



Persistence and Mobility (defined as organic-carbon partitioning) do not correlate to the detection of substances found in surface and groundwater: Criticism of the regulatory concept of Persistent and mobile substances



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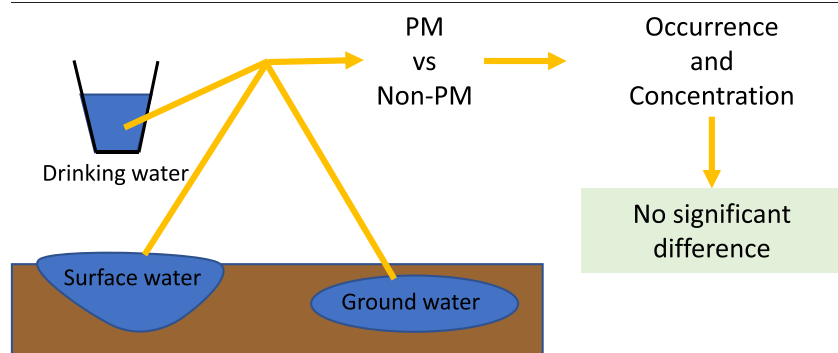
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HIGHLIGHTS

- PM substances do not have a higher likelihood of detection than non-PM substances.
- Likelihood of detecting a P substance in water is independent of the M criterion.
- There is no indication that PM substance accumulate in water bodies.
- No evidence of log Dow or log Koc as driver of contamination of water.

GRAPHICAL ABSTRACT



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ABSTRACT

The Chemical Strategy for Sustainability (CSS) includes actions to ensure the protection of drinking water resources from chemical pollution. To proactively identify potential pollutants, the German Environment Agency (UBA) proposed the Persistent and Mobile (PM) concept according to which Persistence (criteria of REACH Annex XIII) and Mobility ($\log K_{oc} < 4$) would be proxies for a substance's degradation potential and transport velocity, two processes believed to drive the potential for contamination of surface and groundwater as drinking water sources. Two studies identified hundreds of PM substances while three subsequent studies have selected some of these substances for monitoring in surface, ground- and/or drinking water to support the concept. In the present work, the Persistence of the aforementioned substances was reassessed based on all experimental data publicly available. Depending on the exact study examined, it was found that 15 % to 40 % of the substances were erroneously concluded as P. The reinterpretation of the data indicates that a PM substance does not have a higher likelihood to be detected in surface or groundwater than a non-PM substance. In addition, the PM properties do not have any influence on the level of contamination. Twenty-six to 75 % of the substances selected because they were identified as PM were not found in surface or ground water despite being selected for their high emission pattern. Regulations based primarily on the PM concept, like the CLP and possibly REACH and UN-GHS, are unlikely to appropriately identify substances of concern for drinking water sources. It is more likely that chemical presence in surface and groundwater is driven by

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emission patterns or local factors. The development of specific exposure models would better contribute to the protection of drinking water resources and consumers.

1. Introduction

As part of the Chemical Strategy for Sustainability (CSS) roadmap published in October 2020, the European Commission presented in May 2021 its ambition « Towards Zero Pollution for Air, Water and Soil » by 2050 (EC, 2020). Pollution was defined by the European Commission as “the direct or indirect introduction, as a result of human activity, of substances, vibrations, heat or noise into air, water or land which may be harmful to human health or the quality of the environment, result in damage to material property, or impair or interfere with amenities and other legitimate uses of the environment.” Zero Pollution for substances was defined as the reduction of direct and indirect emissions of chemicals “to levels no longer considered harmful to health and natural ecosystems and that respect the boundaries our planet can cope with, thus creating a toxic-free environment” (EC, 2021).

The protection of drinking water sources is one of the cornerstones of this roadmap. Thus, many researchers have highlighted a new type of ‘contaminant of emerging concern’ found in surface and groundwater, which may be used for the production of drinking water (Lapworth et al., 2012; Bunting et al., 2021). Reemtsma et al. (2016) hypothesized that the substances of highest concern for drinking water contamination are the Persistent and Mobile Organic Compounds (PMOCs). Based on the properties of Persistence and Mobility, the researchers indicate that these substances would likely be in drinking water resources and potentially remain undetected by conventional analytical methods (Zahn et al., 2020). Therefore, this family of substances is considered of increasing concern by the drinking water production sector and signals a need for regulation (Nödler and Scheurer, 2019).

In response to this concern, the UmweltBundesamt (UBA, German Environment Agency), in collaboration with the Norwegian Geotechnical Institute (NGI), developed the Persistent and Mobile (PM) concept and the related PMT/vPvM criteria, which were intended to parallel the Persistent, Bioaccumulative and Toxic (PBT) and very Persistent and very Bioaccumulative (vPvB) criteria of the various European environmental legislations. PMT stands for Persistent, Mobile and Toxic, while vPvM stands for very Persistent and very Mobile. The underlying rationale of the PM concept is that chemicals that migrate through natural barriers like soils and sediment faster than they degrade have the potential to reach water resources, in particular, those used for drinking water production (Hale et al., 2020a, 2020b). These researchers asserted that Persistence and Mobility are a combination of intrinsic properties that drive the potential of a chemical present in the environment to infiltrate natural barriers (e.g., riverbank, soil...) and conventional means of drinking water production such that humans are exposed via drinking water (Hale et al., 2020a, 2020b). The potential for a substance to pass through natural filtration is the net result between weakly retarded transport velocity (i.e., high Mobility) and low degradation rate (i.e., Persistence). The degradation rate is mostly driven by microbially-mediated biodegradation and abiotic processes such as hydrolysis. The retarded transport velocity is the net result between the speed of the carrier (water) and the physicochemical processes which remove (temporarily or definitively) the chemical from water (e.g., adsorption, ionic binding, entrapment in organic matter, covalent binding to particles, etc.).

The criteria for the identification of Persistent and Mobile (PM) substances were initially proposed in 2014 by Kalberlah et al. (2014) and later adjusted in 2019 by Neumann and Schliebner (2019) for implementation under Regulation EC 1907/2006 (REACH) and used by Arp and Hale (2019) to screen REACH-registered substances. For the sake of simplicity, Neumann and Schliebner (2019) proposed the P/vP criteria of REACH Annex XIII to describe degradation and proposed to simplify the retarded transport velocity – termed « Mobility » – with the single physicochemical process of adsorption. The organic carbon-water partition coefficient (Koc) and the n-octanol-water distribution ratio (Dow) were proposed as metrics

for neutral and ionizable chemicals, respectively. Cut-off criteria for M were derived from a benchmarking exercise comprised of a large dataset of surface and groundwater monitoring data (Neumann and Schliebner, 2019). Non-detects and detection below 0.1 µg/L represented the vast majority of the results but were disregarded in the benchmarking exercise as only substances exceeding the 0.1 µg/L limit of the EU's Drinking Water Directive (Directive (EU) 2020/2184; DWD) were considered. Detection of a chemical in surface or groundwater was considered sufficient, irrespective of measured concentrations and rates of detection, and given the same weight regardless of intrinsic toxicity, actual human exposure or risk.

