A tiered assessment of human health risks associated with exposure to persistent, mobile and toxic chemicals via drinking water

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HIGHLIGHTS

- We present a tiered human health drinking water risk assessment for PMT substances
- Tolerable risk margins shown for all substances when water treatment is in place
- Higher tier spatially-resolved exposures used a 1 km gridded routing model
- Highest risk predicted in highly populated areas with low dilution under low flow
- Overall, human health risks posed by the 22 PMT substances evaluated are low

GRAPHICAL ABSTRACT
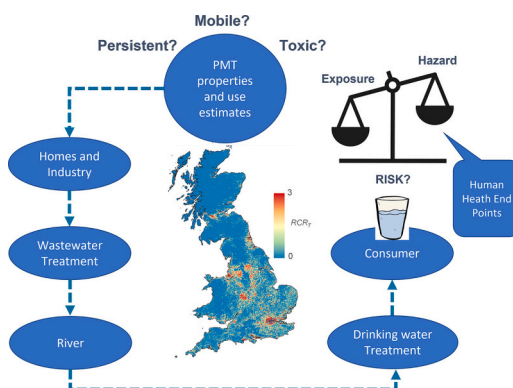
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ABSTRACT

There is increasing interest in chemicals which are persistent, mobile and toxic (PMT), primarily to protect drinking water. We present a tiered assessment of drinking water exposure and associated human health risks for 22 PMT substances. Worst-case exposure via drinking water is assumed to occur when wastewater is discharged to rivers which are then abstracted for water supply. Screening-level exposures assume daily *per capita* emissions based on REACH tonnage estimates, with removal in wastewater treatment calculated using SimpleTreat and no

riverine dilution. Removal in water treatment was estimated for each substance assuming either conventional or advanced treatment processes. Higher tier spatially-resolved exposures used a gridded routing model which transfers chemical through the landscape based on flow directions derived from a 1 km digital elevation model. Emission was assumed to be proportional to population and no in-stream degradation was assumed. Exposures were calculated for 296 locations containing drinking water treatment works (WTWs) under mean discharge and Q_{95} (discharge exceeded 95% of the time). At low tiers, risk characterisation ratios (RCRs) were < 1 for all substances, assuming realistic tonnage and conventional treatment. If drinking water is assumed to represent only 20 % of total chemical intake, total RCRs (RCR_T) were > 1 for three substances under conventional treatment but were < 1 for all substances under advanced treatment. Highest exposure and RCRs were predicted in highly populated areas with low dilution. RCR_T values were > 1 for tetrachloroethylene (highest RCR) at up to 18 % of WTW locations under Q_{95} conditions assuming conventional treatment. However, RCR_T was <1 for all locations assuming advanced treatment. Actual exposures will depend on catchment characteristics, but the model usefully allows prioritising higher risk chemicals and WTWs. Overall, the substances evaluated are unlikely to currently pose health risks, provided an appropriate level of water treatment is employed.

1. Introduction

In recent years, there has been an increasing regulatory interest in the adoption of a new chemical hazard class identifying persistent (P), mobile (M) and toxic (T) substances (PMT), and very persistent (vP) and very mobile (vM) substances, primarily with the aim of protecting drinking water supply (Neumann and Schliebner, 2019; ECETOC, 2021). The German Environment Agency (UBA) and the Norwegian Technical Institute (NGI), for instance, have funded several research projects, published reports and held workshops to develop screening criteria for the identification of PMT and vPvM substances (Neumann and Schliebner, 2019; Arp and Hale, 2023; Arp et al., 2023; Arp and Hale, 2022; Hale et al., 2022a, 2022b). The primary goal of the screening criteria is to prioritise chemicals that have the potential to represent a risk to human health as a result of their ability to reach drinking water sources. Consequently, PMT and vPvM have been proposed as additional hazard classes under the European Union's REACH (Registration, Evaluation, Authorisation and restriction of CHemicals) regulation and have recently been incorporated into the EU CLP (Classification, Labelling and Packaging) regulation (Regulation (EC) No 1272/2008) due to their potential threat to water resources. They are considered by many as of *equivalent concern* to persistent hydrophobic organic chemicals that have the potential to bioaccumulate (e.g. Hale et al., 2022a, 2022b). Indeed, in 2020, the European Commission published its 'Chemicals Strategy for Sustainability Towards a Toxic-free Environment' (European Commission, 2020), which commits to the introduction of PMT and vPvM as categories of substances of very high concern (SVHC). Given the vast array of chemicals used in commerce (of the order of tens to hundreds of thousands: Brown and Wania, 2008), hazard-based screening clearly represents an important prioritization step, which can greatly help to focus limited resources on assessing and managing the risks associated with substances of highest concern. Proponents of the PMT/vPvM approach argue that substances which meet the P and M screening criteria have a higher potential to reach drinking water sources because they can pass through natural barriers like soil and rock matrices and have a lower probability of removal during wastewater treatment since they are, by definition, poorly degradable and have low to moderate hydrophobicity. They also have a greater propensity to 'breakthrough' standard drinking water treatment because of their physico-chemical properties (Rüdel et al., 2020), although their potential for removal by advanced treatment processes such as chlorination and ozonation is rarely, if ever, assessed. PMT substances, therefore, represent 'hazards' which could exceed toxicological levels of concern now or at some point in the future (Cousins et al., 2019; Scheringer et al., 2022).

The criteria adopted by the European Commission (2022) for designating PMT substances under the CLP regulation include a degradation half-life in freshwater >40 days for P (>60 days for vP) and a log (K_{OC}) < 3 for M (<2 for vM), with chemicals classed as toxic if they, *inter alia*, are designated as carcinogenic, mutagenic, reprotoxic or endocrine disrupting (European Commission, 2022). These hazard criteria are relatively crude – capturing a very wide array of chemicals, many of which are unlikely to cause harm to wildlife or humans at current exposures. Moreover, these criteria do not account explicitly for the likelihood of chemical removal in drinking water treatment trains.

The alternative to screening chemicals based on hazard criteria (such as PMT and vPvM) is to perform a risk assessment, in which exposure is estimated and compared to a toxicological threshold of concern. This is supported by the World Health Organisation's (WHO) drinking water safety plan principles in assessing threats to drinking water quality (WHO, 2023). A key challenge in this process is the acquisition of data to allow human exposure to chemicals in drinking water to be quantified accurately, largely because of significant uncertainties in emissions. However, we argue here that a tiered approach to exposure assessment in a risk-based screening procedure is possible even when emissions are uncertain. This could better-facilitate the identification of substances of real concern compared with only applying the PMT criteria currently proposed in Europe.

In 2020, the UK regulator, the Drinking Water Inspectorate (DWI), critically reviewed 58 substances which met the PMT criteria adopted by the European Commission (2022) and which were prioritized as potential PMT or vPvM candidates by Neumann and Schliebner (2019). They conducted a preliminary exposure assessment for source waters used for public drinking water supply (DWI, 2020). However, estimates of usage were highly uncertain and conservative (based on the upper bound of REACH tonnage bands) and estimates of removal fractions in water treatment processes were very approximate. In addition, no attempt was made to estimate spatial exposure patterns and identify areas with high potential for elevated concentrations, which may be associated with higher risk. In this paper, we present a more rigorous evaluation of the human health risks associated with PMT and vPvM substances via the consumption of drinking water in Great Britain, with a particular focus on refining estimates of exposure for England and Wales. Specifically, we attempt to estimate (i) the concentrations of PMT substances in surface waters which might be abstracted for drinking water supply; (ii) the likelihood of removal in different drinking water treatment trains; (iii) the risk to human health arising from exposure via drinking water abstracted from different locations and with different water treatment technologies and (iv) the spatial distribution of environmental exposure in Great Britain, with a particular focus on England and Wales, and the potential drivers for high concentrations in untreated water.

