to learn about their potential to also remove yet unknown PMOCs.

Regulation. Given the high risk that PMOCs, because of their intrinsic substance properties, reach drinking water resources, it may be advisable to explore and develop regulatory means to avoid their release into the environment through risk mitigation measures during production and downstream use. This is especially true for PMOCs that are also toxic (T), that is, persistent, mobile and toxic (PMT) compounds. Such compounds could potentially be identified from the data provided as standard information requirements for registration under REACH or similar chemical regulations.[41] PMT compounds may be of equivalent level of concern as PBT substances, and, consequently, in Europe will be identified as SVHCs according to the criteria of Article 57 (f) of the REACH Regulation.[4] In this manner, authorities would help to prevent emissions "upstream" as a complement to finding more technically advanced and more costly water treatment technologies to solve the problem "downstream". Such chemical regulation would support drinking water safety, as it would prevent the future contamination of raw waters used for drinking water production.

Is M the New B? Fifty years after Jensen's discovery of bioaccumulative (B) persistent organic pollutants[1,2] we call for directing scientific attention toward mobile (M) contaminants that are of concern for drinking water quality. It is interesting to ponder what the field of environmental chemistry, treatment technology and regulation would look like today if Jensen had not discovered PCBs in seabirds, but rather PMOCs in drinking water. The design of much of our analytical methods, remediation activities, environmental fate models and regulations would be developed on different, and arguably more complex, technologies and concepts. Now it is time to explore what these concepts should be.

AUTHOR INFORMATION

Corresponding Author

*E-mail: thorsten.reemtsma@ufz.de.

Notes

The authors declare no competing financial interest.

Biographies

Thorsten Reemtsma is Head of the Department of Analytical Chemistry at the Helmholtz Centre for Environmental Research − UFZ in Leipzig and professor for Analytical Chemistry at the University of Leipzig; his expertise is in the analysis and fate of environmental contaminants.

Urs Berger is a senior scientist at the Department of Analytical Chemistry, Helmholtz Centre for Environmental Research - UFZ in Leipzig with a research focus on the development and application of trace chemical analytical methods for emerging contaminants.

Hans Peter H. Arp is a Principal Engineer at the Norwegian Geotechnical Institute in Oslo, Norway, who works with diverse aspects of assessing and remediating chemical pollution.

Hervé Gallard is professor at the Poitiers Chemistry Institute (IC2MP) at the University of Poitiers (France) with focus on chemistry of oxidants in water treatment.

Thomas P. Knepper is Vice President for Research at the University of Applied Sciences Fresenius in Idstein, Germany and professor for analytical chemistry; his expertise is in the analysis and fate of polar pollutants.

Michael Neumann works as scientific officer at the German Environment Agency (UBA) in the department chemical safety with focus on the EU-regulation REACH and is expert in environmental exposure and PBT assessment. Since 2014 he is a member of the Risk Assessment Committee at the European Chemical Agency (ECHA) in Helsinki dealing with harmonised classifications, restrictions and authorizations.

José Benito Quintana works as Associate Professor in Analytical Chemistry at the University of Santiago de Compostela, with focus on the analysis and fate of aquatic and marine pollutants.

Pim de Voogt is professor of environmental chemistry at the Institute for Biodiversity and Ecosystem Dynamics of the University of Amsterdam and principal scientist at KWR Watercycle Research Institute, Nieuwegein, NL; his expertise is in analysis and fate of emerging environmental contaminants.

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