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Research article

Towards a systematic method for assessing the impact of chemical pollution on ecosystem services of water systems

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ABSTRACT

Chemical pollution impinges on the quality of water systems and the ecosystem services (ESs) they provide. Expression of ESs in monetary units has become an essential tool for sustainable ecosystem management. However, the impact of chemical pollution on ESs is rarely quantified, and ES valuation often focuses on individual services without considering the total services provided by the ecosystem. The purpose of the study was to develop a stepwise approach to quantify the impact of sediment pollution on the total ES value provided by water systems. Thereby, we calculated the total ES value loss as a function of the multi-substance potentially affected fraction of species at the HC50 level (msPAF(HC50)). The function is a combination of relationships between, subsequently: the msPAF(HC50), diversity, productivity and total ES value. Regardless of the inherent differences between terrestrial and aquatic ecosystems, an increase of diversity generally corresponded to an increase in productivity with curvilinear or linear effects. A positive correlation between productivity and total values of ESs of biomes was observed. The combined relationships showed that 1% msPAF(HC50) corresponded to on average 0.5% (0.05–1.40%) of total ES value loss. The ES loss due to polluted sediments in the Waal-Meuse river estuary (the Netherlands) and Flemish waterways (Belgium) was estimated to be 0.3–5 and 0.6–10 thousand 2007$/ha/yr, respectively. Our study presents a novel methodology to assess the impact of chemical exposure on diversity, productivity, and total value that ecosystems provide. With sufficient monitoring data, our generic methodology can be applied for any chemical and region of interest and help water managers make informed decisions on cost-effective measures to remedy pollution. Acknowledging that the ES loss estimates as a function of PAF(HC50) are crude, we explicitly discuss the uncertainties in each step for further development and application of the methodology.

1. Introduction

Human activities are considered to be the driving force of chemical pollution (Posthuma et al., 2020). Emissions from anthropogenic activities are directly or indirectly discharged into natural water bodies, leading to contamination of river basins with agricultural, industrial and household chemicals (e.g. heavy metals and organic pollutants) (Goel, 2006). Hazardous substances pose threats to wildlife, ecosystem stability and function as well as human health (Gerbersdorf et al., 2011; Backhaus et al., 2012; Posthuma et al., 2020). Therefore, various monitoring and modelling tools have been developed to assess the pollution of surface water (Hendriks, 1994; Collins and Mcgonigle, 2008; Michaelis, 2012). Prominently, chemical pollutants are ultimately deposited and accumulated in sediments (De Castro-Català et al., 2016). While surface water quality has improved over the years as required by the European Water Framework Directive (WFD) (European Commission, 2000), sullied sediments may become a source of adverse ecological effects on water systems (De Deckere et al., 2011). However, sediments have received less attention due to lower visibility and higher complexity.

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Chemical pollution has been identified as one of the five highest-ranked environmental pressures that negatively affect global biodiversity (Hirsch, 2010). Ecological effects of chemicals are usually evaluated with the potentially affected fraction (PAF) of species, a toxic pressure metric derived from species sensitivity distributions (SSDs) (De Zwart and Posthuma, 2005; Del Signore et al., 2016). The PAF represents the estimated fraction of species affected at a given exposure to chemicals in the environment (Posthuma et al., 2001). As a standard indicator in risk assessment, the ecological relevance of the PAF has been demonstrated by comparisons with community indicators such as species diversity (De Vries et al., 2010), (mean) species abundance (Posthuma and De Zwart, 2012; Hoeks et al., 2020) and ecological status under the WFD (Posthuma et al., 2020). Recently, Posthuma et al. (2019b) have substantially expanded the RIVM e-toxbase, allowing to employ SSDs for 12,836 chemicals, while distinguishing SSDs for protection targets (SSD-NOEC) and biodiversity impact levels (SSD-HC50), respectively. Consequently, the number of substances covered by SSDs has increased, and estimations of the relative and cumulative impacts of present pollutants on population levels have improved.

In addition to standard indicators for water and sediment quality (e.g. PAF), the ecosystem service (ES) concept has gained more ground in research, policy and applications since the release of the Millennium Ecosystem Assessment (MEA; Millennium Ecosystem Assessment (2005)) (Daily et al., 2009; Maes et al., 2016; Pascual et al., 2017). ESs are defined by the MEA as 'the benefits that people can obtain from ecosystems', and they are further classified by CICES (Common International Classification of Ecosystem Services) into three categories: *Provisioning Services*, *Regulating and Maintenance Services*, and *Cultural Services* (Haines-Young and Potschin, 2018). As a connecting medium between ecosystems via the water cycle, sediments provide a variety of valuable ESs within aquatic systems (e.g. habitat provision, nutrient recycling and flood protection) (Apitz, 2012), while contaminated sediments have the potential to pose ecological risks. Therefore, strategies for evaluating chemical risks in waterways should consider effects on ESs regarding the sustainability of ecological, socio-cultural and economic objectives (Backhaus et al., 2012).