2. Methods

2.1. Substance selection and review

A tiered assessment was conducted for 22 of the 58 chemical substances considered by DWI (2020). Some substances were excluded from the original list if either (i) their REACH tonnage was <10 t a^{-1} or (ii) they were already under regulatory scrutiny and, therefore, were being

considered via separate assessments. For example, many poly- and perfluoroalkylated substances (PFAS) are considered to meet the PMT criteria and represent high priority substances for monitoring and management. However, they are currently being evaluated elsewhere. Five additional substances were included because they were reported in surface water monitoring with high frequency of detection or were identified by Arp and Hale (2019) as having high priority for follow up. These were 1,3-dichlorobenzene, APN, N-butylbenzenesulphonamide, saccharin and triclosan. Of these, triclosan is probably not strictly a PMT substance (although it is listed as vPvM in Arp and Hale, 2019) because it has a log(K_{OC}) > 3 and has been reported to degrade rapidly in river water (Sabaliunas et al., 2003). However, it does present a potential risk due to its toxicity and reported occurrence in surface waters (e.g. Cap-devielle et al., 2008). In addition, hexamethyldisiloxane and tricarbonyl (methylcyclopentadienyl) manganese were originally classified as mobile by DWI (2020) because they used a log K_{OC} threshold of 4 rather than 3. All other substances have log K_{OC} values <3 (see Supplementary Materials Table S1 for further details of chemical properties). Persistence classifications were derived from DWI (2020). Note that trichloroethylene was classed as not persistent by DWI (2020) but was classed as P by Berger et al. (2018). For those chemicals not originally included by DWI (2020), we report the P classifications designated by Arp and Hale (2019). Many more substances will have properties that meet the PMT/vPvM criteria. The list of substances evaluated in this study (Table 1) represents a subset of all this more complete set of substances, which is used to exemplify the approach.

At all stages we assume *a priori* that surface water abstraction from rivers poses the highest potential risk and that the primary route of emission is via wastewater discharge to rivers. All wastewater was assumed to be treated by a minimum of secondary treatment (activated sludge), but with negligible biodegradation during treatment. In all cases, *per capita* use was calculated from assumed tonnage using a population of 67.22 million for the UK (Office for National Statistics, 2021). A value of 150 L cap^{-1} d^{-1} was assumed for domestic water use (Statista, 2022). This value is low compared to many actual wastewater treatment plant (WWTP) inflow rates, which also receive trade effluent and urban runoff (e.g. see Holt et al., 1998). This means that predicted

concentrations in sewage effluent are likely to be conservative.

We also analysed publicly-available surface water quality monitoring datasets for England and Wales to collate available evidence on the presence of the substances evaluated. These include semi-quantitative screening (GC–MS and LC-MS) data for 13 substances and fully quantitative targeted analysis of three substances (tetrachloroethylene, trichloroethylene and dinoseb). No monitoring data appear to be available for six substances (tricarbonyl(methylcyclopentadienyl) manganese; 2-phosphonobutane-1,2,4-tricarboxylic acid; 4-hydroxy-2,2,6,6- tetramethylpiperidinoxyl; 2-morpholinoethanol, 2,2′-azobis[2-methylbutyronitrile]; APN and chlorendric anhydride). Summary data are presented in the Supplementary Materials (Tables S11 and S12). These data were used to sense-check predicted exposures but a formal validation was not performed.

2.2. Removal of chemicals in WWTPs

Removal in WWTPs (Table 1) was calculated based on physico-chemical properties (collated from public data sources: see Supplementary Materials) using SimpleTreat v4 (Struijs, 2014; Lautz et al., 2017). This is believed to be superior to other models, such as the STP model embedded in EpiWIN (e.g. Clark et al., 1995; Wang et al., 2007) and earlier versions of SimpleTreat (Struijs, 1996) for ionisable substances, because it explicitly describes chemical dissociation and the different behaviours of the dissociated species (e.g. ionised and neutral). Biodegradation was assumed to be zero during treatment, and the calculated removal fractions arise from sorption to sludge and volatilisation. Note that for some chemicals zero removal in WWTPs was predicted. Again, this is likely to be conservative because a small fraction of all chemical substances will be removed in the pore water of wet sewage sludge.

2.3. Removal of chemicals in drinking water treatment plants

Human exposure to PMT chemicals via drinking water was predicted after allowing for removal in different drinking water treatment trains (see also Bevan et al., 2012; Crump et al., 2013) and accounting for

Table 1
Removal rates in WWTPs (from SimpleTreat v4) and in conventional and advanced water treatment for the 22 PMT substances considered along with PM classifications. M classifications were based on K_{OC}, with respect to the thresholds adopted by the European Commission (2020). nM is not mobile (i.e. log(K_{OC}) > 3). Persistence classifications were taken from DWI (2020). pvP is potentially vP. nP is not persistent. Bold text indicates classification by Arp and Hale (2019). *from the ECHA registration dossier. Also shown are the Health-Based Guidance Values (HBGVs).

Compound	Removal Rate in WWTP (%)	Removal Rate Conventional WTW (%)	Removal Rate Advanced WTW (%)	PM class	HBGV (µg kg bw^{-1}d^{-1})
2,4,7,9-tetramethyldec-5-yne-4,7-diol	7.99	94.60	99.76	pvP, M	1.50
1,4-dioxane	0.67	15.39	89.62	vP, vM	16.0
Trichloroethylene	91.96	47.71	94.99	nP, vM	0.14
Tetrachloroethylene	94.13	47.69	97.17	vP, M	4.70
Melamine	0.00	73.72	95.12	vP, vM	200
Dinoseb	52.27	87.56	99.95	vP, M	0.78
Dapsone	0.60	80.83	99.35	pvP, M	4.20
Amantadine	41.03	83.00	96.60	vP, M	1.50
Hexamethyldisiloxane	77.77	99.00	99.98	pvP, nM	270
Tricarbonyl(methylcyclopentadienyl) manganese	0.00	49.33	99.04	vP, nM	1.50
Cyanuric Acid	0.00	40.17	94.13	vP, vM	1500
2-phosphonobutane-1,2,4-tricarboxylic acid	0.28	0.00	49.00	P, vM	1875
Chlorendic anhydride	59.77	77.97	99.16	pvP, vM	0.17
4-hydroxy-2,2,6,6- tetramethylpiperidinoxyl	0.03	0.00	50.00	pvP, vM	200
4-aminophenol	0.41	80.98	99.20	pvP, vM	100
2-morpholinoethanol	0.37	61.94	97.28	pvP, vM	1.50
2,2′-azobis[2-methylbutyronitrile]	1.68	95.08	99.87	vP, M	1.50
1,3-dichlorobenzene	82.12	45.29	97.87	vP, nM	20.0
2,2′-dimethyl-2,2′-azodipropiononitrile (APN)	0.18	71.33	98.21	vP, vM	10.0
N-butylbenzenesulphonamide	2.05	43.78	97.29	vP*, vM	260
1,2-benzisothiazol-3(2H)-one 1,1-dioxide (saccharin)	0.09	99.00	99.83	vP, vM	3800
Triclosan	51.97	48.96	98.99	vP, nM	400

riverine dilution (Tier 4 only). Two treatment scenarios were considered. The first was 'conventional treatment' consisting of coagulation-flocculation, filtration and chlorination. The second was 'advanced treatment' consisting of coagulation-flocculation, filtration, ozonation, activated carbon and chlorination. Advanced treatment is commonly employed at many treatment plants where source waters are impacted by micropollutants (e.g. via wastewater emissions or agricultural runoff). Removal rates for each treatment scenario were estimated from physico-chemical properties. No removal was assumed for coagulation. Removal by filtration with granular activated carbon (GAC) in advanced treatment was estimated from K_{OW} and charge at pH 7 (Drewes, 2007). Removal via chlorination and ozonation was estimated using an application of a 3D-QSPR model (Lei and Snyder, 2007). Details of the approach are given in the Supplementary Materials (S2). The removal rates derived are shown in Table 1.

2.4. Exposure assessment in tiers 1–3

The nature of use and estimates of amounts released in the UK are detailed in the Supplementary Materials (S3). Scenarios for the exposure assessment are summarised in Table 2. For assessment tiers 1–3 no dilution in receiving waters is assumed (i.e. treated WWTP effluent is consumed) which is clearly highly conservative. In most cases, tonnages in Tier 3 were the same as or lower than those employed in Tier 2, except for three substances (chlorendic anhydride, 4-aminophenol and saccharin) for which the refined Tier 3 tonnage was higher than for Tier 2.

