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A single indicator of noxiousness for people and ecosystems exposed to stable and radioactive substances

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Abstract

Inspired by methods used for life cycle impact assessment (LCIA), we constructed a series of indicators to appreciate the noxiousness of radioactive materials and wastes for human and ecosystem health. According to known potential human health and ecological effects of such materials, six main impact categories were considered to initiate the development of the method: human cancer and non-cancer effects vs. ecotoxicity, considering both chemotoxicity and radiotoxicity. For ecosystems, the noxiousness indicator is based on the concept of Potentially Affected Fraction (PAF), used as a damage indicator at the ecosystem level. The PAF express the toxic pressure on the environment due to one substance. It has been enlarged to mixtures of substances as multi-substances PAF (ms-PAF), and applied to a mix of stable and radioactive substances. Combining ecotoxicity data and a simplified model of exposure of fauna and

flora, we proposed a chemotoxicity indicator and a radiotoxicity indicator, ultimately aggregated into a single indicator simply by addition.

According to acknowledged practices in LCIA and corresponding available data, we suggested implementing to human health an approach similar to that applied to ecosystems. We produced eight basic indicators combining effects categories (cancer and non cancer), exposure pathways (ingestion and inhalation) and substances (chemicals and radionuclides). The principle of additivity supporting the whole proposed approach allows their complete aggregation into a single indicator also for human health. Different source terms may be then easily directly compared in terms of human and ecological noxiousness.

Applied to the time evolution of a High Level radioactive Waste (HLW), the method confirmed over 1 million years the dominance of the radiotoxicity in the noxiousness of the material for both humans and environment. However there is a change with time in the ranking of the most noxious substances, with stable metals contribution going progressively up. Finally, the HLW global noxiousness, integrating human health and ecological aspects, was assessed through time at three stages and showed a temporal decrease as expected from the dominance of the radiotoxicity.

Capsule: The ecological noxiousness of chemicals and radionuclides can be expressed by a single indicator, potentially transposable to human health

Keywords: noxiousness, integration, human health, ecosystem, chemotoxicity, radiotoxicity

1. INTRODUCTION

It is today consensually acknowledged that stressors occur usually in mixture in the environment, and may impact simultaneously human and ecological health. This reality generates for any living organism complex and uncertain exposure situations for which potential health and ecological consequences are difficult to assess. Most often, the risk assessment for human health is conducted independently, as well as the ecological risk assessment, ignoring other risks (psycho-social, politic, etc.). The awareness of this situation by the general public result in a growing demand for risk integration that should be reflected in regulatory developments. The notions underlying risk assessments are changing to integrate this new dimension. This can only be done progressively, as it supposes to concily concepts and methods specific to each domain involved as for example within the TRIAD approach, the procedure recommended by the ISO for site-specific ecological risk assessment of soil contamination (ISO, 2015).

At present, it is possible to deal operationally with the complex question of the assessment of a global impact of pollutant mixtures. Effect models exist that would make it possible with some improvements to grasp risks associated with the releases of complex mixtures and subsequent exposure of plural targets in the environment. The approach we propose here should be seen as a proof of concept for a framework to characterize complex source terms for the environment through a unique noxiousness indicator. This indicator aims to integrate in an harmonized way human health and ecological aspects for two categories of toxicity, chemotoxicity and radiotoxicity. The final objective is to obtain a unique operational indicator of the noxiousness of these

complex source terms that would be a first-rate help for the management and communication of the covered risks.

The Life Cycle Impact Assessment (LCIA) approach provides a source of inspiration with regard to integration in terms of risks, and more specifically of ecological risks in freshwaters (ISO 2006a, 2006b). The corresponding indicator of ecosystem damages is the Potentially Affected Fraction (PAF) of species in the ecosystem, mostly obtained from a Species Sensitivity Distribution (SSD) (Pennington *et al*, 2004). The ms-PAF was developed as the next step to express the ecotoxic pressure of mixtures of substances (De Haes *et al*, 2002; Van de Meent and Huijbregts, 2005; Solomon *et al*, 2008). Based on a common toxic unit, this concept opens the door to ambitious integrative approaches. We took a first step in that direction in 2007 (Garnier-Laplace *et al*, 2009), looking for the assesment as a whole of the radioactive and chemical risks for fauna and flora exposed to liquid releases from nuclear facilities. We have recently deepened the principles of this approach (Beaumelle *et al*, 2017), with an investigation of the combined use of SSDs and additivity models. On the strength of these developments, we propose an operational method that produces additive indicators for both chemotoxicity and radiotoxicity, applied to human and ecological health. Our objective was to demonstrate the feasibility of an integrated single indicator for the global noxiousness of different stressors to which people and wildlife can be simultaneously exposed. We present first the construction of this indicator. We defined a toxicity factor F_{tox} as a simplification of the Effect Factor used in Life Cycle Impact Assessment, which relies for a given substance on its hazardous concentration for 50% of the species. This F_{tox} parameter is then combined with an information on the substance quantity to obtain a basic Noxiousness Indicator