ECETOC (2021a, 2021b) reviewed the publicly available part of this dataset and additional monitoring surveys, and they concluded that non-detects and detection below 0.1 µg/L (considered as a proxy for risk in the European Plant Protection Products regulation (Regulation EC 1107/2009; PPPR)) represented the vast majority of the results (99.2 %). Neumann and Schliebner (2019) concluded that regardless of the degradation potential of the substance, most chemicals detected had a log Koc or log Dow <4 and concluded that this was the threshold below which a chemical had the intrinsic property to reach surface or groundwater (M). A threshold of log Koc or log Dow <3 was proposed for very Mobile substances (vM).

In November 2022, the exact criteria for the regulation of PM substances were still being debated at the European Competent Authorities for REACH and CLP (CARACAL) for their inclusion as new hazard classes under the European Classification Labelling and Packaging Regulation EC 1278/2008 (CLP).

Since the proposal for PM criteria (Neumann and Schliebner, 2019), several analyses have been conducted to test the appropriateness of the PM concept. For instance, Schulze et al. (2019) monitored 57 chemicals in 14 different water sources across Europe. These chemicals were selected mostly from a QSAR-derived PM list of 936 REACH-registered substances with the highest likelihood for environmental emissions (Schulze et al., 2018). Schulze et al. (2019) detected 43 out of 56 chemicals at least once in natural waters, of which 23 had never been investigated before, and they concluded that the PM approach was thus fit for purpose. Neuwald et al. (2021) performed an analytical screening analysis on surface water across multiple catchments in Germany and were able to identify and quantify 64 PM substances among a list of 1300 suspected PM substances which included the list of Schulze et al. (2018). Huang et al. (2021) collected monitoring data on 432 pharmaceuticals and personal care products (PPCPs) across 75 monitoring studies in China. These chemicals have by definition wide-dispersive uses. One-hundred thirty-eight of these chemicals (31.5 %) were detected at least once in surface water. Among these 138 compounds, the researchers considered that 86.2 % were (potential) PM substances. One hundred six of these chemicals were detected at least once in drinking or ground water (data from both were combined). Among these 106 compounds, the researchers considered that 89.6 % were (potential) PM substances. These studies were considered as a proof of concept by their respective authors but simultaneously raised concern for the additional undetected suspected PM substances in water. However, on the basis of a large monitoring dataset, ECETOC (2021a, 2021b) did not find any trend between log Dow and detection of chemicals above 0.1 µg/L. In addition, ECETOC (2021a) analyzed a set of 62 chemicals monitored in surface and groundwaters of Great Britain, of which 24 % were not persistent. The other chemicals were either confirmed as persistent based on degradation half-lives (DT50) obtained in experimental simulation tests or considered (potentially) persistent based on negative screening tests. On the basis of these data, the authors (ECETOC 2021a, 2021b) concluded that PM substances did not have a higher likelihood of being found in surface or groundwater than non-PM substances. The conclusion by ECETOC (2021b) is greatly important since it is the first and only attempt to date

to make an objective comparison between PM and non-PM substances. All other published work focused solely on the detection of suspected PM substances in surface and groundwater and did not aim at monitoring non-PM substances for comparison. The ECETOC (2021a, 2021b) review raises the question if the PM concept as defined by UBA/NGI needs more work to establish an undisputable and objective proof of concept as a truly protective measure of drinking water sources.

In the available studies listed above, the identification of substances having both P and M properties (as defined in the UBA/NGI concept) was key for proving the concept and justifying concerns. Since these included hundreds of substances to screen, the researchers chose to mostly rely on high-throughput QSAR-based P and M assessments (Schulze et al., 2018). However, even though Persistence QSARs are useful for screening purposes, it is well-known that their applicability domain is limited for certain chemical classes. Consequently, any Persistence prediction should be carefully evaluated on a case-by-case basis with regards to the applicability domain and the available experimental studies (e.g., ready / inherent biodegradability and biodegradation simulation tests available in REACH dossiers).

These studies constitute the basis of the PM concept which is now proposed for inclusion as new hazard classes under the CLP regulation in the European Union, with the aim to be proposed under the United Nations Global Harmonized System (GHS) for classification and labelling at a later stage (Hale et al., 2020a, 2022; Jin et al., 2020; EC, 2020). Subsequently, this inclusion will cascade to sector legislations such as REACH, the DWD, and the Groundwater Directive (Directive 2006/118/EC; GWD). It is therefore of utmost importance that any concept proposed for inclusion into any legislation is based on sound science to ensure adequate and targeted management of chemicals of true concern while avoiding unnecessary costs linked to mislabeled substances (unnecessary risk management measures, monitoring campaigns, watch list inclusion, etc.).

As it was unclear if the QSAR predictions were compared to available experimental data, the objective of this work was to conduct an in-depth P/vP reassessment of the substances investigated in Schulze et al. (2018, 2019, 2020), Montes et al. (2019, 2022), Teychene et al. (2020), Neuwald et al. (2021), and Kolkman et al. (2021) by inclusion of publicly available testing information supporting an assessment of Persistence in an effort to clarify the science behind the proposed concept. No reassessment of the Mobility assignment of the substances was performed in this work, since there is no comparably available Mobility data. Toxicity of the chemicals was not taken into account to remain aligned with the original work done in the publications reviewed as part of this work. Moreover, while toxicity is important for risk assessment, toxicity will not influence the probability of a substance reaching surface or groundwater; therefore it is not a critical parameter to be considered within the scope of this work. In June 2022, no other study on PMT or Persistent and Mobile Organic Chemicals (PMOC) relevant for the purpose of this work (i.e., monitoring a well-identified list of substances labelled either "PM" or "PMOC" in several surface and/or groundwaters) was found. The reassessment of the Persistence status supported a review of the substance profile investigated in the environmental monitoring studies, which were the basis for raising the following questions:

- i) Do substances characterized as PM have a higher potential than substances characterized as non-PM to reach potential sources of drinking water (surface and groundwater)?
- ii) Do PM-characterized substances accumulate in these potential sources of drinking water?
- iii) What could possibly drive the presence of these chemicals in these potential sources of drinking water?

2. Material and methods

2.1. Methodology of Persistence reassessment

The list of substances monitored in Schulze et al. (2018, 2019, 2020), Montes et al. (2019, 2022), Teychene et al. (2020), Neuwald et al. (2021), and Kolkman et al. (2021) were retrieved from the Supplementary

Information of each publication. For each substance, experimental data related to ready biodegradability, inherent biodegradability and biodegradation simulation tests (OECD 307/308/309) were retrieved, when available, from the disseminated information on the website of the European Chemical Agency (ECHA; accessed between the 8th and 21st of January 2022). When a substance was not found to be registered under REACH, attempts were made to identify the registration dossier of a closely related substance (e.g., positional isomers) for which experimental biodegradation information was available. Where these attempts were unsuccessful, experimental biodegradation data in peer-reviewed scientific literature were considered. The studies were considered relevant and used for the assessment when they were assigned a score of Klimisch 1 or 2 (reliable without restriction or reliable with restriction), when they were conducted using inoculum as allowed in the OECD Test guidelines, and when the concentration of inoculum was within the limits of the corresponding OECD Test guidelines. When the amount of information was limited (e.g., dossiers submitted under the Notification of New Substances (NONS) scheme of Directive 67/548/EEC in place before REACH), the study was not used for the assessment even if the Klimisch score was of 1 or 2 (Klimisch et al., 1997). For read-across, only read-across from the acid form to the base form, or vice-versa, were used with the exception of toluene sulfonate for which the read-across is based on a positional isomer (see more details below).