The tonnage for melamine was assumed to be the sum of direct emissions of melamine and half of the tonnage estimated for the substance 1,3,5-triazine-2,4,6(1H,3H,5H)-trione, with 1,3,5-triazine-2,4,6-triamine (1:1). This substance is essentially a 50/50 mix of melamine and cyanuric acid. The remaining 50 % of the estimated tonnage of this substance in each assessment was evaluated as cyanuric acid. The substance tricarbonyl(methylcyclopentadienyl) manganese (sometimes referred to as MMT) is assumed to dissociate in water into methyl-cyclopentane and tricarbonyl manganese. The risk assessment was performed using the tonnage for the whole chemical but with toxicity and physico-chemical properties for methylcyclopentane only to predict

Table 2
Summary of assumptions in the first three tiers of the exposure assessment. In all cases, removal in WWTPs was estimated using SimpleTreat 4.0. For details of Tier 3 tonnage estimates see Supplementary Materials (S3). Suffixes A, B and C represent the level of drinking water treatment. Tiers 1–3 reflect increasing refinement of tonnage estimate. For example, Tier 1 A reflects an extreme, worst-case scenario (100 % of EU tonnage used in UK and no drinking water treatment), whilst tier 3C reflects a refined UK tonnage based on best available information and considers advanced drinking water treatment.

Tier	Tonnage	Riverine dilution of treated wastewater	Drinking water treatment
1 A	100 % EU tonnage	No	None
1B	100 % EU tonnage	No	Conventional
1C	100 % EU tonnage	No	Advanced
2 A	9 % EU tonnage	No	None
2B	9 % EU tonnage	No	Conventional
2C	9 % EU tonnage	No	Advanced
3 A	Refined from Tier 2	No	None
3B	Refined from Tier 2	No	Conventional
3C	Refined from Tier 2	No	Advanced
4 A	As for Tier 3	Spatially explicit for Q_{mean} and Q_{95}	Conventional
4B	As for Tier 3	Spatially explicit for Q_{mean} and Q_{95}	Advanced

removal in WWTPs and in different water treatment processes and for the risk assessment.

2.5. Estimation of toxicity

A toxicological evaluation was undertaken to identify the most sensitive point of departure (PoD) for each substance, which was then used to determine a Health-Based Guidance Value (HBGV) expressed as the amount of a chemical that may be ingested regularly (i.e. through food and drinking water) over a lifetime without appreciable risk. HBGVs (expressed as tolerable intake per unit body mass per day: μg kg bw^{-1} d^{-1}) are designed to be protective of health for the entire population, including sensitive sub-populations. Details of the derivation of HBGVs are given in the Supplementary Materials (S5). Briefly, these were based on a review of publicly-available data published by regulatory bodies and supplemented by studies in the peer-reviewed literature. The HBGV used in the risk assessment was selected as the lowest cancer or non-cancer HBGV. Where the non-cancer endpoint was lower than the cancer endpoint, the non-cancer HBGV will in most cases be protective of the cancer effect.

2.6. Calculation of risk characterisation ratios

In all the scenarios described above, no dilution in the receiving water body was assumed. This conservative assumption means that substances can be screened from the risk assessment process if Risk Characterisation Ratios (RCRs) are <1 at this stage. The *RCR* is defined as

$$RCR = \frac{C.I.M^{-1}}{HBGV} \tag{1}$$

where C is the concentration of chemical consumed in drinking water (μg L^{-1}), I is the daily drinking water intake for the individual being assessed (L cap^{-1} d^{-1}), M is the body mass of the individual being assessed (kg cap^{-1}) and *HBGV* is the health based guidance value for daily intake (μg kg^{-1} d^{-1}) which is derived from the toxicology assessments. Values of I for infants, children and adults were assumed to be 0.75, 1 and 2 L cap^{-1} d^{-1}, respectively, and values of M for infants, children and adults were assumed to be 5, 10 and 60 kg, respectively (e. g. after WHO, 2011). Note that the body mass assumptions for adults and children used here are lower than those used elsewhere (e.g. by EFSA, 2012) and are, thus, relatively conservative. An infant with a body mass of 5 kg corresponds to an age of approximately eight weeks and a child with a body mass of 10 kg corresponds to an age of approximately two years, assuming median growth curves for girls in the UK (RCPCH, 2012). The concentration of chemical, C, is derived from

$$C = \frac{T.\delta}{P_{UK}.w}.(1 - r_{WWTP}).(1 - r_{WTW}) \tag{2}$$

where T is the annual tonnage assumed for the UK, P_{UK} is the population of the UK, w is the water use *per capita* (150 L d^{-1}), r_{WWTP} is the removal rate in secondary sewage treatment, r_{WTW} is the removal rate in drinking water treatment and δ is a factor to convert tonnes a^{-1} to μg d^{-1}.

Two sets of *RCR* were generated for each chemical in each scenario: (i) *RCR* assuming 100 % of exposure to the chemical arises from the consumption of drinking water (Eq. 1) and (ii) RCR_T assuming only 20 % of exposure to the chemical arises from the consumption of drinking water (i.e. the Relative Source Contribution, RSC = 0.2: e.g. Brown and Foos, 2009). Note that the latter assumption is somewhat arbitrary and is intended to add an additional factor 5 to the conservatism of the risk assessment to account for unknown additional exposures (e.g. via food or the environment) rather than being based on a specific assessment of exposure via multiple sources. The total exposure including additional exposure from unknown sources can be calculated as

$$RCR_T = \frac{C.I.M^{-1}}{0.2*HBGV} \qquad (3)$$

2.7. Tier 4: Spatially-referenced modelling of riverine concentrations

A spatially-referenced gridded model of riverine exposure was constructed for Great Britain. The model operates on a grid for the whole of Great Britain, at a resolution of 1 km, similar to that described by Keller et al. (2007) and Whelan et al. (2012) but with updated data on river discharge and population density.

For all PMT chemicals, surface water exposure was assumed to occur via the "down the drain" pathway (from both industrial and domestic sources). Concentrations were predicted from population density, based on a *per capita* use rate and removal in WWTPs, as above, but combined with dilution based on gridded water balance model predictions. It should be noted that although exposures are predicted for all 1 km grid cells in Great Britain, it is assumed that abstraction takes place only in cells containing a surface water abstraction point.

For surface water exposure, we employed algorithms described by Whelan et al. (2012). Chemical load is routed through a flow direction network (derived from a digital elevation model: DEM), discounting for instream degradation (biodegradation, photodegradation or hydrolysis) if applicable. This is assumed to take place according to first order kinetics. For the PMT chemicals considered here, very high persistence was assumed (i.e. a DT_{50} of 10,000 h) which results in negligible in-

stream losses for the velocities and travel distances typically encountered in British rivers. Steady state chemical emissions in each cell were calculated from population density and an assumed *per capita* chemical consumption (U, g cap^{-1} day^{-1}), discounted by the fraction of chemical removed during sewage treatment and adjusted by the fraction of the population served by secondary sewage treatment. In the case of Great Britain, we assumed that 100 % of the population is served by at least secondary treatment. This is based on an estimate of 96 % of the UK population connected to WWTPs in 2002, with most of the remainder served by small private treatment works, cesspits or septic tanks (Defra, 2002). More recent estimates (e.g. GWI, 2018) suggest that the figure is close to 100 %. This will be the case in general, but this assumption does not account for the (intermittent) emission of untreated wastewater (e.g. in Combined Sewer Overflows: CSOs). Although CSOs are usually designed to occur only under wet-weather conditions (which provide an associated additional dilution), this may not always be the case (Whelan et al., 2022).