Despite debate and controversy on economic valuation methods and accuracy (e.g. Gómez-Baggethun and Ruiz-Pérez (2011); Laurans et al. (2013); Martín-López et al. (2014)), the expression of ESs in financial terms has become an essential tool for a more comprehensive assessment regarding trade-offs between ecological benefits and land use options (De Groot et al., 2012; Costanza et al., 2017). However, most ES valuation applied a book-keeping and pragmatic approach based on applicability to each specific ES and data availability (Brouwer et al., 2013; Bartkowski, 2017), which is unfeasible for countless pressures and ecosystems. Consequently, quantification is restricted to a small number of ESs, calculated for a few regions only. Besides, ES assessments for water systems focus on a limited number of pressures (e.g. eutrophication, embankments, shipping, climate change and flood defence measures) (Brouwer et al., 2007, 2008; Gilvear et al., 2013; Grizzetti et al., 2016; Koopman et al., 2018). In the Netherlands, assessments of chemical emission reduction only considered increasing economic costs in specific sectors (e.g. agriculture, commercial shipping), with ES-related cost savings focusing solely on wastewater treatment (Brouwer et al., 2008).

While PAF is a central endpoint in chemical risk assessment, its relationship with total ES value has not yet been explored. Instead of pragmatically achieving reasonable estimates of specific ESs, we here explore opportunities for an overarching approach. We hypothesise that the PAF-ES methodological framework would aid valuing the impacts of chemical contamination on ecological benefits. The outcomes of the PAF-ES methodology could ultimately allow water managers to make informed decisions on handling water and sediment pollution by comparing project alternatives (e.g. ecological benefits against remediation costs of contaminated waterways).

Hence, our study aimed to develop and apply a systematic outline to quantify the economic impacts of chemical pollution on ESs provided by water systems. To this end, we estimated the total ES loss in monetary units as a function of a chemical pollution indicator (i.e. PAF) by subsequently extrapolating PAF to diversity, productivity and total ES value. We applied the derived relationship between PAF and total ES value loss to polluted sediments of waterways in Waal-Meuse river estuary (the Netherlands) and Flanders (Belgium), as monitoring concentration data in sediments were sufficient. We discussed the uncertainties for further development and application of the methodology.

2. Materials and methods

To assess the impacts of sediment pollution of waterways on ESs, we developed a stepwise approach consisting of five steps from (1) to (5) (Fig. 1). Sediment concentrations were first converted into water levels, a common procedure for comparing toxicity data (Section 2.1). The multi-substance PAF of the whole aquatic community (ranging from algae to fish) at the HC50 level was subsequently calculated (msPAF (HC50), Section 2.2). The msPAF(HC50) was linked to diversity based on previous studies (Section 2.3). The association between diversity and total ES value was based on literature review and data analysis, using productivity as a proxy for ESs (Section 2.4 and 2.5). The quantitative relationships between msPAF(HC50) and total ES loss were finally applied to the Waal-Meuse river estuary and Flemish waterways to gain insights into impact magnitudes (Section 2.6).

2.1. Conversion from sediment concentrations into water concentrations

Historically, direct measurement of concentrations in sediment pore water was extremely challenging due to low detection limits, sampling artefacts and chemical interferences (ASTM, 1994; Nolan et al., 2003). Compared with the direct analysis in sediment pore water, total sediment concentrations were easier to measure. Therefore, sediment pore water (hereafter water) concentrations were predicted from measured whole sediment concentrations based on equilibrium partitioning relationships in this study.

2.1.1. Sediment concentrations

The Waal-Meuse river estuary. From 1992 to 1999, sediment pollution was monitored in the Waal-Meuse river estuary. Seven regions were included for field sampling (i.e. Haringvliet (1995), Hollandsch Diep (1993), Nieuwe Merwede (1992), Dordtsche Biesbosch (1993), Brabantsche Biesbosch (1994), Amer (1998), Sliedrechtse Biesbosch (1999)). Sampling methods were described for each location in Postma and Den Besten (2001), and the chemical analyses were performed as described by Den Besten et al. (1995). Concentrations of 46 chemicals were measured in the top-layer (10 cm) of the sediment (Table A1 in Appendix A).

Flemish waterways. The available sediment monitoring data from 2000 to 2015 were derived from the TRIAD assessment programme of the Flemish Environment Agency (www.vmm.be). Locations throughout Flanders were sampled (40 L sediment) approximately every four years in spring using a Van Veen grab sampler (De Deckere et al., 2000). In total, 42 chemicals were analysed (Table A2 in Appendix A).

2.1.2. Conversion from sediment to water concentrations

Due to a lack of information on *in-situ* burrowing behaviour of aquatic species, all species were assumed to be exposed to chemicals in water. As sampling data did not entail information on speciation forms of chemicals, total sediment concentrations ($[C]_{sed}$, mg/kg) were converted to total water concentrations ($[C]_{aq}$, μg/L) via equilibrium partitioning (Van Der Kooij et al., 1991):

$$[C]_{aq} = \frac{r \times [C]_{sed}}{K_{SW}} \quad (1)$$

Fig. 1. Steps developed to assess the environmental impact of chemical mixtures in sediments on ecosystem services (ESs) of waterways. Step (1): Convert sediment concentrations into water concentrations. Step (2): Calculate the multi-substance potentially affected fraction msPAF(HC50) of aquatic species based on water concentrations. Step (3): Link msPAF(HC50) to diversity. Step (4): Identify the quantitative diversity-productivity relationship. Step (5): Update the correlation between productivity and the total value of ESs.

where r is an empirical concentration ratio for suspended matter (taken as 2 and 1 for organics and metals, respectively (Van Der Kooij et al., 1991)) and K_{SW} is the solid-water partition coefficient in L/g.