The Persistence assessment was conducted using the criteria of Annex XIII of REACH according to the methodology described in the ECHA PBT guidance document (ECHA, 2017). Additional information on the methodology and the scientific rationale of this Persistence assessment are available in the Supplementary Information (section S1). To minimize the errors in assessment, the analysis was conducted by an anonymous reviewer, and validated against an independent analysis by an external contractor with sole instructions to follow ECHA's PBT guidance (ECHA, 2017). The Environment Agency (UK) also independently reviewed the list of Schulze et al. (2019). Due to technical constraints, the very recent publication of Montes et al. (2022) was solely reviewed by the anonymous reviewer.

2.2. Reassessment of Persistence of previously suspected or identified PMOC/PM

To evaluate the relationship between PM substances and drinking water contamination, it is important to verify that the substances are truly Persistent. A number of research papers which aimed at providing evidence of proof of concept for the PM criteria proposed in Neumann and Schliebner (2019) were included in this review of data. The Arp et al. (2017) paper established an initial list of 2292 REACH-registered (potential) PMOCs. Although available in their Supplementary Information, this list was not considered in this work as the main aim was to reassess the monitoring data. Instead, we considered the sub-list of 936 PMOCs established by Schulze et al. (2018). This list includes the substances of Arp et al. (2017) that had the highest likelihood to be found in water according to Schulze et al. (2018).

2.2.1. Schulze et al. (2018)

An initial screen based on expert judgment was performed on the list of substances established by Schulze et al. (2018). As this list encompassed 936 substances, the resources to analyze them all in detail were not available. This limitation is not considered critical for this work since the reassessment and discussion focuses on the monitoring studies. If the structure was deemed unlikely to readily biodegrade (expert judgment), we conservatively considered that the P assessment performed by the researchers was correct. If the structure was deemed likely to readily biodegrade, we performed an in-depth assessment of Persistence as described in the Supplementary Information (Section S1).

2.2.2. Schulze et al. (2019)

Schulze et al. (2019) investigated a set of 64 chemicals, mostly derived from the list of Schulze et al. (2018). For the purpose of this reanalysis, Table S1 and Fig. S6 of the Supplementary Information of Schulze et al.

(2019) were used. Table S1 captured the list of the 64 selected substances, identified by both name and CAS number. Schulze et al. (2019) were able to develop adequate analytical methods for water measurements for 57 of the substances, with the remaining seven not investigated due to limitations in analytical separation methods. Monitoring of the substances was performed on 12 freshwater and groundwater samples from different locations in Germany, Spain and the Netherlands. Fig. S6 of their paper depicts the frequency of detection (max. 14) for the 43 substances detected at least once and the maximum concentration detected for each substance, reported in ranges of <0.01 µg/L, <0.1 µg/L, <1 µg/L, and >1 µg/L.

2.2.3. Schulze et al. (2020)

Schulze et al. (2020) investigated a sub-set of 15 substances selected from those previously monitored in Schulze et al. (2019). The dataset is limited to 6 samples: two surface waters, two groundwater bodies, one raw water and one drinking water in Germany. The list of chemicals is available in the Supplementary Information of Schulze et al. (2020), along with the monitored concentrations for each sampling location.

2.2.4. Montes et al. (2019)

Montes et al. (2019) investigated a sub-set of 23 substances selected from those listed in Arp et al. (2017) and Schulze et al. (2018). The dataset consists of 9 samples in surface water, 3 samples in drinking water and 3 samples of bottled water in Spain. We focused our analysis on the surface and drinking water monitoring because no substance was found in the bottles. The list of chemicals and the measured concentrations are available in the Supplementary Information of Montes et al. (2019).

2.2.5. Montes et al. (2022)

Montes et al. (2022) performed two campaigns of semi-targeted screening of over 3500 compounds on samples from 29 sites located in Portugal and Spain encompassing rivers, estuaries, and treated effluents of wastewater treatment plants. They identified a total of 343 chemicals detected at least once, of which 153 could be considered PMT or vPvM according to their own assessment against the UBA criteria. They expressed the results as “detected” or “not detected” for each substance in each of the 55 samples (information available in the supplementary information of Montes et al. (2022)). Since the number of substances investigated was high and covered by various regulations (cosmetics, pharmaceuticals, pesticides, industrial chemicals, etc.), it was not technically possible to review the Persistence of all these substances. We decided to focus on the 101 REACH-registered substances detected at least once (68 in January and 81 in June), excluding substances where biodegradation data were absent, as is often the case for intermediates for instance. It was considered relevant to focus on REACH-registered substances because these have been the most widely used to support the relevance of the PM concept (Arp et al., 2017; Arp and Hale, 2022).

2.2.6. Teychene et al. (2020)

Teychene et al. (2020) investigated the water filtration efficiency of reverse osmosis on a set of 18 substances selected from the Schulze et al. (2019) list. We reviewed this study to determine the rate of errors in the Persistence assessment of 17 of these compounds (the last one had an erroneous identifier and was removed from the analysis).

2.2.7. Neuwald et al. (2021)

Neuwald et al. (2021) performed a suspect screening of 1310 substances in 11 surface waters in Germany. These chemicals were mostly selected from the lists of Arp et al. (2017) and Schulze et al. (2018). They detected and identified a total of 64 PM candidates (list available in the Supplementary Information of Neuwald et al. (2021)). The list of substances includes two inorganic substances which were not considered further in the present paper as they fall out of the scope of PM substances.

2.2.8. Kolkman et al. (2021)

Kolkman et al. (2021) monitored a set of 32 substances in 24 samples of surface, ground and drinking water in the Netherlands and Belgium.

Twelve substances were detected in at least one sample, while another 12 substances were also detected in at least one sample by non-target analysis. The list of 32 substances was not reviewed further in this work because no unambiguous identifier (e.g., CAS number) was provided. We were however able to review the list of 12 non-target substances (identified with a CAS number) and reassess their Persistence.

2.3. Revisiting the role of Persistence and adsorption on the contamination of surface and groundwaters

A significant proportion of non-Persistent substances was found in the datasets of Schulze et al. (2019, 2020), Montes et al. (2019, 2022), and Kolkman et al. (2021). In order to determine whether PM substances have a higher likelihood to be present in surface and groundwater than non-PM substances (nPM), we used the raw data available in the publications reviewed and compared the nPM/PM distributions between the substances included in the monitoring campaigns and the substances actually detected in the samples (as given in the original publications). No monitoring campaign was conducted as part of this work and none of the data from the original publications was reassessed (detection, concentration, etc.). A non-statistically significant nPM/PM difference of distribution would mean that surface and groundwater contaminations do not depend substantially on the P property and that other factors are more important in determining the propensity of the substance to reach these waters, whether exclusively or in combination with P. This hypothesis was tested with a chi-square for the five datasets ($p < 0.05$). The chi-square is an appropriate statistical test to determine whether two frequency distributions are significantly different from each other. The chi-square is a straightforward calculation, and a simple online calculator was used (<http://testchideux.awardspace.info/?v=calcul>). To align with the publications of the researchers, a substance was conservatively considered as detected if found at least once in a sample, regardless of the number of sampling locations or the lack of information to inform on the relevance of the finding (e.g., information on the proximity of a source).