Concentrations in each 1 km grid cell were calculated from cumulative chemical loads using discharge estimates derived by a cumulative routing of runoff (discharge per unit area of contributing catchment) through the grid using flow direction vectors derived from a digital elevation model (DEM: CEH, 2022). We used model estimates of monthly discharge between 1960 and 2015 for a 1 km grid of Great Britain to derive a mean discharge (Q_{mean}) and a 95 % exceedance flow (Q_{95}) for each grid cell (see Fig. 1). The original monthly model
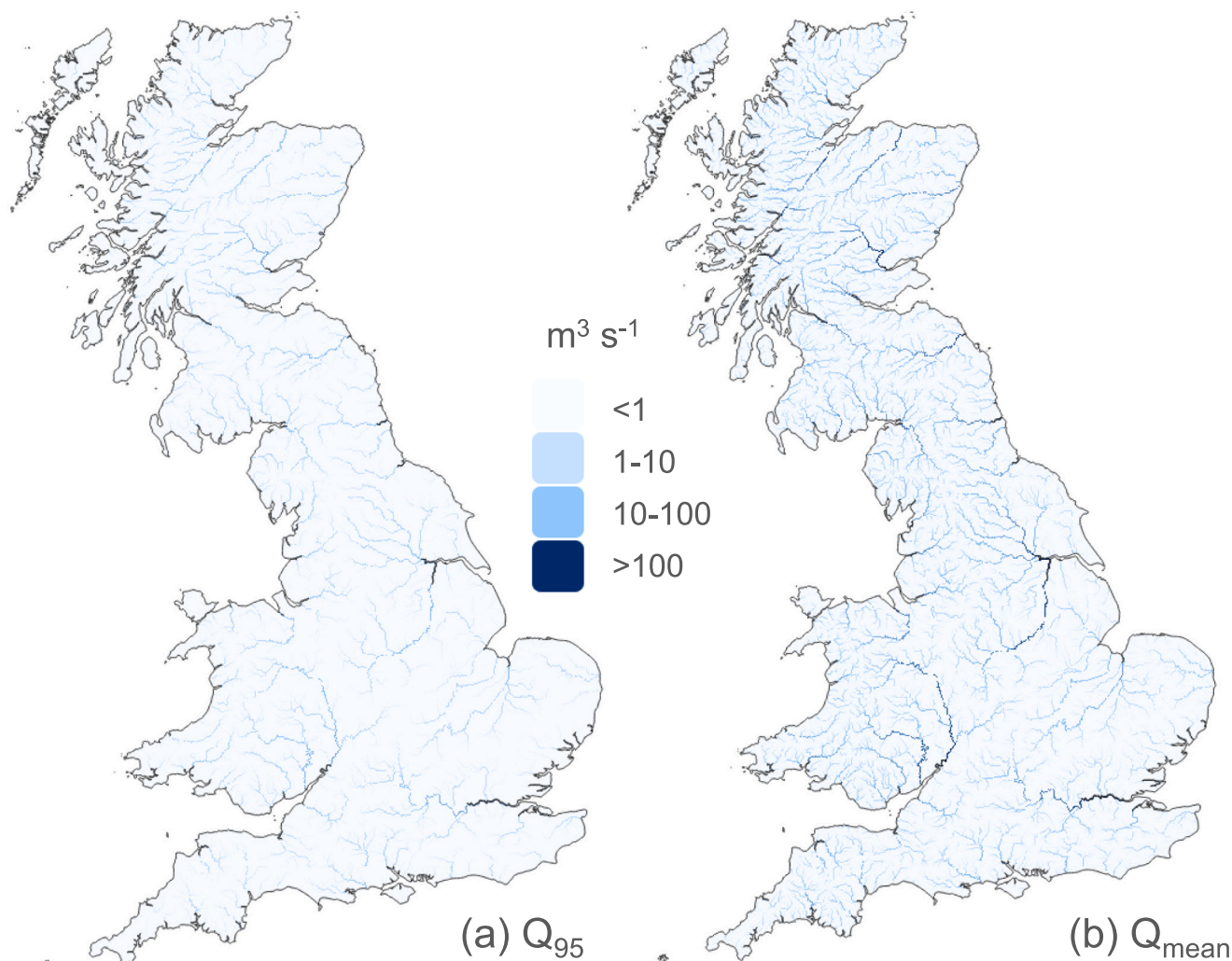

Fig. 1. (a) Q_{95} and (b) Q_{mean} for Great Britain in a 1 km grid estimated by the G2G model using gridded rainfall data and MORECS evapotranspiration estimates (Bell et al., 2018).

estimates were produced using the CEH Grid-to-Grid (G2G) model for the MaRIUS (Managing the Risks, Impacts and Uncertainties of drought and water Scarcity) project using gridded rainfall data (CEH- Gridded Estimates of Areal Rainfall) and MORECS (Met Office Rainfall and Evaporation Calculation System) evapotranspiration estimates (Bell et al., 2018 but see also Bell et al., 2009). These data were processed using library routines in the NetCDF4 Python package to derive grids for Q_{mean} and Q_{95}. Flow directions were derived using the method of Paz et al. (2006) as applied by Davies and Bell (2009) from the hydrologically corrected 50 m integrated-hydrological-digital-terrain-model (IHDTM: Morris and Flavin, 1990, 1994). Flow directions use the D8 method (Jenson and Domingue, 1988), which has eight drainage directions. Population density estimates (Fig. S3) were derived from the UK Gridded Population 2011, based on the 2011 Census and Land Cover Map 2015 (Reis et al., 2017). These data were trimmed to a consistent grid (Great Britain - not including Northern Ireland) and imported into the chemical routing model with a consistent spatial reference system.

For non-persistent chemicals, loads can be discounted for degradation, assuming first order kinetics with travel time, calculated as the quotient of the stream path length and a nominal water velocity, allowing for stream channel sinuosity:

$$L_x = L_0 . e^{-k.\tau} \tag{4}$$

where L_x is he chemical load at a particular point in the river, L_0 is the load at a distance x (m) upstream along the flow path, k is the first-order degradation rate constant for the chemical of interest and τ is the travel time (h) calculated as:

$$\tau = \frac{x}{v} . c \tag{5}$$

where x is the distance along the channel (m), c is a factor to convert from seconds to hours (2.78×10^{-4}) and v is water velocity (m s^{-1}). For cell to cell transfer in the horizontal and vertical directions, x was calculated as

$$x = z.s \tag{6}$$

and for diagonal transfers, x was calculated from Pythagoras theorem as

$$x = \sqrt{2.z^2} . s \tag{7}$$

where z is the cell width (1000 m) and s is river sinuosity, defined as the ratio of river length to down-valley length (Leopold and Wolman, 1957). This varies substantially depending on geomorphological setting with reports ranging from ca 1 (nearly straight) to >3 (Stolum, 1996; Stolum, 1998) with averages for natural channels of about 2 (e.g. Frascati and Lanzoni, 2009). Here, we assumed that all rivers have a sinuosity of 3 to account for the fact that valley lengths are also longer than the straight-line lengths between grid cells used for flow routing in the model.

Water velocity was calculated from discharge using the empirical equation derived by Round et al. (1998):

$$v = 10^a . Q_{mean}{}^b . \left(\frac{Q}{Q_{mean}} \right)^c \tag{8}$$

where Q is the river discharge (m^3 s^{-1}), Q_{mean} is the mean river discharge for the reach in question (m^3 s^{-1}) and a, b and c are empirical parameters reported to be -0.583, 0.283 and 0.495, respectively.

2.8. Concentration estimates at drinking water abstraction points

The locations for Water Treatment Works (WTW) in England and Wales were supplied by the DWI. These were assumed to be representative of drinking water abstraction points although, in practice, many WTWs treat water from multiple (and often) distal sources. After the removal of groundwater abstraction points, duplicate locations and duplicates within grid cells, there were 296 cells containing either a surface water or mixed water abstraction point. Note that the abstraction point in the Scilly Isles was also removed from this list. The locations of these abstraction points are shown in Fig. S2.

Predicted concentration in raw (untreated) drinking water was assumed to be the concentration predicted for the 1×1 km grid square within which the abstraction point was located. These concentrations were then reduced in accordance with the estimated removal rates in different drinking water treatment trains (conventional or advanced, as described above) to generate a distribution of concentrations in treated drinking water, and associated RCRs, under Q_{mean} and Q_{95} conditions.