(NI). This has to be done without prior selection for each substance, each exposure pathway, each toxicity endpoint. Basic NIs can be summed in different ways to produce intermediate indicators, relevant for a category of toxicity, an exposure pathway or a toxicity endpoint. A second level of integration is proposed to obtain the final indicator, the global noxiousness indicator. In a second part we applied our approach to a High Level radioactive Waste, a mixture of radionuclides and stable metals ideal to test our indicator. The selection of substances was then done according to the availability of the data required by the indicator calculation, our guideline being to privilege recognized and homogeneous sources of data.

The framework presented in this paper to address the risk assessment of mixtures of stable and radioactive substances for both man and the environment was conceived to meet an operational need for comparison between complex source terms, i.e. in a purely comparative perspective between related sources of toxicity. This main goal allowed simplification in the processes taken into account; it required robust but overconservative assumptions in order to achieve the integration which are not compatible with a realistic risk assessment, but mainly answer the need for comparison. To enhance realism in the selected scenarios, our method as presented here should be seen as a starting point to develop a more consistent and homogeneous approach, within a collaborative process associating experts from each domain.

2. MATERIAL AND METHODS

The method described hereafter was initially developed for ecosystems. Its extension to human health was logically considered in a second time but is less accomplished. We

considered its achievement will require the collaboration of experts and specialists in the field of human toxicity. We decided to briefly present what we thought to be the required associated concepts and basics to invite beyond this feasibility exercise the most suitable audience to contribute to its improvement.

The detailed mathematical formulations of the factors and indicators presented below, as well as the fully referenced data sources, are provided as supplemental data.

2.1 Elementary bricks: Fate and Effect Factors

Most of the method was described in a previous work (Garnier-Laplace *et al*, 2009). Directly inspired from the LCIA methodology, it combines a fate-analysis step and an effect-analysis step respectively described by a fate factor (FF) and an effect factor (EF) (Pennington *et al*, 2006). These factors allow to express for each substance one-by-one the change in exposure from a given release and the change in effect per unit change of exposure. In summary, FF is defined per substance as the equilibrium ratio between the concentration within the receiving compartment and the concentration in the donor. EF is defined as the ratio $\Delta\text{PAF}_i/\Delta\text{C}_i$, also equal to the ratio $0.5/\text{HC}_{50i}$, where HC_{50i} is the Hazardous Concentration of the substance i affecting 50% of species at their 50% effect, as recommended to compare impact categories (Pennington *et al*, 2006). The variation in ΔPAF is expressed as $0.5\Sigma_i(\Delta\text{C}_i/\text{HC}_{50i})$. Theories, assumptions and options related to the ΔPAF , EF, their relationship and their use in a multi-stressor context have been presented and discussed elsewhere (e.g. Van de Meent *et al*, 2005 ; Garnier-Laplace *et al*, 2009). The adaptation of the method to radionuclides needs to convert radiotoxicity data expressed in $\mu\text{Gy/h}$ into an HC_{50} expressed in mol/L , as for stable substances. The conversion process described by Garnier-Laplace *et al* (2009) is replicated.

2.2. *The toxicity factor Ftox*

An additional step is taken toward the operational aspect of the method, by further simplifying the EF expression. Focusing on comparisons, any proportionality factor independent of the substance is useless and therefore removed. As such, we define a new EF for ecosystems, called Toxicity Factor (Ftox) and simply equal to $1/HC_{50i}$.