The datasets of Schulze et al. (2019, 2020), Montes et al. (2019, 2022) and Kolkman et al. (2021) allowed for the calculation and comparison of mean frequencies of detection between PM and nPM substances. Moreover, the datasets of Schulze et al. (2019) and Montes et al. (2019) also allowed for the calculations and comparisons (PM vs nPM) of mean contamination scores, calculated as the estimated maximum measured concentration (Schulze et al., 2019) or the mean measured concentration (Montes et al., 2019) multiplied by the frequency of detection for each substance. Since Schulze et al. (2019) did not report the exact concentrations in their study, the maximum measured concentrations were rounded to the upper bound of the reported ranges.

With the Schulze et al. (2019) dataset, it was also possible to determine the distributions of detected/non-detected among the PM substances as a function of log Dow. The log Dow values used (modelled with the Chemaxon software) were the ones reported in the study and were not checked because experimental data on log Dow are not publicly available. The distributions were compared by chi-square, with the hypothesis that, according to the PM concept, detection rate should increase with decreasing log Dow.

The Schulze et al. (2019, 2020), Montes et al. (2019), and Kolkman et al. (2021) datasets also allowed for determination of the rate of false positives (percentage of PM substances not found in surface, ground and/or drinking water) and/or false negatives (percentage of nPM substances found in surface, ground and/or drinking water). In addition, we have included an analysis based on the data available in Huang et al. (2021). Although the P assessment of this study was not reassessed and the substances included in the analysis are not necessarily REACH-registered, this publication provides valuable insights into false negatives as it is almost the only publication which has not preselected PM substances first but analyzed contaminants and then assessed them for their PM properties. Within each study, the detection of a substance at any level in one or more samples was sufficient to consider it as detected, in line with the

original position taken by the researchers in their papers. Yet, the significance of a detection in the context of the development of a concept aiming at regulating chemicals deserves a critical review. For this reason and where possible, we also made some comparative analysis using the 0.1 µg/L threshold defined in the Plant Protection Products regulation (EC 1109/2008) above which groundwater risk assessment is considered.

3. Results & discussion

3.1. Reassessment of Persistence of substances previously suspected or identified PMOC/PMT

In the present study, the Persistence conclusions of the substances investigated by Schulze et al. (2018, 2019, 2020), Montes et al. (2019, 2022), Teychene et al. (2020), Neuwald et al. (2021) and Kolkman et al. (2021) were reassessed and are available in the Supplementary Information. Apart from Kolkman et al. (2021) and Montes et al. (2022), these lists consisted of sub-selections of substances from Schulze et al. (2018) which was an emission ranking exercise of 1811 substances, leading to a priority list of 936 REACH-registered substances ranked by emission likelihood on the basis of REACH dossier data. These substances were all assumed to be persistent by Schulze et al. (2018), as they were concluded as Persistent and Mobile by modeling (Arp et al., 2017). When experimental data on biodegradation were available from the ECHA website or literature, this was used in the reassessment, leading to 15–40 % of the substances included in the different lists of monitored substances being reassigned as not P based on valid biodegradation tests (Table 1).

While the values reported in Table 1 for nP substances represent substances for which it was possible to conclude with certainty that they were erroneously concluded P in the original study due to uncertainty of model predictions and lack of verification of existing data, the substances concluded as P include those concluded P with a high level of certainty and those that were concluded as potentially P, as in general, insufficient data were available to definitely conclude (e.g. failed ready biodegradability test but no simulation tests available). Data from ECETOC (2021a) are also reported; the Persistence assessment methodology used by ECETOC was similar to the present study.

3.1.1. Schulze et al. (2018)

From the initial expert judgment screening, 24 inorganic substances were identified and removed from the list, leaving 912 substances to reassess. From the remaining 912 substances, around 25 % required an in-depth Persistence reassessment. Focusing exclusively on information disseminated on the ECHA website, 4 substances did not have a registration dossier and 15 % were reassessed to be non-persistent (Table 1). Most of these reassessments were concluded based on ready biodegradability data (Supplementary Information; Table S1).

Table 1

Distribution of (potentially) P and nP substances in the datasets reassessed for Persistence after exclusion of inorganic substances.

	(Potentially) P	nP	Total (concluded P by the researchers of the original publications, except ECETOC, 2021a)
Schulze et al. (2018)	777 ^a (85 %)	135 (15 %)	912 ^b
Schulze et al. (2019)	45 (79 %)	11 (21 %)	56
Schulze et al. (2020)	9 (60 %)	6 (40 %)	15
Montes et al. (2019)	15 (65 %)	8 (35 %)	23
Montes et al. (2022)	72 (71 %)	29 (29 %)	99 ^c
Neuwald et al. (2021)	51 (82 %)	11 (18 %)	62
Kolkman et al. (2021)	8 (67 %)	4 (33 %)	12
Teychene et al. (2020)	11 (65 %)	6 (35 %)	17
ECETOC (2021a)	47 (76 %)	15 (24 %)	–

^a Not all 777 substances were thoroughly re-evaluated but were initially screened on expert judgment – 85 % of P substances is likely an overestimation.

^b 24 of the 936 substances were inorganic compounds and were removed from the reassessment.

^c 2 substances were considered nP by the researchers.

3.1.2. Schulze et al. (2019)

Of the 56 substances listed in Schulze et al. (2019), four did not have a REACH dossier (acesulfame K, naphthalene-1-sulfonic acid, dimethylbenzenesulfonic acid, and 3,5-di-tert-butylsalicylic acid). Ready biodegradability test (RBT) results were retrieved from REACH dossiers of the isomers of the two sulfonic acids, which used the latter as source substance in a read-across. For acesulfame K, data concluding non-Persistence in soils are available (Biel-Maeso et al., 2019). However, Li and McLachlan (2019) showed that it is persistent in freshwater, therefore, it was concluded as P. For 3,5-di-tert-butylsalicylic acid, no data were found in other REACH dossiers or in the literature. Based on its chemical structure (presence of tert-butyl groups on an aromatic ring), it was not expected to be concluded as readily biodegradable. The closest related substance identified was 4-tert-butylbenzoic acid which was not readily biodegradable; however, the structure lacks both the hydroxy- and a tert-butyl group compared to 3,5-di-tert-butylsalicylic acid. Hence, 3,5-di-tert-butylsalicylic acid was conservatively considered as (potentially) persistent.

Almost all REACH dossiers of the 52 remaining substances included a ready biodegradability test performed with the registered substance (see Supplementary Information; Table S2). A discrepancy was observed for toluenesulfonamide, for which Schulze et al. (2019) monitored both ortho- and para- isomers simultaneously and reported a combined concentration. According to their respective REACH dossiers, toluene-4-sulfonamide (para) is readily biodegradable (OECD 301D) whilst toluene-2-sulfonamide (ortho) failed an OECD 301C test. However, it has been shown that the OECD 301C is the most stringent of the six test systems of the OECD 301 series (Dick et al., 2016; Takekoshi et al., 2021). In addition, the OECD 301D performed on the para- congener showed microbial inhibition at 4 mg/L. Since OECD 301C uses initial concentrations of 100 mg/L and does not offer any option to mitigate toxicity, it is highly likely that this concentration inhibited mineralization. Therefore, the ortho-congener is likely readily biodegradable and was considered unlikely to be Persistent.