3. Results and discussion

3.1. Tiers 1–3

Predicted drinking water concentrations and RCRs for each chemical in each emission scenario considered in Tier 3 are shown in Table 3. Equivalent data for Tiers 1 and 2 can be found in the Supplementary Materials (Tables S5 and S6). In all cases, the results for the scenario representing an infant ($M = 5$ kg; $I = 0.75$ L d^{-1}) are presented, as this is the most conservative for risk assessment. In all cases, removal in WWTPs was considered but no dilution in the receiving water body was assumed. In these scenarios, drinking water was assumed to be the only intake route for the chemical. Chemical removal in drinking water treatment was assumed to be zero for Scenario A, the rate predicted for conventional treatment was used for Scenario B and the rate predicted for advanced treatment was used for Scenario C.

For Tier 1 (100 % of EU tonnage assumed to be used in the UK), most chemicals have RCRs >1 for scenarios 1 A (no treatment) and 1B (conventional treatment) but only seven chemicals have an RCR > 1 for Scenario 1C (advanced treatment). This suggests that, for most chemicals, advanced treatment would be sufficient to reduce human health risks to acceptable levels (RCR < 1) even given the most extremely conservative assumptions in exposure estimation. When further refinement of tonnage was employed (Tiers 2 and 3), the number of substances with an RCR exceeding unity decreased. When assuming the most realistic estimates of emission (Tier 3) and conventional drinking water treatment provision (Scenario 3B), no substances were predicted to have an RCR > 1. Some margin of safety (RCR < 0.1) was predicted when advanced water treatment trains were assumed (Scenario 3C). This suggests that an acceptable level of risk for drinking water consumption can be achieved for these substances, provided that conventional treatment trains are in place. Even in the absence of any water treatment, potential risks (RCR > 1) were only identified for tetrachloroethylene and chlorendic anhydride (1,4,5,6,7,7-hexachloro-8,9,10-trinorborn-5-ene-2,3-dicarboxylic anhydride).

When the more conservative assumption that only 20 % of chemical intake arises from the consumption of drinking water (again, assuming no riverine dilution), RCRs increase (see RCR_T values in Table 3), with data for Tiers 1 and 2 presented in the Supplementary Materials (Tables S7 and S8). Even in this case, just three chemicals have $RCR_T > 1$ for conventional water treatment (Tier 3B), although all chemicals have $RCR_T < 1$ for the advanced treatment scenario. The three chemicals of potential concern under conventional treatment were included in Tier 4. These were 1,4-dioxane; tetrachloroethylene and chlorendic anhydride. All other chemicals (including dapsone, melamine and APN, which had $RCR_T > 1$ for the highly conservative Tier 3 A) were not considered further.

The highest RCRs were generated for tetrachloroethylene, which was selected to illustrate the Tier 4 spatial assessment. These RCR values were driven by a combination of tetrachloroethylene's toxicity (HBGV: 4.7 µg kg bw^{-1} d^{-1}) and moderate tonnage (2500 t a^{-1}). Targeted and quantitative monitoring data (based on between 2099 and 26,680 samples from the year 2000 onwards) for surface waters in England, collected by the Environment Agency, are summarised for tetrachloroethylene, trichloroethylene and dinoseb in the Supplementary Materials

Table 3
Predicted drinking water concentrations (C) in µg L^{-1}, *RCR* and *RCR$_T$* values for each chemical in Tier 3. Note *RCRs* are shown here for infants. Values for children and adults are lower. Cells coloured orange with bold text have *RCR* or *RCR$_T$* > 1. Cells coloured green have *RCR* or *RCR$_T$* < 1. RCRs calculated assuming exposure only via drinking water. *RCR$_T$* calculated assuming exposure via drinking water represents 20 % of total exposure. Concentrations shown as 0.0 indicate values <0.01 µg L^{-1} *RCR* or *RCR$_T$* values shown as 0.00 indicate values <0.001.

Compound	No DW Treatment Tier 3A			Conventional Tier 3B			Advanced Tier 3C		
	C	RCR	RCR$_T$	C	RCR	RCR$_T$	C	RCR	RCR$_T$
2,4,7,9-tetramethyldec-5-yne-4,7-diol	0.6	0.06	0.29	0.0	0.00	0.02	0.0	0.00	0.00
1,4-dioxane	61.5	0.58	**2.88**	52.1	0.49	**2.44**	1.6	0.08	0.30
Trichloroethylene	0.0	0.00	0.00	0.0	0.00	0.00	0.0	0.00	0.00
Tetrachloroethylene	39.9	**1.27**	**6.36**	20.9	0.67	**3.33**	5.7	0.55	0.18
Melamine	484.2	0.36	**1.82**	127.3	0.10	0.48	50.6	0.00	0.09
Dinoseb	0.4	0.07	0.34	0.0	0.01	0.04	0.0	0.00	0.00
Dapsone	25.4	0.91	**4.53**	4.9	0.17	0.87	0.2	0.00	0.03
Amantadine	1.6	0.16	0.80	0.3	0.03	0.14	0.0	0.00	0.03
Hexamethyldisiloxane	54.4	0.03	0.15	0.5	0.00	0.00	0.0	0.00	0.00
Tricarbonyl(methylcyclopentadienyl) manganese	0.0	0.00	0.00	0.0	0.00	0.00	0.0	0.00	0.00
Cyanuric Acid	81.5	0.01	0.04	48.8	0.00	0.02	0.1	0.00	0.00
2-phosphonobutane-1,2,4-tricarboxylic acid	2438.6	0.20	0.98	2438.6	0.20	0.98	27.0	0.00	0.50
1,4,5,6,7,7-hexachloro-8,9,10- trinorborn-5-ene-2,3-dicarboxylic anhydride	1.4	**1.23**	**6.14**	0.3	0.27	**1.35**	0.0	0.00	0.05
4-hydroxy-2,2,6,6- tetramethylpiperidinoxyl	244.5	0.18	0.92	244.5	0.18	0.92	2.5	0.00	0.46
4-aminophenol	8.4	0.01	0.06	1.6	0.00	0.01	0.0	0.00	0.00
2-morpholinoethanol	0.5	0.05	0.24	0.2	0.02	0.09	0.0	0.00	0.01
2,2'-azobis[2-methylbutyronitrile]	1.6	0.16	0.80	0.1	0.01	0.04	0.0	0.00	0.00
1,3-dichlorobenzene	0.1	0.00	0.00	0.0	0.00	0.00	0.0	0.00	0.00
2,2'-dimethyl-2,2'-azodipropiononitrile (APN)	20.1	0.30	**1.51**	5.8	0.09	0.43	0.1	0.00	0.03
N-butylbenzenesulphonamide	124.3	0.07	0.36	69.9	0.04	0.20	0.1	0.00	0.01
1,2-benzisothiazol-3(2H)-one 1,1-dioxide (saccharin)	223.2	0.01	0.04	2.2	0.00	0.00	0.0	0.00	0.00
Triclosan	0.0	0.00	0.00	0.0	0.00	0.00	0.0	0.00	0.00

(Table S12). The frequencies of detection for these substances (> limit of detection, LOD) were 18 %, 15 % and 16 %, respectively. For tetrachloroethylene, the maximum measured concentration was >7000 µg L^{-1}, although the 98th percentile concentration was just 1.38 µg L^{-1}. This is an order of magnitude lower than the concentration predicted in treated wastewater effluent for Tier 3 tonnage (40 µg L^{-1}). However, in most situations, wastewater effluent will be diluted by river water. If we assume a dilution factor of 10 (as recommended by TGD, 2003), predicted river water concentrations for tetrachloroethylene are within a factor 3 of the 98th percentile measured concentration, suggesting that the model represents a reasonable worst-case exposure for this substance. For dinoseb, the maximum measured concentration was 70 µg L^{-1} and the 98th percentile concentration was 0.59 µg L^{-1}(0.35 µg L^{-1}). However, the predicted values are still fairly reasonable if we assume a dilution factor of 10. This would place the predicted exposure between the 50th and 98th percentile measured concentrations. For trichloroethylene, modelled Tier 3 exposures (<0.01 µg L^{-1}) are substantially lower than the range of concentrations reported above the limit of detection (0.01–76 µg L^{-1}), suggesting that the Tier 3 tonnage for this chemical significantly under-estimates emissions. However, if we assume Tier 1 or Tier 2 tonnages, predicted concentrations in treated wastewater effluent are much higher (2340 and 197 µg L^{-1}, respectively) and, after dilution, the Tier 2 exposures would be within the 98th percentile and the maximum measured concentrations. This suggests that higher tier exposure assessments may not be conservative for all compounds and highlights the need for better emissions estimates. Data on frequencies of detection for semi-quantitative non-targeted screening analyses of English surface water samples reported by the Environment Agency between 2017 and 2021 are also presented in the Supplementary Materials (Table S11). High detection frequencies (>30 %) were observed for 2,4,7,9-tetramethyldec-5-yne-4,7-diol, melamine, amantadine, saccharin and triclosan. Low detection frequencies (<5 %) were reported for tetrachloroethylene, dinoseb, hexamethyldisiloxane, 1,3-dichlorobenzene and N-butylbenzenesulphonamide. For the three compounds which are regularly monitored using quantitative targeted analyses, these non-target detection frequencies are lower (especially in the case of dinoseb) which highlights that these screening data should be