While K_{SW} values show a great variability depending on physico-chemical factors (Van Der Kooij et al., 1991), only the average binding capacity of metals to sediments was taken into account. We obtained $K_{SW,metal}$ values for metals from Van Der Kooij et al. (1991) assuming to represent the standard 11% and 25% clay (Flanders and Netherlands, respectively) and 5% and 10% organic substances (Flanders and Netherlands, respectively) (Crommentuijn et al., 1997; De Deckere et al., 2000):

$$K_{SW,metal} = \frac{\sum_1^n K_{SW,metal,n}}{n} \quad (2)$$

where n is the sampling locations in the predefined ecosystems/rivers by Van Der Kooij et al. (1991).

Given the high diversity in hydrophobicity for polycyclic aromatic hydrocarbons (PAHs, $10^3 < K_{OW} < 10^7$), $K_{SW,organic}$ for organic chemicals was calculated via the octanol-water partition coefficients (K_{OW}, dimensionless, obtained from Mackay et al. (2006) and Kim et al. (2016)) and the fraction of organic substances (f_{OS}, dimensionless):

$$K_{SW,organic} = 0.6 \cdot K_{OW} \cdot f_{OS} \quad (3)$$

where 0.6 is the empirical conversion factor in L/g (Karickhoff et al., 1979).

2.2. The multi-substance potentially affected fraction of aquatic species

The toxic pressure exerted by chemical mixtures to aquatic communities is expressed as multi-substance PAF at the HC50 level (msPAF (HC50), %), representing the fraction of field species likely affected (De Zwart and Posthuma, 2005). The msPAF(HC50) was calculated based on estimated concentrations of pollutants in water (Section 2.1.2) and SSDs constructed from the laboratory-based toxicity data.

Species sensitivity parameters for each chemical included the hazardous concentration at which 50% of the species in the SSD is affected (HC50, μg/L) for population-level relevant effect criterion (e.g. reproduction, growth, development) and the respective SSD slope (β, dimensionless). Species sensitivity parameters were collected from the RIVM e-toxbase (Posthuma et al., 2019b) (Table A3 in Appendix A). The toxic pressure was first calculated within each chemical group j (i.e. eight individual metals, PAHs, polychlorinated biphenyls (PCBs) and persistent organochlorine pesticides (OCPs)), assuming concentration additivity as an approximation of mixture impacts (De Zwart and Posthuma, 2005):

$$msPAF_j = \frac{1}{1 + e^{-\log\left(\sum \frac{[C]_{aq,i}}{HC50_{aq,i}}\right)/\overline{\beta_j}}} \quad (4)$$

where $[C]_{aq,i}$ is the water concentration and $HC50_{aq,i}$ is the hazardous concentration for the chemical i within the same chemical group. $\overline{\beta_j}$ is the average SSD slope of the chemical group j.

The response addition method was then applied to predict the mixture toxicity across the groups of chemicals (De Zwart and Posthuma, 2005):

$$msPAF(HC50) = 1 - \prod_{j=1}^{11}\left(1 - msPAF_j\right) \quad (5)$$

2.3. Relationship between potentially affected fraction and diversity

Empirical research has shown the ecological relevance of the msPAF (HC50) that the value of diversity indicators (e.g. Shannon-Wiener) reduces when the msPAF(HC50) increases in surface water (De Vries et al., 2010). In the present study, an increase in the msPAF(HC50) was assumed to result in a proportional reduction in benthic diversity (D', %) in various metrics, including species richness, species evenness and functional diversity:

$$D' = 100\% - \text{msPAF(HC50)} \quad (6)$$

2.4. Relationship between diversity and productivity

2.4.1. Rationale

Productivity is related to many ecosystem services such as food or wood provisioning (Millennium Ecosystem Assessment, 2005). Productivity (or aboveground plant biomass in experimental studies) has been reported in diversity-ecosystem functioning research at multiple scales (Costanza et al., 2007), and it has been correlated with the total ES value (Costanza et al., 1998). While using a large number of indicators of ESs (e.g. nutrient cycling, organic matter decomposition) might improve accuracy, the assessment process would become more intensive in terms of data requirements. To explore the full cause-effect chain shown in Fig. 1, we used productivity as a proxy for ESs.

2.4.2. Dataset description

The dataset used in this paper is a filtered and updated version of the datasets published in several meta-analyses (i.e. Cardinale et al. (2006); Balvanera et al. (2006); Daam et al. (2019); Duffy et al. (2017)) reporting experimental and observational evidence of diversity-ecosystem functioning relationships in both terrestrial and aquatic realms. Details on literature search and filtering procedures are described in Appendix B. Our database included 75 studies, 51 of which were conducted in experimental settings (32 in terrestrial and 19 in aquatic ecosystems) and 24 in observational settings (17 in terrestrial and 7 in aquatic ecosystems) (Tables A4–A6 in Appendix A).

