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Reliability of methods used for the risk assessment of mixture of micropollutants in aquatic environments: from theoretical concepts to field observations

Summary

For several years now, the scientists as well as the society is concerned by the aquatic risk organic micropollutants may pose. Indeed, several researches have shown the toxic effects these substances may induce on organisms living in our lakes or rivers, especially when they are exposed to acute or chronic concentrations. However, most of the studies focused on the toxicity of single compounds, i.e. considered individually. The same also goes in the current European regulations concerning the risk assessment procedures for the environment of these substances. But aquatic organisms are typically exposed every day simultaneously to thousands of organic compounds. The toxic effects resulting of these “cocktails” cannot be neglected. The ecological risk assessment of mixtures of such compounds has therefore to be addressed by scientists in the most reliable and appropriate way.

In the first part of this thesis, the procedures currently envisioned for the aquatic mixture risk assessment in European legislations are described. These methodologies are based on the mixture model of concentration addition and the use of the predicted no effect concentrations (PNEC) or effect concentrations (EC50) with assessment factors. These principal approaches were applied to two specific case studies, Lake Geneva and the River Rhône in Switzerland, including a discussion of the outcomes of such applications. These first level assessments showed that the mixture risks for these studied cases exceeded rapidly the critical value. This exceeding is generally due to two or three main substances. The proposed procedures allow therefore the identification of the most problematic substances for which management measures, such as a reduction of the entrance to the aquatic environment, should be envisioned. However, it was also showed that the risk levels associated with mixtures of compounds are not negligible, even without considering these main substances. Indeed, it is the sum of the substances that is problematic, which is more challenging in term of risk management. Moreover, a lack of reliability in the procedures was highlighted, which can lead to contradictory results in terms of risk. This result is linked to the inconsistency in the assessment factors applied in the different methods.

In the second part of the thesis, the reliability of the more advanced procedures to predict the mixture effect to communities in the aquatic system were investigated. These established methodologies combine the model of concentration addition (CA) or response addition (RA) with species sensitivity distribution curves (SSD). Indeed, the mixture effect predictions were shown to be consistent only when the mixture models are applied on a single species, and not on several species simultaneously aggregated to SSDs. Hence, A more stringent procedure for mixture risk assessment is proposed, that would be to apply first the CA or RA models to each species separately and, in a second step, to combine the results to build an SSD for a mixture. Unfortunately, this methodology is

not applicable in most cases, because it requires large data sets usually not available. Therefore, the differences between the two methodologies were studied with datasets created artificially to characterize the robustness of the traditional approach applying models on species sensitivity distribution. The results showed that the use of CA on SSD directly might lead to underestimations of the mixture concentration affecting 5% or 50% of species, especially when substances present a large standard deviation of the distribution from the sensitivity of the species. The application of RA can lead to over- or underestimations, depending mainly on the slope of the dose-response curves of the individual species. The potential underestimation with RA becomes important when the ratio between the EC50 and the EC10 for the dose-response curve of the species composing the SSD are smaller than 100. However, considering common real cases of ecotoxicity data for substances, the mixture risk calculated by the methodology applying mixture models directly on SSDs remains consistent and would rather slightly overestimate the risk. These results can be used as a theoretical validation of the currently applied methodology. Nevertheless, when assessing the risk of mixtures, one has to keep in mind this source of error with this classical methodology, especially when SSDs present a distribution of the data outside the range determined in this study

Finally, in the last part of this thesis, we confronted the mixture effect predictions with biological changes observed in the environment. In this study, long-term monitoring of a European great lake, Lake Geneva, provides the opportunity to assess to what extent the predicted toxicity of herbicide mixtures explains the changes in the composition of the phytoplankton community next to other classical limnology parameters such as nutrients. To reach this goal, the gradient of the mixture toxicity of 14 herbicides regularly detected in the lake was calculated, using concentration addition and response addition models. A decreasing temporal gradient of toxicity was observed from 2004 to 2009. Redundancy analysis and partial redundancy analysis showed that this gradient explains a significant portion of the variation in phytoplankton community composition, even when having removed the effect of all other co-variables. Moreover, some species that were revealed to be influenced positively or negatively, by the decrease of toxicity in the lake over time, showed similar behaviors in mesocosms studies. It could be concluded that the herbicide mixture toxicity is one of the key parameters to explain phytoplankton changes in Lake Geneva.

To conclude, different methods exist to predict the risk of mixture in the ecosystems. But their reliability varies depending on the underlying hypotheses. One should therefore carefully consider these hypotheses, as well as the limits of the approaches, before using the results for environmental risk management.