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Enhancement of the impact score assessment of micropollutants release from WWTP using predicted characterization factors

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Abstract

During wastewater treatment, incomplete elimination of micropollutants occurs: effluents released to the environment still contain a non-negligible part of these substances. Recently, the potential impacts on human health and aquatic environment of the release of some micropollutants have been studied. The high impacts were mainly due to the (eco)-toxicological potential of a few numbers of compounds, not to their mass emitted in the environment. Furthermore, the impact was estimated for less than 1/3 of the initial list of substances due to a lack of concentration and toxicological data. In the present communication, we used already developed machine learning models to complement these impact calculations. The conclusions were not modified for the impact on aquatic environment, but were different for the human health impact: the higher toxicological impact could be driven by a high-emitted mass, and a high number of compounds could take a significant part of the overall impact.

Keywords

Characterization factors; continental freshwater; (eco)-toxicity; human health; Life Cycle Assessment; molecular descriptors.

INTRODUCTION

For several years, the presence of organic micropollutants (pharmaceuticals, pesticides, Polycyclic Aromatic Hydrocarbons (PAH)...) in the effluents of wastewater treatment plants (WWTP) is ubiquitous and has raised increasing concerns. More than thousands of active substances are identified in wastewater and their treatment within the plants remains incomplete (Aemig et al., 2021). In parallel, with increasingly accurate and efficient analytical technologies, more and more compounds are detected at low concentrations (ng.L^{-1} to $\mu\text{g.L}^{-1}$) in rivers, groundwater, surface water, and drinking water. The identification and quantification of the potential impact of toxic substances induce the development of new sustainable process technologies of targeted substances and represent a great challenge for the safety of aquatic ecosystems (Rosenbaum et al., 2007).

Recently, Aemig et al. (2021) gathered data on 261 organic micropollutants in the effluents of French WWTP from literature. The toxicological and ecotoxicological impacts of 94 and 88 micropollutants, respectively, were quantified by multiplying the emitted mass of the compound in the total volume of effluents from French WWTPs by their characterization factors (Lindim et al., 2019). Characterization factors (CF) are used in the Life Cycle Assessment (LCA) framework to represent the human toxicity or the freshwater ecotoxicity. Results showed that a molecule can be highly toxic for the aquatic environment without necessarily being toxic for human health, and vice versa. Moreover, a high concentration does not necessarily lead to a high impact, and in the same way, a molecule with a low concentration can lead to a significant impact. The CF seemed to be the most important variable to explain the potential impact of a substance because the differences in terms of CF were more important than in terms of mass. The authors also pointed out that there was a significant lack of toxicological and ecotoxicological CF for many substances to draw conclusions. Up to now, if a CF is missing for any substance, the potential impact of these substances could not be estimated (*i.e.* is set equal to zero) leading to an underestimation of the

potential overall impacts.

Recently, Servien et al. (2022) developed a modeling method based on machine learning approaches and 40 easy-to-obtain molecular descriptors. This approach allowed the prediction of toxicological and ecotoxicological CF in continental freshwater with an acceptable margin of error. Applying these models to estimate the missing CF could bring the assessment of the overall potential impacts closer to reality.

The objective of this communication was thus to predict missing toxicological and ecotoxicological factors using the machine learning models developed by Servien et al. (2022). This allowed a completed assessment of the overall potential impacts (on human health and on aquatic environment) of 153 organic micropollutants in continental freshwater at the scale of France.

MATERIALS AND METHODS

Molecules

The 261 organic micropollutants identified by Aemig et al. (2021) came from (i) the Water Framework Directive (WFD, Directive 2008/105/CE), (ii) the RSDE national action for survey and reduction of hazardous substances in water (INERIS, 2016), and (iii) the AMPERES French project (Martin Ruel et al., 2012). Among these 261 organic micropollutants, the emitted mass of 153 was estimated with 90% of the measured data above the limit of quantification.

Characterization factors

Among the 153 micropollutants, Aemig et al. (2021) identified approximately 90 compounds with characterization factors for aquatic environment (CF_{ET}) and/or for human health (CF_{HT}) for emissions in continental freshwater in the USEtox[®] (Rosenbaum et al., 2007) database, version 2.12. USEtox[®] is an international consensual for characterizing human and ecotoxicological impacts of chemicals (UNEP-SETAC, 2019). To perform the calculation of the impacts of the 153 compounds, we computed the missing ecotoxicological and toxicological characterization factors, using the machine learning models developed by Servien et al. (2022). These models predict ecotoxicological or toxicological characterization factors using 40 selected easy-to-obtain molecular descriptors, chosen in Servien et al. (2014). These models are based on a comparison between global and cluster-then-predict approaches with partial least squares, support vector machines, and random forest. The best approaches were then selected for each cluster and each characterization factor. Then, the models selected in Servien et al. (2022) were applied without any modification, assuming they had been previously tested on a large diversity of molecules covering the diversity of the compounds assessed in the present study.

Calculation of molecular descriptors

CHEM-3D of ChemOffice Ultra 12.0 (2017) molecular modeling software was used to build three-dimensional chemical structures (3D-structures) in order to calculate the quantum-chemical molecular descriptors. The Excel function of ChemOffice was then used to calculate the molecular weights and the Connolly surface areas. Finally, the constitutional (except the molecular weight) and the topological descriptors were calculated with Dragon 7.0 (2017).

Quantification of the potential impacts

The concentrations and masses are those of the Supplemental Material of Aemig et al. (2021). Total impacts on human health and aquatic environment in continental freshwater were quantified by summing the impacts of all the compounds, as it is usually done in LCA. Human health impact is expressed in DALY (Disability-Adjusted Life Years) representing the number of negatively impacted human years, and the ecotoxicological impact is expressed in PDF (Potentially

Disappeared Fraction of species) representing the potential fraction of disappeared species.