2.4.3. Data analysis

Relationships between diversity and productivity were characterised by the effect direction (i.e. positive or 0) and function form (e.g. Michaelis-Menten, linear functions) (Fig. A1 in Appendix A). To allow for comparison and facilitate interpretation of the quantitative diversity-productivity relationships, we performed data scaling and curve fitting for experimental studies in terrestrial ecosystems. Experimental terrestrial studies were chosen since most diversity-productivity research focused on terrestrial systems in the past decades (Loreau et al., 2001;

Wardle, 2016; Daam et al., 2019), and quantitative patterns in aquatic (freshwater, transitional and marine) and terrestrial ecosystems were similar (Fig. A1 in Appendix A and details in Appendix B). Only studies that explicitly reported the mathematical function between diversity and productivity were included in the data scaling and curve fitting.

Data scaling. The reported diversity-productivity relationships were scaled into a 0–100% range without affecting the shape of curves through a linear transformation. The minimum (P_{min}) and the maximum (P_{max}) values of productivity (e.g. aboveground biomass in g/m^2) were calculated from the minimum (D_{min}) and the maximum (D_{max}) value of diversity measurements (e.g. species richness) according to the reported mathematical functions. The values of productivity P and diversity D were scaled to P' and D' in the range of 0–100% according to:

$$P' = (P - P_{min})/(P_{max} - P_{min}) \times 100\% \quad (7.1)$$

and

$$D' = (D - D_{min})/(D_{max} - D_{min}) \times 100\% \quad (7.2)$$

Curve fitting. The average, minimum and maximum impact (the upper and lower boundary encompassing all the reported diversity-productivity curves) of diversity loss on productivity were identified through optical fitting. The decreasing power function ($P' = D'^{\theta}$, $\theta < 1$), Michaelis-Menten and linear functions represented the average, minimum and maximum impact, respectively.

2.5. Relationship between productivity and total ecosystem services

2.5.1. Rationale

While productivity has been related to the total ES value (Costanza et al., 1998), estimates of productivity and total ES value for each biome have changed over the years due to environmental changes (e.g. climate change, El Niño events) and improved estimation methods. Therefore, an updated quantitative linear correlation between the logarithmically transformed value of mean productivity (P, kg/m^2/yr) and total ES value (V, 2007$/ha/yr) across biomes was provided (Costanza et al., 2007):

$$\log_{10}(V) = a + b \times \log_{10}(P) \quad (8)$$

wherein a and b are empirical fitting constants (Costanza et al., 2007).

2.5.2. Dataset description

The fitting constants a and b were determined based on literature data. Papers that reported the total ES value for each biome (i.e. the sum of the mean value of each ES within the biome) were included (Costanza et al. (1997); De Groot et al. (2012); Costanza et al. (2014)) (Fig. A2 in Appendix A). A total of 12 biomes and 17 ESs were included (Table A7 in Appendix A). To update the quantitative correlation between P and V, we chose to use the most recent available data from Costanza et al. (2014) and calculated the geometric mean of productivities for each biome (Table A8 in Appendix A).

2.6. Application of derived relationships

Based on the stepwise approach described above, the relative total ES value loss was estimated as a function of msPAF(HC50). To obtain an absolute value of ES loss due to chemical mixtures in waterways, we considered the sediments in the Waal-Meuse river estuary and Flemish waterways as estuarine/freshwater sediments (Den Besten et al., 2003; De Deckere et al., 2011). Therefore, the geometric mean of ES value provided by lakes/rivers, estuaries, and swamps/floodplains reported in Costanza et al. (2014) was applied to represent the sediment ecosystems in the present study.

3. Results

3.1. Multi-substance potentially affected fraction

The Waal-Meuse river estuary. The average mixture toxic pressure (msPAF(HC50)) for sediment samples in the Waal-Meuse river estuary was estimated to be 18.5% (±6.3%) (Table 1). Metals (10.1%) and PAHs (8.7%) were the main contributors to the total pressure, while OCPs (0.5%) and PCBs (0.2%) had limited contribution.

Flemish waterways. The msPAF(HC50) for Flemish sediments was estimated to be 35.6% (±21.1%) (Table 2). The mixture toxic pressure in the Waal-Meuse river estuary (18.5%) was lower than that of the Meuse river in Flanders (25.5%), as the relatively clean Waal river diluted the Meuse river. It was estimated that the toxic pressure of three rivers in Flanders was lower than that of all waterways, indicating that other waterways (e.g. canals, streams) were more heavily polluted. The chemical groups exerting the highest pressure to the ecosystems were those of PAHs, nickel and copper.

3.2. Relationship between diversity and productivity

Through curve fitting, the 'average', 'minimum' and 'maximum' diversity loss impact on productivity were represented with the power ($P' = D'^{0.35}$ as approximately 50% and 80% of productivity was still maintained at 15% and 50% of pristine species richness, respectively), Michaelis-Menten ($P' = 1.04 \times \frac{D'}{D'+0.04}$) and linear functions ($P' = D'$), respectively (Fig. 2). In other words, x% of msPAF(HC50) (i.e. (100-x)% of remaining diversity) would relate to an average of $\left(1-\left(1-\frac{x}{100}\right)^{0.35}\right) \times 100\%$ (from minimum $\frac{4x}{104-x}$% to maximum x%) of productivity loss.

