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Ecological risk assessment of mixtures of radiological and chemical stressors: Methodology to implement an msPAF approach☆

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ABSTRACT

A main challenge in ecological risk assessment is to account for the impact of multiple stressors. Nuclear facilities can release both radiological and chemical stressors in the environment. This study is the first to apply species sensitivity distribution (SSD) combined with mixture models (concentration addition (CA) and independent action (IA)) to derive an integrated proxy of the ecological impact of combined radiological and chemical stressors: msPAF (multisubstance potentially affected fraction of species). The approach was tested on the routine liquid effluents from nuclear power plants that contain both radioactive and stable chemicals. The SSD of ionising radiation was significantly flatter than the SSD of 8 stable chemicals (namely Cr, Cu, Ni, Pb, Zn, B, chlorides and sulphates). This difference in shape had strong implications for the selection of the appropriate mixture model: contrarily to the general expectations the IA model gave more conservative (higher msPAF) results than the CA model. The msPAF approach was further used to rank the relative potential impact of radiological versus chemical stressors.
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1. Introduction

The impact of multiple stressors on ecosystems is a challenging research area. While nuclear facilities release both radiological and chemical stressors to the environment under normal operating conditions, ecological risk assessment procedures still focus on the separate risks engendered by single types of stressors. It is widely recognised that (i) the effect of a mixture of substances or of stressors is different from the simple summation of the individual effects of each of its components, and (ii) that a mixture can exert a significant toxicity to organisms even if all of its components are present at concentrations below the thresholds that individually do not provoke any toxicity (i.e. NOECs: No observed effect concentration) (Kortenkamp et al., 2009; Backhaus et al., 2010). In a recent extended inter-laboratory experiment on various species, Carvalho et al. (2014) demonstrated that regulatory safety concentrations may not provide sufficient protection when chemicals occur in mixture. Posthuma et al. (2016) further demonstrated that chemical mixtures significantly impacted aquatic communities in several field-based studies. It is thus crucial to develop ecological risk assessment tools accounting for the impact of mixture of stressors.

Two classical mixture models proved their worth predicting the effect of chemical mixtures: concentration addition (CA (Löewe and Muischnek, 1926)) and independent action (IA (Bliss, 1939)). In a recent review, Vanhoudt et al. (2012) have concluded that no conceptual limitation prevents the use of these two general concepts to model the effects of mixture of radioactive and stable substances and address the challenging issue of assessing the environmental impact of mixtures that include radioactive substances. CA and IA both formulate the assumption of additivity or non-interaction (i.e. the components of the chemical mixture act without diminishing or enhancing each other's toxicity). Although this assumption has been challenged both theoretically and empirically (e.g. for mixture of metals (Vijver et al., 2011) and mixture of radionuclides and metals (Margerit et al., 2015)), recent reviews concluded that synergistic effects (i.e. positive interaction between substances leading to a higher mixture effect than predicted by the additivity models) were rare at environmentally relevant concentrations (Cedergreen, 2014; Kortenkamp et al., 2007). Additivity models are thus not expected to severely underestimate the effect of chemical mixtures.

The CA model has been repeatedly recommended as a reference model for risk assessment purposes (Cedergreen, 2014; Backhaus

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et al., 2010). The rationale is that CA generally provides more conservative predictions than IA (i.e. CA predicts higher mixture effects than IA). The CA model assumes that the substances in the mixture have a similar mode of toxic action. This assumption is difficult to verify because for a number of chemicals the toxic mode of action is unknown and varies according to the species, the life stage, or even from one organ to another within the same organism (Syberg et al., 2009; Kortenkamp et al., 2009; Margerit et al., 2016). Thus, in the case of complex mixtures such as the releases of nuclear facilities, it is not clear whether the CA model is the best option. The IA model assumes that all the compounds in the mixture have different toxic modes of action. Cedergreen et al. (2008) showed that both CA and IA models are equivalent in terms of prediction accuracy, thus the choice of the mixture model to apply is not straightforward. Both models can be used to provide a prediction window in which the 'true' effect of a mixture can be expected to fall (Backhaus et al., 2010; Altenburger et al., 2013). Other approaches also rely on combination of CA and IA models in which CA is applied on groups of substances sharing a common mode of toxic action and IA is applied across groups of substances with distinct modes of toxic action (e.g. de Zwart and Posthuma, 2005; Smetanová et al., 2014).