RESULTS AND DISCUSSION

Global analysis

The addition of new molecules more than doubled the total emitted mass (from 71.1 and 64.5 tons to 147.1 tons) of micropollutants released into freshwater in one year. We can notice that the added compounds have globally a higher CF than those of Aemig et al. (2021), but that the more extreme compounds, with the highest CF values, were already included in their study.

Impact on the aquatic environment

Aemig et al. (2021) showed that 99% of the total impact was induced by only 2% of the total emitted mass and 10 molecules. Thus, as a comparison with the addition of 65 molecules:

- 99% of the impact is now induced by 24 molecules and 21% of the total mass,
- the 5 molecules with the highest impact represent 95% of the impact and 0.9% of the total mass,
- among the 5 most impactful, one was different from the previous study (Table 1),
- the total impact has increased by only 4%.

The total impact is still due to only a small number of molecules and a small amount of the total mass. It could have been expected that the addition of a set of compounds with a global higher CF_{ET} would increase a lot the global impacts. However, these impacts are mainly due to extreme values of CF_{ET} that are not present in our new set of compounds.

Table 1. List of the 5 molecules with the highest aquatic environment impact. The underlined molecules were not taken into account in Aemig et al. (2021).

CAS Number	Name	Impact (%)	Emitted mass (%)	Emitted mass (kg)	CF _{ET} (PDF/kg)	PDF
52315-07-8	Cypermethrin	79.21	0.47	696.3	7.17E+07	4.99E+10
37680-73-2	PCB 101	11.15	0.03	47.1	1.49E+08	7.03E+09
50-28-2	17- β -estradiol	2.28	0.01	9.6	1.50E+08	1.44E+09
<u>154-21-2</u>	<u>Lincomycin</u>	1.43	0.31	450.0	2.00E+06	9.01E+08
26787-78-0	Amoxicillin	1.09	0.07	100.0	6.87E+06	6.87E+08
Total		95.16	0.89	-	-	-

Impact on Human health

As a comparison, with Aemig et al. (2021) with the addition of the 59 new molecules:

- 94% of the impact is now induced by 26 (prev. 8) molecules and 41% (prev. 4%) of the total mass,
- the 8 molecules with the highest impact represent 70% of the impact and 27% of the total mass,
- on the 20 molecules that are identified as the most impactful, 13 molecules were not incorporated in the previous study. This now represents 92% of the impact and 32% of the total mass,
- the total impact has nearly doubled.

The main conclusion of the study of Aemig et al. (2021) does not stand anymore for the CF_{HT} . First, the high impact of the most impacting compound (valsartan) is due to its very high emitted mass, nearly 18% of the total mass (Table 2). Depending on the compound, a high impact could be driven by a high mass, a high CF_{HT} , or a combination of both. Second, the number of molecules needed to reach 94% of the total impact has been multiplied by more than three (8 to 26) highlighting a more equal distribution of the impacts among the compounds. In the ten most impactful molecules, six have been added by the present study. Among these six compounds, two are anti-hypertensive drugs, valsartan, and irbesartan, which contributed to the impact thanks to their high masses released in the environment that could be linked to their high uses and low removal in WWTP. Then, contrarily to the aquatic impacts, the total impact on human health has nearly doubled

compared to the results of Aemig et al. (2021), leading to the conclusion that evaluating the human health impacts based only on the available CF_{HT} could provide very underestimated impacts.

Table 2. List of the 10 molecules with the highest human health impact. The underlined molecules were not taken into account in Aemig et al. (2021).

CAS Number	Name	Impact (%)	Emitted mass (%)	Emitted mass (kg)	CF _{HT} (DALY/kg)	DALY
<u>137862-53-4</u>	<u>Valsartan</u>	14.959	17.7	26067.4	6.54E-05	1.71E+00
205-99-2	Benzo(b)fluoranthene	14.621	0.04	61.9	2.69E-02	1.67E+00
207-08-9	Benzo(k)fluoranthene	8.182	0.04	52.7	1.77E-02	9.33E-01
53-86-1	Indomethacin	7.581	0.64	940.2	9.19E-04	8.64E-01
115-32-2	Dicofol	6.847	0.17	243.8	3.20E-03	7.81E-01
<u>138402-11-6</u>	<u>Irbesartan</u>	6.600	8.05	11848.7	6.35E-05	7.52E-01
193-39-5	Indeno(1,2,3-cd)pyrene	6.283	0.04	55.5	1.29E-02	7.16E-01
<u>465-73-6</u>	<u>Isodrin</u> <u>1,3,5,7,9,11-</u>	4.526	0.04	54.9	9.41E-03	5.16E-01
<u>25637-99-4</u>	<u>Hexabromocyclododecane</u>	4.059	0.20	296.2	1.56E-03	4.63E-01
32534-81-9	Pentabromodiphenylethers	3.094	0.01	12.5	2.82E-02	3.53E-01
Total		76.75	26.9	-	-	-

CONCLUSION

Using a new modeling approach, we were able to estimate new characterization factors and then calculate the impact of 153 organic micropollutants. It has been shown that, depending on the substances, a high potential impact could be due to a high emitted mass and/or a high characterization factor, especially for human health. These results could be used to select substances on which a special effort should be made on the tertiary treatments to implement in WWTP. Therefore, to estimate the impacts of 100% of the initial dataset of substances, the lack of data on the mass emitted in the environment is now the only remaining limitation. These data could be predicted with consumption models and WWTP fate models. It has to be underlined that this whole methodology can be adapted to any other compartment and any other geographical context, with predictive models still to develop.

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