3.3. Relationship between productivity and total ecosystem service value

An increase of productivity generally led to an increase of total ES value of each biome (Fig. 3, note the logarithmic axes for both variables). Regardless of the differences between terrestrial and aquatic systems, the fitted relationship between the geometric mean of productivity (P, kg/m^2/yr) and total ES value (V, 2007$/ha/yr) was: $\log_{10}(V) = 4.14 + 1.40\log_{10}(P)$ with $R^2 = 0.53$. In other words, x% of productivity loss resulted in $(1-(1-\frac{x}{100})^{1.4}) \times 100\%$ of total ES value loss.

3.4. Application of derived relationships

The relationships between msPAF(HC50) and diversity, between diversity and productivity in terrestrial ecosystems, and between pro-

Table 1
The msPAF(HC50) (%) for sediments in the Waal-Meuse river estuary.

Chemical groups	msPAF per distributary (%)[b]							$\overline{msPAF}$
	HV	HD	DB	NM	AM	BB	SB	(%)[c]
Metals[a]	7.0	9.4	13.2	10.3	10.7	9.8	11.3	10.1 (±1.8)
PAHs	8.0	8.0	10.4	7.6	9.9	10.0	7.6	8.7 (±1.2)
OCPs	0.1	1.1	7.1	2.0	0.2	0.1	0.4	0.5 (±2.4)
PCBs	0.1	0.1	0.5	0.7	0.2	0.1	0.3	0.2 (±0.2)
msPAF(HC50)(%)	14.6	17.6	28.0	19.4	19.9	19.0	18.6	18.5 (±6.3)

[a] Metals include arsenic, cadmium, chromium, copper, mercury, nickel, lead and zinc.
[b] Distributary code: Haringvliet (HV), Hollandsch Diep (HD), Dordtsche Biesbosch (DB), Nieuwe Merwede (NM), Amer (AM), Brabantsche Biesbosch (BB), Sliedrechtse Biesbosch (SB).
[c] The geometric mean and ±1 standard deviation based on msPAF per distributary.

Table 2
The msPAF(HC50) (%) and ±1 standard deviation for three rivers and all waterways in Flanders.[a]

Chemical groups	msPAF (%)			
	Meuse river	Scheldt river	Yser river	All waterways in Flanders
Metals[b]	9.2 (±1.7)	10.1 (±3.7)	9.3 (±2.3)	9.4 (±4.3)
PAHs	17.3 (±2.5)	20.2 (±5.2)	13.6 (±4.0)	23.1 (±10.9)
PCBs	0.6 (±0.2)	0.6 (±0.4)	0.2 (±0.2)	0.7 (±1.0)
OCPs	0.2 (±0.1)	5.0 (±9.4)	2.6 (±4.2)	7.0 (±19.2)
msPAF(HC50) (%)	25.5 (±11.1)	32.2 (±18.6)	23.9 (±12.1)	35.6 (±21.1)

[a] All waterways in Flanders included the Meuse, Scheldt, Yser rivers and other waterways in Flanders.
[b] Metals include arsenic, cadmium, chromium, copper, mercury, nickel, lead and zinc.

ductivity and total ES value were combined. Consequently, an x% of msPAF(HC50) implies an average reduction in total ES value (V_{loss}, %) of (Fig. 4):

$$V_{loss,avg} = \left(1 - \left(1 - \frac{\text{msPAF(HC50)}}{100}\right)^{0.49}\right) \times 100\% \quad (9.1)$$

with minimum ES loss of

$$V_{loss,\max} = \left(1 - \left(1 - \frac{0.04 \times \text{msPAF(HC50)}}{104 - \text{msPAF(HC50)}}\right)^{1.4}\right) \times 100\% \quad (9.2)$$

and maximum ES loss of

$$V_{loss,avg} = \left(1 - \left(1 - \frac{\text{msPAF(HC50)}}{100}\right)^{1.4}\right) \times 100\% \quad (9.3)$$

As an outcome of the complete set of assessment steps, 1% of msPAF (HC50) was estimated to correspond to on average 0.5% (minimum-maximum: 0.05–1.40%) of total ES value loss. Chemical mixtures in sediments of Waal-Meuse river estuary and all waterways in Flanders would reduce 9.5% (1.20–24.9%) and 19.4% (2.9–46.0%) of total ES value, resulting in an ES loss of 0.3–5 and 0.6–10 thousand 2007$/ha/yr, respectively (Table 3).

4. Discussion

So far, the economic impacts of chemical pollution on ESs provided by the ecosystem have not been quantified. Therefore, we developed a novel methodology for assessing the impacts using a stepwise approach, based on msPAF(HC50)-diversity-productivity-total ES value relationships. The present study is, to our best knowledge, the first to derive and apply a holistic outline to estimate changes in total ES value as a function of changes in the chemical pollution indicator (i.e. mixture toxic pressure, msPAF(HC50)). The feasibility and potential utility of the methodology was illustrated by an application to chemical pollution in Dutch and Flemish sediments. The economic outcome may serve as a necessary component in the cost-benefit analysis regarding waterway quality improvement measures. With sufficient monitoring data, our generic relationships can be applied to quantify the economic impacts of chemical pollution on water systems for other chemicals and regions of interest. Below, we discuss the three main steps of our study, i.e. the msPAF-diversity, diversity-productivity, and productivity-total ES value relationship, respectively.