used with caution. In any case, making comparisons between frequency data and model predictions of concentrations in wastewater is difficult and there was no apparent correlation in rank order. It should be noted that that for triclosan, the Tier 3 tonnage was assumed to be zero due to a recent removal of this substance from the UK market. However, monitoring data go back to 2017 when triclosan emissions may have been higher than they are today.

3.2. Tier 4

The predicted spatial pattern of surface water exposure for all the substances investigated are very similar because they are calculated using the same exposure assumption and data sets (population density, topographically-derived flow routing and river discharge). Only one example for tetrachloroethylene is shown here, for illustrative purposes. This compound had the highest *RCR$_T$* value at Tier 3B (despite having a reasonably high predicted removal rate in both WWTPs and WTWs and not having the highest predicted exposure in river water) due to a combination of moderate tonnage (2500 t a^{-1} at Tier 3) and a fairly low HBGV (4.7 µg kg^{-1} d^{-1}). It has several different dispersive uses and has a relatively large body of environmental occurrence data in English rivers. Predicted Tier 4 riverine concentrations of tetrachloroethylene are shown in Fig. 2 and predicted patterns of *RCR$_T$* values are shown in Fig. 3 for both Q_{mean} and Q_{95} conditions. Predictions of spatial patterns of riverine concentrations for the other two chemicals with *RCR$_T$* values greater than one in tier 3B (20 % of total exposure from drinking water) are shown in the Supplementary Materials (S6).

Predicted riverine concentrations of tetrachloroethylene were as high as 39 µg L^{-1} under Q_{95} conditions. This is close to the concentration calculated for WWTP effluent – suggesting little dilution at low flow. Highest concentrations are, principally, predicted in densely populated areas including Greater London, the West Midlands, the Mersey basin (Manchester and Liverpool), West Yorkshire, the Northeast of England and the Central belt of Scotland.

Values of *RCR$_T$* for tetrachloroethylene extracted from cells containing WTWs with surface water abstraction points for drinking water supply are displayed as cumulative probability distributions in Fig. 4 for
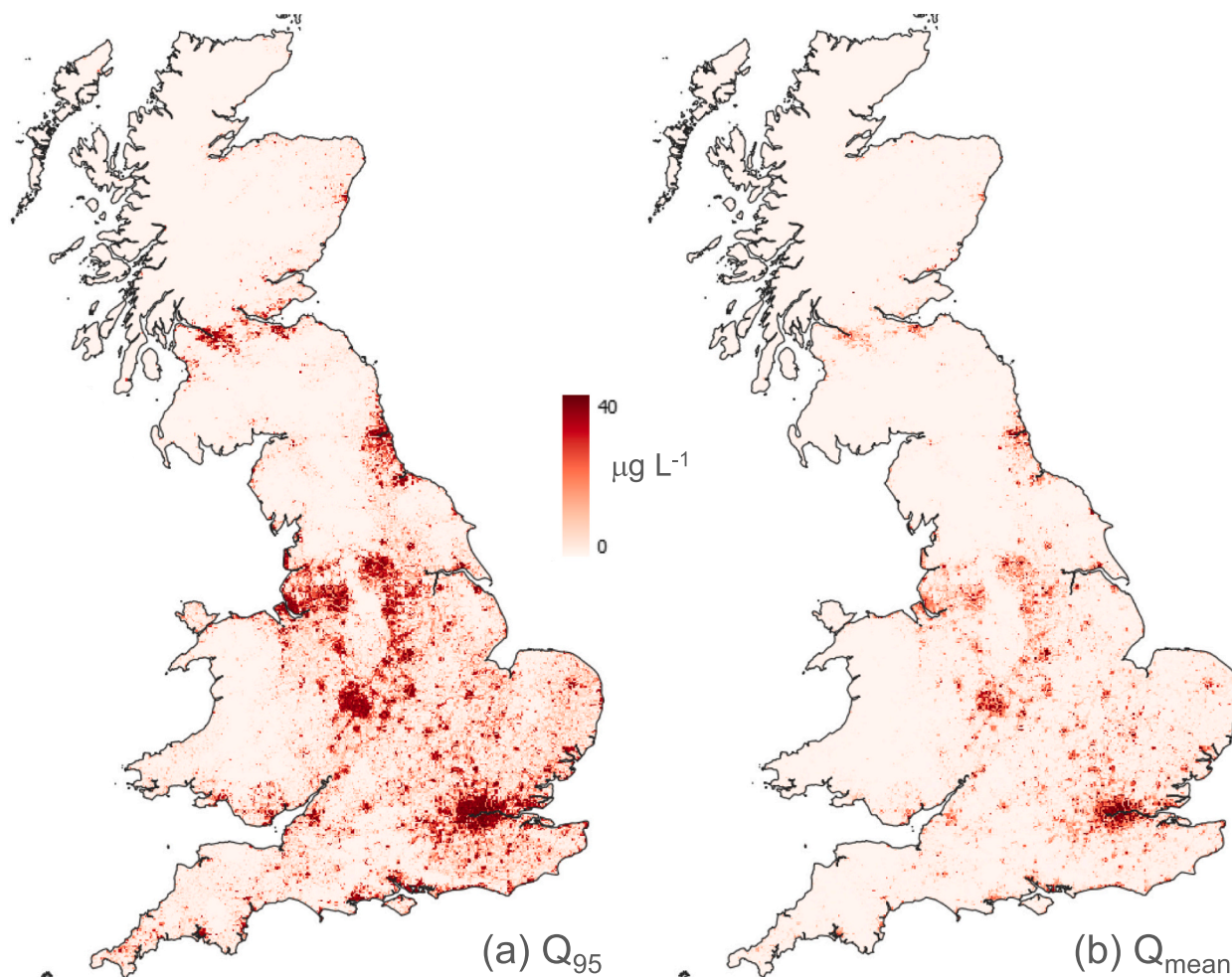
Fig. 2. Predicted spatial patterns of the concentration of tetrachloroethylene in river water for Great Britain under (a) Q_{95} and (b) Q_{mean} conditions.

conventional water treatment trains at Q_{mean} and Q_{95}. The percentage of abstraction points predicted to have RCR_T values >1 for all three chemicals considered in Tier 4 are shown in Table 4.

For conventional treatment, 15 (5.07 %) and 54 (18.24 %) treatment works (out of 296) were predicted to have RCR_T greater than unity for tetrachloroethylene under Q_{mean} and Q_{95} conditions, respectively.

Model predictions for the other two chemicals with top three RCR values are shown in the Supplementary Materials. In summary, for 1,4-dioxane 2.03 % and 12.5 % of WTWs were predicted to have $RCR > 1$ under Q_{mean} and Q_{95} conditions, respectively. For chlorendic anhydride, 0 % and 4.39 % of treatment works were predicted to have RCR_T greater than one under Q_{mean} and Q_{95} conditions, respectively. Those WTWs identified as presenting higher potential risks to human health could be targeted for more detailed monitoring to confirm or dismiss the risks calculated by the model.