Following the reassessment of the biodegradation data, 12 substances (21 %) were concluded as not persistent (Table 1). The relative proportions of (potentially) P and nP substances within the set of 56 monitored substances detected above 0.1 µg/L were not statistically different (Table 2). The average contamination scores and the average detection rates were almost identical between P and nP substances (Tables 4 and 5). The average detection rates and contamination scores of persistent substances tended to remain constant or slightly decrease with decreasing log Dow (Table 6).

3.1.3. Schulze et al. (2020)

Of the 15 substances captured in the list of Schulze et al. (2020), only acesulfame K and the two dimethylbenzylsulfonic acids did not have a REACH dossier (see Supplementary Information; Table S3). Six substances (40 %) had experimental data for which a non-Persistent conclusion could be drawn (Table 1). Considering all sampled waters together, the researchers detected 9 substances in at least one sample. The distributions of PM/nPM substances in the set of 15 monitored and the set of 9 detected

Table 2

Comparison of the distribution of (Potentially) Persistent and non-Persistent substances among all substances investigated and all substances detected at any concentration or above 0.1 µg/L at least once. The chi-square tests indicate that the distributions are not significantly different. Detection in surface or groundwater is independent from the PM properties.

		(Potentially) P	nP	Total	Chi-square	p-value
Schulze et al. (2019)	Number of substances investigated	45 (80 %)	11 (20 %)	56		
	Number of detected substances	33 (79 %)	9 (21 %)	42	0.047	0.828
	Number of detected substances >0.1 µg/L	14 (74 %)	5 (26 %)	19	0.376	0.540
Schulze et al. (2020)	Number of substances investigated	9	6	15	–	–
	Number of detected substances	5	4	9	0.046	0.830
Montes et al. (2019)	Number of substances investigated	15 (65 %)	8 (35 %)	23	–	–
	Number of substances detected in surface water	11 (65 %)	6 (35 %)	17	0	1
	Number of substances detected >0.1 µg/L in surface water	2 (33 %)	4 (67 %)	6	1.994	0.158
	Number of substances detected in drinking water	2 (40 %)	3 (60 %)	5	1.096	0.295
	Number of substances detected >0.1 µg/L in drinking water	0	0	0	–	–
Montes et al. (2022)	Number of substances investigated in January	48 (71 %)	20 (29 %)	68		
	Average number of substances detected in January	11 (58 %)	8 (42 %)	19	1.096	0.295
	Number of substances investigated in June	58 (72 %)	23 (28 %)	81		
	Average number of substances detected in June	15 (66 %)	7 (34 %)	22	0.098	0.754

substances were not statistically different (Table 2). Among the 9 monitored PM substances, 4 were not detected in any source of water (false positive rate of 44 %).

3.1.4. Montes et al. (2019)

All of the 23 substances of the list of Montes et al. (2019) had already been assessed in the above-mentioned studies. Considering all sampled waters together, the researchers detected 17 substances in at least one sample (see Supplementary Information; Table S4). The distributions of PM/nPM substances in the set of the monitored and the set of the detected substances were not statistically different (Table 2). False positive rates, mean frequencies of detection and contaminations scores are reported in Tables 4, 5, and 7.

3.1.5. Montes et al. (2022)

On the basis of the ready biodegradability studies found in the REACH dossiers, we found 29 non-Persistent substances in the set of 101 substances detected at least once in the 55 investigated samples, only two of which (caprolactam and diethylphthalate) had also been considered non-Persistent by Montes et al. (2022). For more information, see Supplementary Information Tables S9 and S10.

3.1.6. Teychene et al. (2020)

One substance (2-toluenesulfonic acid) had a mismatch between its name and the chemical formula presented in the study. This substance was excluded from the analysis. All the 17 remaining substances had already been assessed in Schulze et al. (2019). For more information, see the Supplementary Information; Table S5.

3.1.7. Neuwald et al. (2021)

Fifty-two of the 62 substances reassessed had a REACH dossier. For some substances with no REACH registration, biodegradation information was retrieved from the literature. Four substances did not have any REACH or literature data available. Three (3,5-di-tert-butylsalicylic acid, bis(trifluoromethyl-sulfonyl)imide, tetracyanoborate) were considered Persistent or potentially Persistent based on their chemical structures (expert judgment). We were unable to assess the Persistence of olmesartan by expert judgment and conservatively concluded it as Persistent. Eleven non-Persistent (18 %) substances were identified among the 62 monitored substances (Table 1). For more information, see the Supplementary Information; Table S6.

3.1.8. Kolkman et al. (2021)

Nine of the 12 non-targeted substances detected and identified in the study by Kolkman et al. (2021) had a REACH dossier or could be assessed based on read-across from another REACH-registered substance presenting structural similarities (positional isomers). For the remaining 3 substances,

biodegradation information was retrieved from the literature, except for the substance metoprolol, for which no relevant information was found, and hence was conservatively concluded as Persistent. Among the 12 detected substances, 4 substances were concluded as non-persistent based on experimental data (Table 1). For more information, see Supplementary Information; Table S7.

3.1.9. Conclusions of the Persistence reassessment

The reassessment of the Persistence conclusions in the seminal work by Arp et al. (2017) and Schulze et al. (2018) revealed significant discrepancies when compared to available data. The examination and inclusion of experimental data available in the disseminated REACH dossiers and the literature leads to the identification of approximately 15 % of not persistent substances (most of which were found to be readily biodegradable) out of the 912 prioritized organic substances (Table 1). Twenty-four inorganic substances were identified, a chemical class for which Persistence assessment is not considered under REACH, and there are also well-known positive reference substances for biodegradability testing in the OECD TG 301 series, like sodium benzoate and various citrates and acetates, present in the list. These substances were also considered as PMOCs and ranked in Schulze et al. (2018). Although it is logical that the original screening was done using automated methods, the P conclusions from Arp et al. (2017) were meant to also use available experimental data and prioritized experimental data over estimation methods. More importantly, although Arp et al. (2017)'s original work was a screening and specifically mentioned that some of the assessments were to be used with caution, subsequent research did not reassess the conclusions before proceeding to the monitoring campaigns. As is normal practice in science, other researchers have already cited these publications as a reference list for the identification of Persistent and Mobile substance to interpret their observations. For example, Gustavsson et al. (2022) considered polyethylene glycol (CAS: 25322–68-3), *N,N*-bis(carboxymethyl) alanine (CAS: 164462–16-2) and poly(oxy-1,2-ethanediy)l, *n*-decyl-hydroxy (CAS 68439–46-3) as the most widely emitted Persistent and Mobile substances because they were listed as PMOCs in Arp et al. (2017). Yet, according to the experimental data available in their respective REACH dossiers, all these substances are not Persistent (based on readily biodegradable results from OECD 301 tests or weight of evidence). Similarly, Montes et al. (2019) considered methyl sulfate, xylene sulfonic acid or caprolactam and several others as Persistent, while those substances are also readily biodegradable. As many researchers (e.g., Schulze et al., 2019, 2020; Montes et al., 2019, 2022; Teychene et al., 2020; Neuwald et al., 2021; Kolkman et al., 2021) based their first hypothesis on the list provided in Arp et al. (2017), it would be prudent to reconsider the conclusions of these publications, in particular if they are considered as key publications for the justification of regulatory action like the inclusion of PMT/vPvM hazard classes under the CLP regulation (and UN-GHS subsequently).