4.1. The msPAF-diversity relationship

The local mixture toxic pressure (msPAF(HC50) of species) was calculated from converted water concentrations and SSDs, combined with standard correction of bioavailability and mixture modelling. An average msPAF(HC50) of 18.5% in the Waal-Meuse river estuary was close to the median of 20% estimated by Posthuma and De Zwart (2012) using a smaller toxicity database for the same region in the Netherlands.

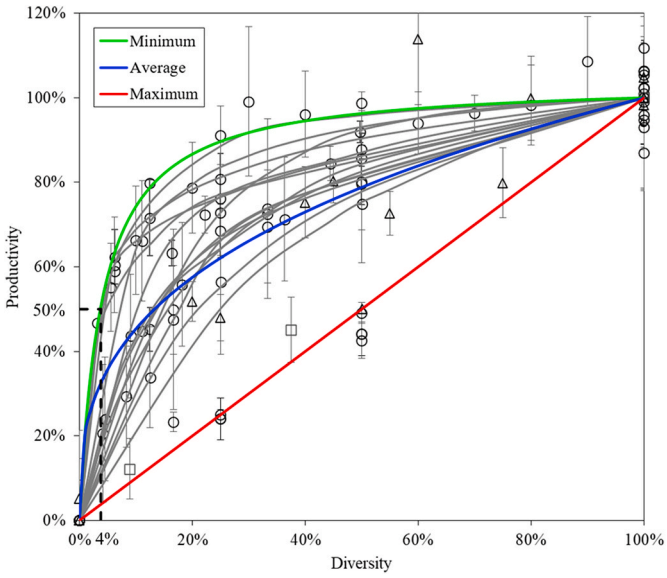

Fig. 2. Relationships between diversity (D', %) and productivity (P', %) in experimental terrestrial ecosystems based on reported scaled relationships (solid grey curves). The average, minimum and maximum diversity loss impact on productivity are shown in solid blue, green and red curves, respectively. Minimum impact shows that 96% of diversity loss results in 50% of productivity loss (dashed black line). Type of diversity is classified as species richness (open circles), functional richness (open triangles) and species evenness (open squares). Means and standard errors are shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Notably, toxicity is associated with the fraction of the chemical that is freely bioavailable (i.e. bioavailability), which was roughly approximated via the average binding capacity of metals. Toxicity may also vary substantially depending on the different forms that chemicals exist in Vink (2009). This is prominent for metals, whereas only total sediment and aqueous concentrations were available. We envision that a proportion of variance in the msPAF (Tables 1 and 2) may characterise uncertainties due to unknown speciation and bioavailability.

To date, the ecological relevance of the msPAF is being explored mainly for surface water. For instance, the value of the biodiversity indicators generally reduces when the msPAF(HC50) increases (De Vries et al., 2010). The ecological status of European surface waters was demonstrated to increase with a decrease of msPAF(HC50) (Posthuma et al., 2020). Such correlations are increasingly underpinned by mechanistic approaches, relating msPAF(HC50) to mean species abundance (MSA) and field occurrences (Hoeks et al., 2020; Thunnissen et al., in preparation). For sediments, the fraction of benthic taxa with the observed abundance reduction of more than 50% was almost proportional with the calculated msPAF(HC50) (Posthuma and De Zwart, 2012). Based on the evidence, a proportional diversity loss of taxa was

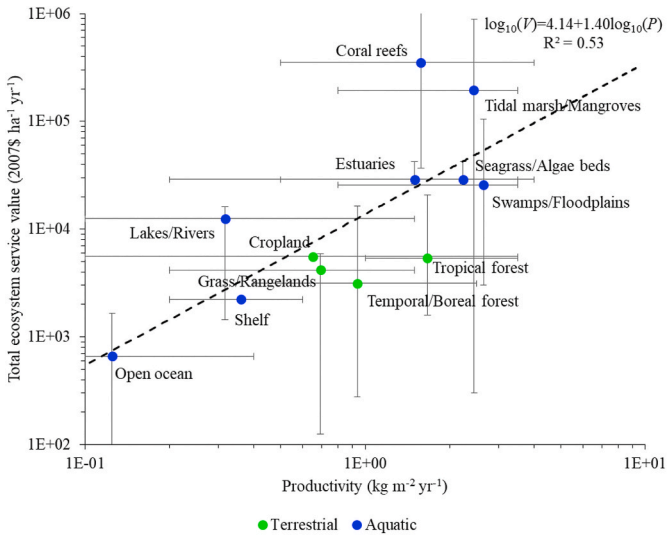

Fig. 3. Relationship between productivity (P, kg/m^2/yr) and total ecosystem service value (V, 2007$/ha/yr). Bars show the range of productivity and the total value of ecosystem services per biome based on Whittaker (1975) and De Groot et al. (2012), respectively. The black dashed line is a regression of total ecosystem service estimates from Costanza et al. (2014) on the calculated geometric mean of productivity based on literature listed in Table A8 in Appendix A.