Note that predicted RCR values (exposure via drinking water only) were less than unity for all abstraction points under both flow conditions for conventional treatment and RCR_T values (i.e. with an additional hypothetical five-fold exposure to these compounds from other sources) were less than unity for all abstraction points under both flow conditions when advanced treatment was assumed. Our modelled data suggest that conventional drinking water treatment trains are sufficient to manage risks to human health from the PMT substances studied. However, it is important to acknowledge the high uncertainty associated with our exposure estimates – principally because of the lack of accurate data on amounts of these substances used in the UK. In any case, if advanced treatment is in place, acceptable margins of safety are likely be achieved for total exposure.

Proponents of chemical screening based solely on hazard criteria, such as PMT and vPvM, maintain that risk assessment is not possible if there is high uncertainty in emission rates. We argue here that a tiered exposure assessment can make a useful contribution to the screening process – allowing many chemicals which are unlikely to pose harm to humans (e.g. via drinking water and, by extension, harm to wildlife) to be eliminated from additional evaluation and allowing resources to be directed towards better understanding (and, where appropriate, managing) chemicals of higher concern. The inclusion of an assessment of chemical removal in different drinking water treatment trains is a particularly useful addition to the screening process because protection of drinking water is a primary driver for screening chemicals via P and M (e.g. Arp and Hale, 2019).

The results of the screening risk assessment for a set of example PMT candidates at lower tiers suggest that most of the substances considered are likely to pose negligible risk to humans via drinking water consumption. RCRs were generally <1, even with highly conservative assumptions regarding use and release and assuming no dilution in receiving waters and no advanced drinking water treatment.

RCR_T values from lower tier assessments for three substances: 1,4-dioxane, tetrachloroethylene and chlorendic anhydride, were > 1, indicating a potential risk. This was explored further using a spatially-referenced higher tier assessment employing a gridded exposure model for the whole of Great Britain. This model generated estimates of surface water concentrations based on emissions which were assumed to be proportional to population density and routed through the landscape via a 1 km resolution DEM, under two flow scenarios: Q_{mean} and Q_{95}.

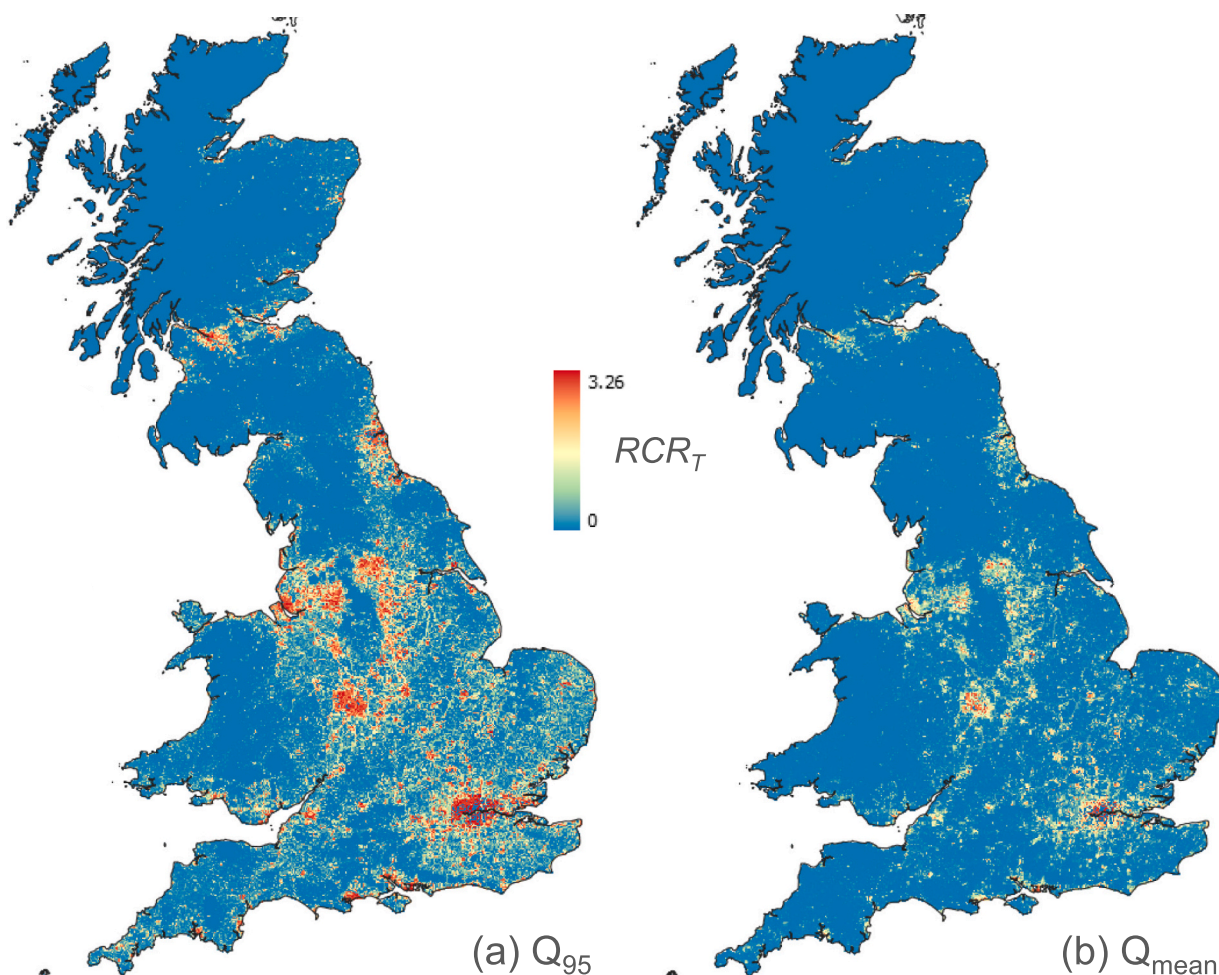In the higher tier assessment, all three substances had $RCR > 1$ at

Fig. 3. RCR_T derived from predicted concentrations of tetrachloroethylene assuming an infant exposure and conventional water treatment, for all cells shown in Fig. 2, under (a) Q_{95} and (b) Q_{mean} conditions.
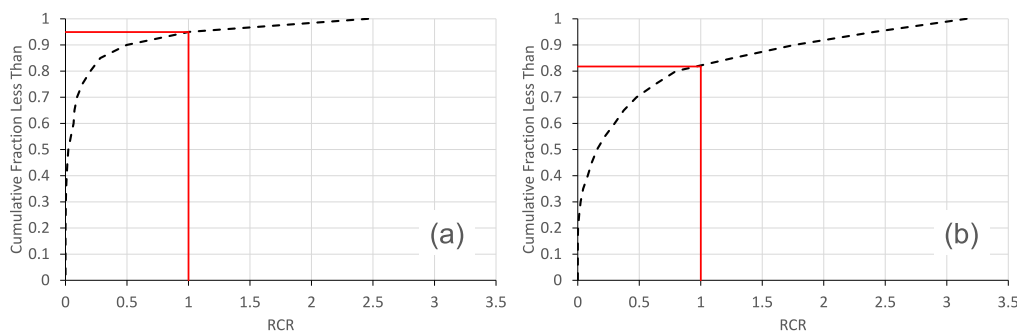

Fig. 4. Cumulative distributions of predicted RCR_T values for tetrachloroethylene in drinking water for cells containing WTWs with surface water abstractions, assuming conventional treatment (dashed lines) under (a) Q_{mean} and (b) Q_{95} flow. The red lines show the cumulative fraction less than $RCR_T = 1$.

Table 4
Predicted percentage of cells containing WTWs with surface water abstraction points which have $RCR_T > 1$ for Q_{mean} and Q_{95} conditions under the assumption of conventional water treatment.

Chemical	Q_{mean} conditions	Q_{95} conditions
1,4-dioxane	2.03	12.50
Tetrachloroethylene	5.07	18.24
Chlorendic anhydride	0	4.39

some abstraction sites when assuming negligible degradation in the stream network and only conventional drinking water treatment technologies. For tetrachloroethylene, predicted river concentrations in cells containing WTWs with surface water abstraction points generated RCR_T values >1 for approximately 5 % and 18 % of sites for Q_{mean} and Q_{95} scenarios, respectively, under conventional drinking water treatment. For 1,4-dioxane the RCR_T values were > 1 for approximately 2 % of abstraction points at Q_{mean} and 12.5 % of points under Q_{95} conditions. For chlorendic anhydride, RCR_T values were < 1 for all abstraction points at Q_{mean} and for 4.4 % of points at Q_{95}. However, all RCR_T values were < 1 if advanced treatment was assumed.