3.2. Revisiting the role of Persistence on the contamination of surface and groundwater

Once the Persistence reassessment was completed, the proportions of PM and nPM substances in the original set of chemicals selected for monitoring was compared to the proportions found in the subset of detected substances to determine if Persistent substances had a higher detection likelihood and exhibited higher levels of contamination than non-Persistent substances in the studies (assuming all the substances were correctly evaluated for Mobility).

The Schulze et al. (2019) study was considered as a proof of concept for the PM approach as proposed in Neumann and Schliebner (2019): substances a priori identified as having intrinsic PM properties combined with a high emission score would be detected by monitoring a posteriori in the environment. Schulze et al. (2019) monitored 56 substances identified as PM by Arp et al. (2017) in five European locations, for a total of 14 samples per substance. Since 42 substances were detected in at least one sample, approximately half of which for the first time, the researchers concluded that their work supported that the PM hypothesis was predictive of surface and groundwater contamination. Similarly, Schulze et al. (2020) monitored 17 substances identified as PM by Arp et al. (2017), of which two were inorganics. Out of the 15 organic substances, 9 were detected in surface, ground and/or drinking water.

The presence of these substances within the monitoring program allowed for an evaluation of the role of chemical Persistence in water contamination by comparing the monitoring data available for PM substances against that of nPM substances. The exercise did not include a Mobility assessment and considered that all the substances of the datasets met at least the M criterion used by the researchers (log Dow or log Koc < 4). First, both datasets of Schulze et al. (2019, 2020) showed that the distributions of nPM and PM substances in the subsets of detected and monitored chemicals were not statistically significantly different (Chi-square test, $p > 0.05$; Table 2). This strongly suggests that the likelihood of detection of these substances in water is independent of their PM property. In other words, the P property did not play a major role in the presence of those chemicals in water, nor did the PM properties in extenso.

This conclusion is also in agreement with the monitoring dataset from Huang et al. (2021), who collected surface and groundwater monitoring data in China from 75 different studies which focused on 432 pharmaceuticals and personal care products (PCPPs). They divided their dataset into three categories: “not-PMT/vPvM”, “potential PMT/vPvM” and “very likely PMT/vPvM” (Table 3). The substances were mostly non-REACH registered therefore we could not reassess Persistence nor Mobility, but did the analysis based on the conclusions of the authors. The statistical analysis shows that the distributions of these three categories within the total set of substances, the substances detected in surface water or those detected in groundwater are not different from each other (Table 3).

Finally, our conclusion is also in agreement with the findings of ECETOC (2021a) in which the researchers analyzed an extensive dataset comprised of 62 substances monitored in surface and groundwater bodies of Great Britain (5000 datapoints). The survey included a comparison of nPM vs PM substances with regard to detection rates of substances in ground and surface waters. There was no evidence of any correlation between Persistence and Mobility and detection of chemicals in surface water or groundwater. In a study of 60 groundwater samples in Switzerland, Kiefer et al. (2021) performed a screen based mostly on the

list of PM substances developed by Schulze et al. (2018). For industrial chemicals, they found high concentrations of the readily biodegradable benzenesulfonic acids and p-toluenesulfonic acid. Similarly, in Gustavsson et al. (2022), the PM substances most widely emitted in the environment were readily biodegradable. In Montes et al. (2022), in a set within which 29 % of the substances were readily biodegradable, 40 % of the top 10 most frequently detected compounds were readily biodegradable. Taken altogether and based on the criteria proposed for inclusion into the CLP regulation, the data strongly suggest that Persistent and Mobile substances do not have a higher potential than non-Persistent but Mobile substances to reach drinking water sources, contrary to the suggestion made in Arp and Hale (2022). Based on the re-analysis of the data available, the PM property does not effectively discriminate between substances that may and those that may not reach drinking water sources. Other factors are likely key in determining the propensity of a chemical to reach those sources such as the scale of emissions or the proximity of sources. Those potentially determining factors are discussed in the next sections.

3.2.1. Do PM substances accumulate in the “water cycle”?

It is often argued that contrary to non-persistent and mobile substances, which degrade rapidly, persistent and mobile substances will accumulate in the water “cycle” (Neumann and Schliebner, 2019; Hale et al., 2020a; Rüdell et al., 2020). It is however not explained in these publications the mechanisms by which a PM substance – of concern due to its ability to pass through natural and man-made barriers and hence contaminate sources of drinking water – would immobilize in drinking water so that its concentration would increase over time. An increase in concentration may be seen following an increase in emissions, but an increase due to the substance accumulating in drinking water would contradict the definition of a PM substance. In any case, if the accumulation assumption was correct, PM substances should display higher levels of contamination than nPM substances. Our reassessment of the data from Schulze et al., 2019 shows that the nPM/PM distribution of the substances detected above 0.1 µg/L is not statistically different from that of the monitored dataset (Table 2). This observation agrees with the analysis of a British monitoring survey by ECETOC (2021a) where very similar proportions of PM and nPM substances were detected at least once above 0.1 µg/L in surface water (approx. 70 % of the monitored PM and nPM substances). Likewise, in this survey, very similar proportions of PM and nPM substances were detected at least once above 0.1 µg/L in groundwater (approx. 80 %). The datasets of Schulze et al. (2020) and Montes et al. (2019) also show no indication that average detection rates, detection above 0.1 µg/L or contamination scores could be higher for PM than for nPM substances (Tables 2, 4, and 5). Taken altogether, the data supports a lack of correlation between the PM properties and the assumed accumulation of substances in surface and/or groundwater. Furthermore, it should be kept in mind that the number of substances in the dataset with a Persistent conclusion are over-represented, and as data become available, it is likely that the proportion of substances with non-P conclusions would increase, further emphasizing the lack of correlation identified above.

These observations are again in agreement with the analysis of the British monitoring survey by ECETOC (2021a) where very similar proportions of PM and nPM substances (where log Koc < 3 was used for M criteria) were detected above 0.1 µg/L in at least two thirds of the samples of surface water (approx. 15 %). For groundwater, the PM proportion (27 %) was however higher than the nPM proportion (15 %). Taken altogether, our

Table 3

Comparison of the distribution of PMT/vPvM properties (as defined in the original publication from Huang et al. (2021)) among all substances, among all substances detected in surface water and among all substances detected in groundwater in Huang et al. (2021). The chi-square tests indicate that the distributions are not different. Detection in surface or groundwater is independent from the PM properties.