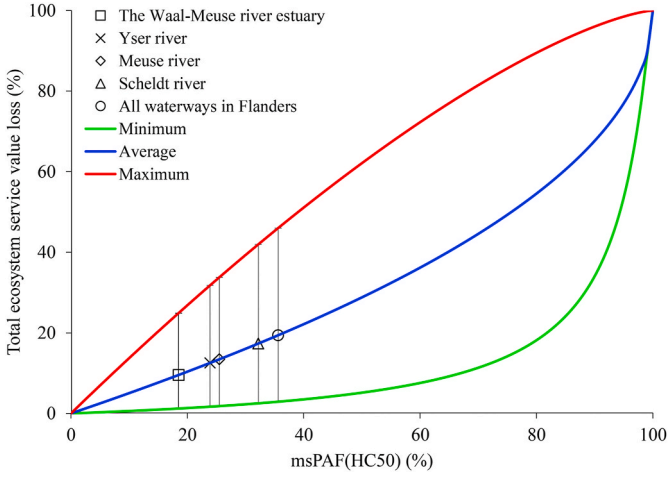

Fig. 4. Relationship between multi-substance potentially affected fraction of species (msPAF(HC50), in %) and total ecosystem service (ES) value loss (in %). The average, minimum and maximum toxic pressure impacts on total ES value loss are shown in solid blue, green and red curves, respectively. Sediment sampling sites are shown in different shapes. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

assumed associated with an increase in msPAF(HC50). However, such relationships in sediments are generally weaker than those in surface water. Therefore, it is recommended to test the direct relationship between msPAF and macrofauna species diversity for various types of sediment under field conditions.

4.2. Diversity-productivity relationship

Regardless of the inherent differences between terrestrial and aquatic ecosystems (e.g. phylogenetic diversity, ecological processes), diversity generally increases with ecosystem productivity, as reported in the present study (Fig. 2) and by others (e.g. Stachowicz et al. (2007); Cardinale et al. (2012); Tilman et al. (2014); Strong et al. (2015)). In the last few decades, the ecological consequences of diversity loss have become a central issue in the field of ecological and environmental sciences, while experimental work has mainly been carried out in grassland ecosystems (Loreau et al., 2001; Giller et al., 2004; Hooper et al., 2005; Gustafsson and Boström, 2011). Early studies have explored two main classes of underlying mechanisms (while contradictory) for how organisms promote productivity (e.g., Loreau et al. (2001); Mulder et al. (2001); Cardinale et al. (2007); Tilman et al. (2014); Daam et al. (2019)). 'Complementarity effects' relate to better performance in diverse communities due to niche partitioning and facilitation in shared resource use, while 'sampling effects' refers to an increased probability of including highly productive dominant species in diverse communities. While a summary of diversity-productivity mechanisms allows a better understanding of the results presented in this paper, a detailed discussion is beyond the scope of this study.

Our study suggested that a 1% diversity loss corresponds to on average 0.35% (0.04–1.00%) productivity loss. The result was consistent with the findings from Costanza et al. (2007), reporting that a 1% change in vascular plant diversity in warm ecoregions corresponds to a 0.173% change in productivity based on multiple regression analysis at the ecoregion scale in North America. Our study also suggested that productivity could increase monotonically with diversity as $P' \sim D'^{\theta}$ ($\theta = 0.35$ in the study). The result of θ was consistent with that reported in other studies, ranging from 0.1 to 0.5 in algae, grasses, shrubs and trees (Liang et al., 2016; Duffy et al., 2017; Chen et al., 2018).

It should be noted that the present quantitative diversity-productivity relationships were derived from evidence in terrestrial experiments. While our study showed similar quantitative diversity-productivity patterns in aquatic realms (freshwater, transitional and marine) and observational studies (Fig. A1 in Appendix A), uncertainties remain. For example, compared to terrestrial systems, aquatic systems are considered more complex due to multitrophic interactions, faster biological processes, greater propagule and material exchange, and often steeper physical and chemical gradients (Giller et al., 2004; Gamfeldt et al., 2015; Daam et al., 2019). As a result, the direct extrapolation of conclusions on diversity effects on productivity from terrestrial experiments might be limited. Besides, positive concave-up diversity-productivity patterns (i.e. exponential) were reported in two large-scale observational studies on deep-sea nematodes and coral reef fishes (Danovaro et al., 2008; Mora et al., 2011). The loss of species is likely to affect the functioning of natural ecosystems more than would be expected from manipulation experiments (Mora et al., 2014). Therefore, given the differences between terrestrial and aquatic ecosystems and

Table 3
Total ecosystem service loss (in 2007$/ha/yr) due to chemical pollution in the Waal-Meuse river estuary and Flemish waterways.

Site	msPAF(HC50) (%)[a]	Productivity loss (%)[b]	Total ES loss (%)[b]	Total ES value (2007$/ha/yr)[c]	Total ES loss (2007$/ha/yr)[b]
The Waal-Meuse river estuary	18.5 (±6.3)	6.9 (0.9–18.5)	9.5 (1.2–24.9)	21023 (±7095)	2005 (255–5242)
Flemish waterways	35.6 (±21.1)	14.3(2.1–35.6)	19.4 (2.9–46.0)		4078 (611–9679)

[a] The average mixture toxic pressure (±1 standard deviation).
[b] The average value (minimum-maximum). The range only reflects the conversion from diversity to productivity based on the average mixture toxic pressure.
[c] The geometric mean (±1 standard deviation) of ecosystem services provided by lakes/rivers, estuaries and swamps/floodplains based on Costanza et al. (2014).

differences between experimental and observational studies, further study is still needed to improve our understanding of the mechanisms behind the diversity-productivity relationships across a range of ecosystems, or diversity-ecosystem functioning relationships in general.