The CA and IA mixture models can be combined with species sensitivity distributions (SSD) models to provide a single indicator of ecological risk associated with mixtures: the multisubstance potentially affected fraction of species (msPAF) (de Zwart and Posthuma, 2005). SSDs are widely used to account for the variation in species' sensitivity when deriving maximum acceptable concentrations of a substance (prospective risk assessment) and in retrospective risk assessment approaches (Jesenska et al., 2013). Based on ecotoxicological data (NOEC, EC10 or EC50 (Effective concentration affecting 10 or 50 % of the test population)) for different species and different taxonomic groups, a SSD model expresses the proportion of species potentially affected by the exposure concentration of a given chemical (PAF). In a recent review, Posthuma et al. (2016) showed the interest of multisubstance SSD as a useful lower-tier risk assessment model providing a good proxy (msPAF) of the potential ecological impact of environmental stressors. The disadvantages of SSD modelling reviewed by Forbes and Calow (2002) notably include a lack of ecological realism (lack of data for representative species, ecological interactions not taken into account). Posthuma and de Zwart (2012) proposed to view msPAF as a metric of the toxic pressure characterising a water sample, that does not predict directly the ecological impact, but whose value can be related to the ecological impact. Indeed, several studies have shown significant relationships between msPAF and observed taxon abundance (Posthuma et al., 2016; Posthuma and de Zwart, 2012) or functional diversity (Smetanová et al., 2014; Jesenska et al., 2013). Finally, the msPAF approach is useful to rank substances and/or stressors within mixture according to their relative contribution to the ecological impact (Posthuma et al., 2016).

The msPAF is thus a promising proxy for screening-level ecological risk assessment (ERA) of chemical mixture, but it has never been applied to mixtures that include radionuclides. Until now, only a few attempts have considered radiological and chemical stressors altogether in ERA. Garnier-Laplace et al. (2009) provided a screening level ERA of the liquid effluents from nuclear power plants that integrated radioactive and stable compounds based on ecotoxicological data. In that study, SSD curves were simplified as linear relationships inspired from a life cycle analysis (LCA) approach (Pennington et al., 2004). That previous study was a first attempt to integrate radiological and chemical stressors in an ERA approach that needs to be refined, notably by including the recent advances in the field. Garnier-Laplace et al. (2009) applied CA but not IA model and concluded that the impact of radionuclides was 5 orders of magnitude lower than the impact of stable chemical substances based on a linear approach. van de Meent and Huijbregts (2005) recommended using the non-linear msPAF approach to estimate effect factors in LCA. Indeed, the msPAF approach best reflects the distribution of ecotoxicological responses, and it further offers the possibility to aggregate other stressors (such as eutrophication or warming) meaningfully (van de Meent and Huijbregts, 2005; van Zelm et al., 2007). Integrating the non-linear patterns of the distribution of species sensitivity could thus (i) affect the ranking of the potential ecological impact of radiological and chemical stressors previously derived, and (ii) provide a ranking that takes advantage of all the information available within ecotoxicological data.

The present study tests and discusses the msPAF approach on the routine releases from nuclear facilities, with the aim to contribute to the improvement of existing ERA tools and framework by explicitly addressing the effect of mixture of radiological and chemical stressors. The msPAF approach was applied for the first time to the case of the liquid effluents released under normal operating conditions in a large river by four nuclear power plants located in the watershed. The two mixture models CA and IA and their combination (i.e. CA applied on separate groups of substances and IA across groups) were compared for different exposure scenarios (dilutions of the liquid effluents in the river). The msPAF approach was further used to rank two categories of stressors within the liquid effluents according to their potential ecological impact: ionising radiation and stable chemicals. The ranking results were compared with the ranking based on the linear approach carried out by Garnier-Laplace et al. (2009).

2. Material and methods

The general methodology of this study is illustrated in a flow diagram in Fig. 1.

2.1. Exposure scenarios

2.1.1. Characteristics of the liquid effluents

This study focused on the liquid effluents from four French nuclear power plants (Bugey (P1), Saint Alban (P2), Cruas (P3) and Tricastin (P4), ordered from upstream to downstream) under normal operating conditions into the Rhône river in 2013. The chemical and radiological composition of the effluents were retrieved from the EDF environment report (EDF, 2013). Table 1 shows the annual released quantities of the 22 stable chemicals and 13 radionuclides. The effluents of these four plants covered a range of different mixture compositions. For example the P3 plant released more stable chemicals than the three other plants (Table 1).

The range of exposure scenarios was further extended using three different dilution scenarios of the effluents into the river: (i) at the mean flow rate of the Rhône near the nuclear plant (465 m^3 s^{-1} for P1, 590 $m^3.s^{-1}$ for P2, 1480 $m^3.s^{-1}$ for P3 and 1167 $m^3.s^{-1}$ for P4 (in the Donzère-Mondragon canal) (Banque Hydro, 2016)), (ii) at the minimum flow rate above which the national nuclear safety authority (ASN) authorises to discharge effluents into the river (130, 255, 300 and 400 $m^3.s^{-1}$ respectively for the plants P1 (ASN, 2013), P2 (ASN, 2014b), P3 (ASN, 2014a) and P4 (ASN, 2008) and (iii) the pure effluents, considered here as a worst-case scenario to test the model predictions at maximal (unrealistic) exposure levels. The concentrations of the different chemicals and radionuclides were obtained considering a simple dilution model in which the releases were averaged by year (Garnier-Laplace et al., 2009):

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