	Not PMT/vPvM	Potential PMT/vPvM	Very likely PMT/vPvM	Chi-square	p-value
Total substances investigated	72 (17 %)	203 (47 %)	152 (36 %)		
Detected in surface water	19 (14 %)	65 (47 %)	53 (39 %)	0.848	0.65
Detected in groundwater	11 (11 %)	46 (44 %)	47 (45 %)	4.362	0.11

Table 4

Comparison of the average contamination score of PM vs nPM substances according to the datasets of Schulze et al. (2019) and Montes et al. (2019). The average scores between PM and nPM substances are qualitatively similar or higher for the nPM substances, showing no evidence of correlation between contamination score and Persistence.

	Average contamination score ($\mu\text{g/L}$)	
	(Potentially) PM	nPM
Schulze et al., 2019	0.39	0.62
Montes et al., 2019 (surface water)	0.003	0.025
Montes et al., 2019 (drinking water)	0.0004	0.0018

Table 5

Average detection rates of PM and nPM in various sources of natural surface and groundwater bodies in several PM monitoring studies. These values are qualitatively similar or higher for the nPM than for the PM substances, showing no evidence of correlation between contamination severity and Persistence.

	Average detection rate within each category	
	(Potentially)PM	nPM
Schulze et al., 2019	44 %	58 %
Schulze et al., 2020	21 %	28 %
Montes et al., 2019 (surface water)	27 %	46 %
Montes et al., 2019 (drinking water)	2 %	11 %
Kolkman et al., 2021	11 %	23 %

analysis strongly suggests that PM substances do not have any potential to accumulate to higher levels compared to nP substances in surface or groundwater.

3.3. Revisiting the role of Persistence combined with Mobility on the contamination of surface and groundwater

To further analyze the robustness of the PM concept, a comparison of the percentage of detects of (potentially) Persistent substances according to their log Dow, a parameter used by Schulze et al. (2019) to assess M, was conducted (Table 6). Different log Dow cut-offs were tested in order to determine whether adsorption potential contributed to the detection rate. A chi-square was used on the distribution of detects vs non-detects to check for any significant trend, and the results indicate that the “detects / non-detects” distribution is not different than the distribution of “P + log Dow < 3.5”.

Substance occurrence (% detects and detection rate) in water does not increase with decreasing adsorption potential, which was an observation also made by ECETOC (2021a) for Persistence and log Koc. These observations contradict the PM concept hypothesis according to which lack of/low affinity for soil organic carbon is the major driver of water contamination if the substance is persistent.

This was further demonstrated by the extension of this analysis to multiple reviews in which the log Dow was reported by the researchers (Schulze et al., 2019, 2020, Huang et al., 2021). In all three studies, the data indicate very high rates of false positives (substances considered PM after our correction of the P assessment and not detected in surface and/

or groundwater) (Table 7). Although in a general assessment, some of these false positives may be due to low emission rates and/or insensitive analytical methods, the substances analyzed by the researchers were specifically selected for their high emission rates (REACH data or wide-dispersive use) and specific analytical methods were developed with limits of detections down to the picogram (Schulze et al., 2019). Therefore, those two aspects were not limiting and did not contribute significantly to the rate of false positives.

3.4. What about the emitted PM substances that are not present in surface and groundwater?

Neumann and Schliebner (2019) defined false positives as the substances fulfilling both P and M criteria and not detected in a monitoring program. These researchers estimated a false positive rate of 25 % from a single monitoring study (Schulze et al., 2019). They considered it sufficiently low for proof-of-concept of PMT, even though Schulze et al. (2019) had monitored the substances with emission scores among the highest in Europe, hence greatly increased their probability to find at least traces of all substances in water. From our own analysis, out of the 56 substances, only 45 substances actually fulfil the PM criteria, among which only 33 were detected at least once in water, making the false positive rate 27 % (substances fulfilling the PM criteria but not found in water), which is slightly higher than originally provided in Neumann and Schliebner (2019). Moreover, even higher false positive rates were found when considering the data of Schulze et al. (2020) where 67 % and 56 % of the PM substances were not detected in surface and groundwaters, respectively. This is taking into consideration that a single detection at any concentration level is sufficient to consider the substance as “detected”. However, in European legislations regulating water quality such as the DWD or the Water Framework Directive (Directive 2000/60/EC), significant detection is defined by a complex framework. Reporting under the WFD implies a number of samples to be taken over an agreed period of time and the data points span several years. A single detect is not sufficient to consider the substance to be a priority. The Joint Agency Groundwater Directive Advisory Group (JAGDAG) considers that significant monitoring data must represent a concentration above the limit of quantification in 5 % of the samples or that the substance is found in a significant proportion of the sampling sites (JAGDAG, 2017). This is necessary to be able to determine if the contamination is driven by point source emissions in the vicinity of the sampling location or by a wide contamination. This also highlights the need for more specific monitoring campaigns which would allow for a better understanding of the fate of the substances by providing enough information to link the source of the emission and the presence/concentration in drinking water. The methods used up to now do not permit to do this link which would be valuable to better address the concern. Therefore, it may be possible that the number of false positives would be even higher taking into consideration the criteria of the DWD and/or the WFD. For instance, in Huang et al. (2021), 67 % and 74 % of the potential or very likely, respectively, PMT/vPvM PPCPs were not found in surface and groundwaters. Thus, false positive rates were within 29 %–67 % in Western Europe for REACH-registered substances and up to 67 %–74 % for PPCPs in China. Furthermore, for the majority of substances re-assessed, only a “potentially P” conclusion could be drawn, and Mobility was not reassessed in the present work. Considering that these studies purposefully selected substances with the highest and widest emissions, and sampling locations were generally in the vicinity of

Table 6

Trends of the percentage of detects, average detection rate and contamination score of P substances according to log Dow (data from Schulze et al., 2019). Log Dow cut-offs are arbitrary.

	Number of detects	Number of non-detects	% Detects	Chi-square	p-value	Average detection rate	Average contamination score
Number of P and log Dow < 3.5	33	12	73 %			44 %	0.39
Number of P and log Dow < 0	24	7	77 %	0.164	0.686	42 %	0.44
Number of P and log Dow < -2	17	5	77 %	0.122	0.727	41 %	0.44
Number of P and log Dow < -3	7	4	64 %	0.407	0.523	30 %	0.36

Table 7

Percentages of false positives in different studies (i.e., % of substances considered PM in the present work and not found in different kinds of water).

	Schulze et al. (2019)	Schulze et al. (2020)	Montes et al. (2019)	Huang et al. (2021)
Total PM substances investigated	45	9	15	355
Not detected in any water investigated (false positives)	27 %	44 %	27 %	Not determinable
Not detected in surface water (false positives)	Not determinable	67 %	27 %	67 %
Not detected in groundwater (false positives)	Not determinable	56 %	Not relevant	74 %
Not detected above 0.1 µg/L	69 %	Not relevant	87 %	Not determinable

wastewater treatment plants, the proportion of (potentially) PM substances not detected in the environment is remarkable. This strongly suggests that for substances with lower emission rates, which represent the vast majority of the REACH-registered substances, false positive rates could be even higher.