4.3. Productivity-total ES value relationship

The differences in total ES value among biomes could be generally explained by the differences in productivity, with higher productivity resulting in higher ES values (this study and others, e.g. Egoh et al. (2008); Thomas et al. (2013)). Therefore, productivity as a proxy for ESs could facilitate and simplify quantitative assessments of ESs as productivity can be measured through remote sensing over space and time (Costanza et al., 2017). However, we consider our productivity-ES relationship as a necessary but still highly uncertain step in the approach (note the logarithmic axes for both variables in Fig. 3). The total ES value for each biome on average varied two orders of magnitude due to the heterogeneity of original data in terms of valuation methods, ES definitions, and socio-economic characteristics in different locations and periods (De Groot et al., 2012). Besides, for most biomes, less than half of the total number of services (potentially 22 recognised services) were valued (De Groot et al., 2012). Consequently, the total ES value shown in Fig. 3 is an underestimate of the economic importance of each biome/ecosystem. Moreover, the economic impacts of polluted sediments were estimated based on the average ES value provided by estuarine or freshwater ecosystems (lakes/rivers, estuaries, and swamps/floodplains). As sediments make up the bottom of these ecosystems, we envision that individual case studies (i.e. the original ES value) considered the essential ecosystem functions of sediments (e.g. habitat provision, flood protection). Nevertheless, in the absence of total ES value specifically for sediments, it is recommended to understand the broad roles of sediments as essential and dynamic components in ecosystems, and to evaluate the ecological benefits of sediments.

To date, ES valuation tends to focus on individual estimates of specific ESs value for a particular biome or ecosystem at local or regional scales for uses such as urban land use planning and specific policy analysis. On one hand, the individual ES estimates for each biome may reflect the interdependencies among ESs, biome types and valuation methods (Schild et al., 2018). On the other hand, the aggregation of ES values for (global) ecosystems or biomes is also necessary, especially in the case of raising awareness of nature's contribution to people (Costanza et al., 2017). The aggregation of ES values is free from data limitations (e.g. explicit information on ecological and socio-economic contexts), and is feasible to explore overarching principles. Therefore, the individual estimates for specific ESs at a local scale and the overall aggregated estimates at a larger scale could supplement each other, allowing for a more comprehensive understanding and assessment on the importance of nature to human well-being.

Overall, 1% of msPAF(HC50) was estimated to correspond to on average 0.5% (0.05–1.40%) of total ES loss. As our study is the first to assess chemical pollution impacts on ES values, a direct comparison of our methodology and results with other studies is difficult. However, ongoing studies corroborate that approximately one-third of the variability in the ecological status of European surface waters can be attributed to chemical mixtures (Posthuma et al., 2019a). In terms of the magnitude of adverse effects of chemical pollution on ecosystem health, our results are similar to those from Posthuma et al. (2019a). Our methodology could potentially serve as an additional risk assessment tool for informed decisions on water quality management, as the economic outcome is understandable to policymakers given the practical decision context.

4.4. Outlook and recommendation

Although this paper explicitly focuses on the use of monetary units to express ESs, we would like to point out our work does not suggest that nature should be treated as private commodities that can be traded in private markets, nor does it imply that nature only exists to 'serve' humans. Instead, our message is simple: ecosystems are essential to human well-being and deserve social recognition. Quantification of ESs in financial terms may help society make better decisions, especially in the many cases where trade-off exists (e.g. land use options). Monetary value is understandable and easy to communicate, and could provide efficient use of limited funds in the context of nature conservation and restoration. Additionally, the explicit valuation of ESs is transparent, which is a crucial component in democratic decision processes. Therefore, the valuation of ESs is inevitable to demonstrate the crucial contribution of ecosystems to societies.

The major achievement of this study is the stepwise approach to quantifying the impacts of chemical exposures on diversity, productivity and total value that ecosystems provide. The case studies of polluted sediments in the Netherlands and Flanders are intended to illustrate the impact magnitude in monetary units and the feasibility of the proposed approach. We consider our study as a first move linking pollutant concentrations to ecosystem service loss. Given the rough estimates in the present study, we identified the main uncertainties in each step and recommended further research for developing the proposed methodology. We encourage the application and refinement of this systematic methodology, which will contribute to thorough assessments, management and communication of chemical risks to ecosystems.

Author contribution

Jiaqi Wang: Methodology, Data Curation, Writing - Original Draft, Visualization. Leonie S. Lautz: Data Curation, Writing - Review & Editing. Tom M. Nolte: Data Curation, Writing - Review & Editing. Leo Posthuma: Data Curation, Writing - Review & Editing. K. Remon Koopman: Writing- Reviewing and Editing. Rob S.E.W. Leuven: Writing-Reviewing and Editing. A. Jan Hendriks: Conceptualization, Writing - Review & Editing, Supervision.

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.jenvman.2020.111873.

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