If drinking water is assumed to represent the only exposure route, *RCR* values were < 1 for all substances at all abstraction points under both conventional and advanced treatment. Overall, these results suggest that the consumption of all the compounds considered via drinking water is unlikely to pose a health risk. However, drinking water may contribute to wider health risks for a small number of substances if substantial additional exposure routes exist. Further consideration of this aspect is beyond the scope of this paper.

It should be noted that the gridded exposure assessments described in this report are extremely crude. Water and chemicals are routed through the landscape at a 1 km resolution, which is too coarse to allow a detailed representation of channel networks and to describe the reality of abstraction point locations with respect to wastewater emissions. In any 1 km cell, there will be several river and stream channels, some of which may receive wastewater and others which will not. In addition, the model assumes that chemicals are emitted to every cell in proportion to the population density. However, in reality, wastewater is collected in sewer networks, with "sewersheds" extending over several km^2, which are routed to WWTPs. This means that most 1 km cells will not receive emissions. This will result in some discrepancies between predicted and actual surface water exposure, although downstream loads should eventually be consistent for persistent chemicals. Similarly, cells were identified containing WTWs which include surface water abstractions. Whilst some of these abstractions will be from rivers receiving wastewater emissions, some will be from reservoirs draining catchments with no wastewater influences, which might be remote from the WTW locations themselves. For example, a WTW situated in an urban area may have a high predicted chemical concentration in raw water, because it has a reasonably high upstream population, but actually draws water principally from uncontaminated distal sources. More refined local assessments would be required to evaluate actual risks at individual WTWs, ideally in collaboration with the WTW operators which can supply detailed information about the nature and ratios of specific sources and, ideally, provide measured data on concentrations in raw and treated water.

There is a high degree of uncertainty associated with the release estimates for all the substances assessed. This is because there is a lack of data available on amounts of each substance supplied or used, either as total tonnage for the UK and/or split by nature of use. Most of the substances we considered have a variety of uses, but there are rarely good data available to indicate how total tonnage is allocated between these applications. Consequently, taking a precautionary approach, we applied worst-case assumptions to estimate emissions. For some substances, specific manufacturing site locations were identified. However, environmental releases of the studied chemicals from these sites are uncertain and they all appear to be downstream of known WTWs with surface water abstractions.

A conservative approach was taken with the selection of the HBGVs associated with 2,4,7,9-tetramethyldec-5-yne-4,7-diol (TMDD), dinoseb and tetrachloroethene. The HBGV for TMDD was derived using the Threshold of Toxicological Concern (TTC) approach. Although a Derived No Effect Level (DNEL: oral, general public) of 250 μg kg bw^{-1} day^{-1} is available for TMDD, this was based on a read-across approach from another substance and is higher than the HBGV selected here for the risk assessment (1.5 μg kg bw^{-1} day^{-1}) so was not used. This is consistent with EFSA guidance on the use of the TTC approach in food safety assessment, which is applicable to drinking water (EFSA, 2019).

A published HBGV was not available for dinoseb. Hence, an HBGV was derived based on the most sensitive PoD from all the available studies, and applying appropriate uncertainty factors. The PoD selected was a comparatively mild effect (an increase in haematocrit in males and decreased extramedullary haematopoiesis of the spleen) when compared with other studies with higher PoDs – making the HBGV for dinoseb relatively conservative.

The HBGV for tetrachloroethene (non-cancer) is also relatively conservative, as it is based on a tolerable daily intake (TDI) derived for inhalation exposure which was converted to an oral exposure value via route-to-route extrapolation. Tetrachloroethene also has a HBGV based on carcinogenicity which is higher than the non-cancer HBGV. The non-cancer HBGV is, therefore, protective of carcinogenicity.

The HBGVs for melamine and 1,4-dioxane (non-cancer) are based on robust TDI's set by EFSA and the WHO, based on well documented studies. They are, therefore, considered to be the most appropriate HBGVs for these substances and the TDI used in the risk assessment is considered to be protective of human health for all individuals. As with tetrachloroethene, 1,4-dioxane has a cancer HBGV which is higher than the non-cancer HBGV meaning that the non-cancer HBGV is protective of the cancer effect.

4. Conclusions

Evaluating the potential health risks to humans from PMT/vPvM substances via drinking water is important. The approach described here could be used to support the development of guidance for water companies on risk assessment and to help design targeted monitoring of these and other substances. It will also be of wider value to inform development of future national and international policies on PMT substances. A relatively small subset of substances that are likely to meet the proposed criteria for PMT/vPvM were examined here. This list was based on their relevance to England and Wales, in order to illustrate the approach and demonstrate the value of incorporating exposure assessment as part of a chemical screening process, even if uncertainties in emissions and chemical properties are high. In Europe, the envelope of environmental emissions estimates can be informed by REACH tonnage bands. These emissions can be subsequently refined using improved information about chemical uses, and confirmed via monitoring of wastewater, river water and drinking water. Even when the most conservative assumptions are made about tonnage (e.g. all European tonnage is used in the UK: Tier 1), some substances can be eliminated from further assessment if consideration is taken about removal during the water treatment train. For Tier 2 tonnage, only 5 out of 22 substances had RCR_T values >1, if advanced treatment was assumed. There are likely to be many chemicals which meet the PMT criteria but which can be screened out because they have acceptable risks to human health via drinking water exposure. Many chemicals known to have PMT properties which are already under regulatory scrutiny were deliberately excluded from this assessment. In particular, many PFAS are known to meet the PMT/vPvM criteria and PFAS are receiving considerable attention in the scientific community, amongst regulators and in the media because of their extreme persistence, high environmental mobility and evidence of harmful effects on health for some substances. As the list of PMT substances of concern for drinking water supply is reviewed and updated, additional substances can be evaluated following the methodology outlined here.

Spatially-referenced risk assessment for 1,4-dioxane, tetrachloroethylene and chlorendic anhydride suggested that these substances could make a potential contribution to human health risk at some locations (especially under low flow conditions) but only when assuming conventional treatment trains are in place and assuming that drinking water (treated by conventional treatment trains) makes a 20 % contribution to total exposure. When advanced treatment processes were assumed, none of the 22 substances had RCR_T values >1. This suggests that high margins of safety are likely to be achieved for human health if advanced treatment is in place. That said, additional work is needed to evaluate model predictions. Ideally, predictions of both raw and treated water concentrations should be compared with measured concentration data from a range of drinking water abstraction points. However, these data are currently not in the public domain and these comparisons have not yet been performed.

CRediT authorship contribution statement

M.J. Whelan: Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **E. Pemberton:** Writing – review & editing, Writing – original draft, Project administration, Methodology, Funding acquisition, Conceptualization. **C.B. Hughes:** Writing – original draft, Project administration, Methodology, Funding acquisition, Conceptualization. **C. Swansborough:** Writing – review & editing, Writing – original draft, Investigation, Formal analysis, Data curation. **E.H. Goslan:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Conceptualization. **T. Gouin:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Conceptualization. **V.A. Bell:** Writing – original draft, Methodology, Formal analysis. **E. Bird:** Writing – original draft, Methodology, Formal analysis, Data curation. **S. Bull:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis. **L. Segal:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Data curation. **S.H. Cook:** Writing – original draft, Methodology, Formal analysis, Data curation. **C. Jephcote:** Writing – original draft, Methodology, Formal analysis. **S. Fane:** Writing – original draft, Project administration, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Michael Whelan reports that financial support was provided to all authors (EXCEPT Vicky Bell and Sarah Fane) by The Drinking Water Inspectorate. All authors declare no known competing or conflicting financial interests or personal relationships in the subject matter contained in the paper, that could have appeared to influence the work reported.

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Appendix A. Supplementary data

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Data availability

Data will be made available on request.

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