Schulze et al. (2019) ranked 936 substances by importance of emission potential (rank 1 was for the substance most emitted). Schulze et al. (2019) chose a subset and did not investigate any substance ranked below 699. In the present study, the score of non-detected substances is considered zero. Acesulfame K was considered an outlier because of its very wide use in consumer goods. Trifluoroacetic acid was considered an outlier because it is a likely degradation product of a multiplicity of precursors. Those sources of contamination could not be considered by Schulze et al. (2019). Plotting the average contamination score of all the other PM and nPM substances considered together as calculated in this work against the emission score ranking from the data of Schulze et al. (2018) shows that when a substance is detected in water, this is mostly because of its production tonnage, emission patterns and uses, regardless of its intrinsic properties (Fig. 1). The demonstration that the contamination score derived in this work is mainly driven by the emission patterns confirms the hypothesis made by ECETOC (2021a) that emissions and vicinity to emissions sources are driving the detection rate of chemicals in surface water. Conversely, a widely emitted substance will not necessarily be found in water in all cases.

4. Conclusions

The PM concept aims at proactively identifying substances that have the highest potential to both reach sources of drinking water and accumulate in the drinking water cycle if emitted into the environment (Neumann and Schliebner, 2019; Hale et al., 2020a). Using monitoring data from surface water, ground water and drinking water to assess the contaminants for properties in common, these researchers have defined a set of criteria based partly on existing regulations. Chemicals meeting these criteria would be expected to be detected in water. False positives (substance meets the criteria but is not detected in water) may be due to the chemicals being used in low amounts or in controlled conditions, and/or to the limits of analytical methods. However, the monitoring studies used in this work were not limited by low-rate emissions as the researchers purposefully selected high emission chemicals (Schulze et al., 2018) or lack of analytical

capability as the researchers developed highly sensitive analytical methods (down to picograms) for each chemical analyzed (Schulze et al., 2019). When existing and newly generated and targeted monitoring data were used to test this hypothesis, the results indicate the inadequacy of the hypothesis. It can be estimated that at least 29 % and maybe >75 % of the PM substances of the chemical space emitted into the environment are actually not detected in surface water, which questions the relevance of such a simplistic approach to solve a complex issue to be included within a regulatory framework at this time.

In addition, the reanalysis of the available data demonstrates that:

- PM substances do not have a higher likelihood than non-PM substances to be detected in surface, ground- or drinking water;
- Among substances fulfilling any M cut-off criterion of sorption-based metrics (Dow or Koc), P substances do not have a higher likelihood to be found in water than nP substances;
- Among these M substances, the severity of contamination is not higher for P substances than for nP substances.

Finally, the reanalysis did not provide any evidence of log Dow or log Koc as drivers of chemical contamination in waters.

Using the available data from the previous works mentioned, the present study shows that PM substances as defined by the UBA-proposed criteria do not have an increased potential to reach sources of drinking water and do not appear to accumulate in the water cycle compared to nPM substances. These criteria do not adequately discriminate between the substances that may and those that may not contaminate drinking water sources. At the very least other factors need to be considered in addition to Persistence (and Mobility) such as scale of emissions and proximity of sources. This confirms the findings of ECETOC (2021a) which highlighted that chemical contamination in water does not depend on the Persistence and Mobility criteria that are being proposed for inclusion into the CLP. The analysis presented in this study greatly suggests that ECETOC (2021a) was correct in hypothesizing that emissions and vicinity to emissions sources are driving the detection rate of chemicals in surface water (Fig. 1). The PM concept was designed on the basis of numerous datasets encompassing hundreds of substances and thousands of datapoints published in several publications. Most of these studies explicitly concluded they did not identify any threat to human health from these contaminations (Stackelberg et al., 2007; Loos et al., 2010; Tröger et al., 2018; Huang et al., 2021). However, Lapworth et al. (2012) and Bunting et al. (2021) expressed concerns about the possible growing concentrations of some chemicals in groundwater in the future. Even some of the studies reviewed as part of this work concluded that the criteria as proposed may not be fit for purpose based on their findings (Neuwald et al., 2021). Based on these previously expressed concerns that emission rates and/or emissions vicinity are key factors, future research on the false positives and negatives could benefit the protection of the sources of drinking water through the targeted reduction of pollutant emissions for example. Furthermore, currently, exposure models (e.g., leaching via riverbank filtration) are lacking to conservatively ensure that drinking water sources are safely protected from unacceptable chemical concentrations. Future research should focus on assessing transport of contaminants in selected locations in order to characterize transport flux to more effectively predict the concentrations in the different environmental compartments, in particular groundwater.

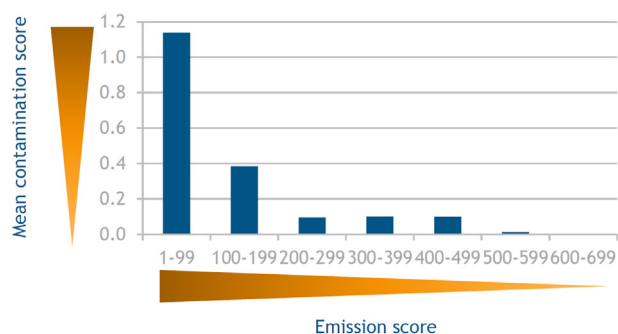


Fig. 1. Average contamination score as calculated in this work from the data of Schulze et al. (2019) against emission score ranking from the data of Schulze et al. (2018).

The Cefic LRI project ECO54 (ECETOC, 2022) aims at closing the gap by developing such models, which could be implemented in regulatory risk assessment tools for a better management of chemicals with regards to the protection of drinking water sources.

Under CLP, as under GHS, hazard classification is independent from exposure considerations (production tonnage, uses, emission patterns, etc.). In addition, the European downstream regulations tend to regulate solely on a hazard basis. The European Commission considers including PMT/vPvM substances as Substances of Very High Concern, which could be generically banned from consumer products. The regulatory consequences of the PM concept are thus mostly hazard-based and do not take exposure into consideration. It can be assumed that the number of PMT/vPvM classified substances which are actually not present in waters – and thus of no concern – could be in the hundreds, if not thousands. At best, an identification as PMT/vPvM could be a screening for further analysis of the potential for drinking water contamination through risk assessment tools and modeling as developed in Pawlowski et al. (2022). Although, the precautionary principle requires Europe to address any uncertainty related to the contamination of European waters, all the available science, including this analysis, should still be weighed in order to increase the likelihood of success of the policy measures proposed.

This work shows that the criteria have a low predictability for detecting drinking water contaminants.

The lack of robustness is of concern for at least two reasons. First substances of no concern may be prioritized for regulatory action. Second and more importantly, potential drinking water contaminants may not be identified by the proposed approach. In consequence there is a true concern regarding the alignment of the policy proposal with the protection goal.

Credit authorship contribution statement

Marie Collard conceptualized, designed, and planned the review and coordinated the work with the anonymous reviewer. She also analyzed the data, developed figures and tables and wrote the original draft.

Pippa Curtis-Jackson provided a validation of the reassessment and provided a thorough review of the manuscript.

Delina Lyon coordinated the review and acted as project manager as well as funded the external review of the data for validation. She also provided through reviews of the draft and final manuscripts.

Louise Camenzuli, Nathalie Vallotton and David Saunders provided support in the conceptualization and execution of the planned review. They also participated in the writing of the manuscript and the review of it.

Data availability

The data is available in the Supplementary Information.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.161228>.

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