Recognized practices in LCIA supported by available data prompt us to suggest the extension of the approach to human health, considering ingestion and inhalation exposures. The corresponding Ftox takes the form of the inverse of the $ED50_{i,e,v}^{whole\ life}$. This is the life dose inducing a 50% increase of the probability to develop the pathology e for individuals exposed to the substance i via the pathway v (kg internalized/ whole life). This concept is shared between human radiotoxicity and chemotoxicity and should be easily expressed in a common unit. But its use will require some care. For example, the duration of what is similarly called “whole life”, a shared notion used in the derivation of ED_{50} values, differs slightly between chemotoxicity (70 years) and radiotoxicity (86 years). The issue of how comparable the various types of toxicity benchmarks (i.e. for radioactive or stable substance) are, must also be solved. Some homogenization work would also be probably necessary to obtain consistent datasets regardless the exposure pathway (ingestion, inhalation) and the pathology (cancer, non-cancer).

2.3. *The Noxiousness Indicator NI*

The notion of Ftox lacks the quantitative dimension attached to the source term, which introduction leads to the definition of the Noxiousness Indicator (NI). NI corresponds, for a given substance s , to the product of its Ftox by its concentration C_i in the source term

under consideration i.e. the ratio of the substance concentration for example in water to its HC_{50} . This dimensionless indicator can be summed for ecosystem on substances to give two basic indicators, either the radiological ($NI-E_{rad}$) or chemical ($NI-E_{chem}$) ecological noxiousness indicator. Summation over toxicity categories produces the final global indicator NI-E (Table 1). These three indicators allow dealing with two of the six impact categories selected (chemo- and radio-ecotoxicity).

The remaining four impact categories (human cancer and non-cancer effects due to chemotoxicity or radiotoxicity), remain to treat consistently. The previous approach has then been applied to human health in a similar however a bit more complex way. In addition to the category of effects (cancer; non-cancer) and type of toxicity (radiological; chemical) the noxiousness has to be characterised also with regard to exposures pathways. Indeed the ED_{50} values for people required by the method are usually reported for a given exposure pathway. A look in some relevant databases showed that data are easily available mainly for ingestion or inhalation. We acknowledged that other exposure pathways such as irradiation at distance or dermal adsorption could be of importance. However we considered that an objective of feasibility demonstration as pursued here could be satisfied by the immediate availability of ingestion and inhalation data. Moreover reducing the application to these two exposures pathways allowed a certain homogeneity in the data required for human and ecosystem health by using a same internationally recognised source. That means finally that eight basic indicators with the same unit have to be produced to cover all the possible combinations (Table 1). These indicators might be aggregated to express the noxiousness in an integrated manner by summing some or all the individual indicators (all substances per effect category, all

effects categories ...) or even the global noxiousness. Basically, each indicator NI-
 $hh_{\text{toxicity, effect, pathway}}$ is calculated for each substance, stable or radioactive. They are then
summed for all substances in a toxicity category (chemo- or radiotoxicity) for a given
exposure pathway and a given effect category to produce the eight basic indicators (i.e.
NI- $hh_{\text{chem, c, ing}}$ indicator related to the cancer effect linked to the ingestion of all the
chemical substances). The first level of integration produces by summing over effects
(cancer and non-cancer) four intermediate indicators that express the noxiousness related
to an exposure pathway for a given category of toxicity (i.e. NI- $hh_{\text{chem, ing}}$ indicator related
to the toxicity by ingestion of all chemicals), and by summing over exposure pathways
four other indicators that express the noxiousness related to an effect category for the
same category of toxicity (i.e. NI- $hh_{\text{chem, c}}$ indicator related to cancer due to all chemicals).
The second level of integration produces two indicators, one per category of toxicity (i.e.
NI- hh_{chem} the indicator related to chemotoxicity including all effects and pathways). The
third and last level of integration gives the final single expected indicator by summing the
two previous ones.

The difficulty relies on ensuring the homogeneity of significance of all these indicators
and expressing them in a same unit.

Table 1. Indicators proposed to characterize the noxiousness of complex source terms for
human health (hh) and ecosystems (E), depending on the level of aggregation

Toxicity	Exposure pathway	Human health			Ecosystem
		Cancer (c)	Non-cancer (nc)	Both	All effects
Chemotoxicity (chem)	Ingestion (ing)	NI- $hh_{\text{chem, c, ing}}$	NI- $hh_{\text{chem, nc, ing}}$	NI- $hh_{\text{chem, ing}}$ ^a	n.a.
	Inhalation (inh)	NI- $hh_{\text{chem, c, inh}}$	NI- $hh_{\text{chem, nc, inh}}$	NI- $hh_{\text{chem, inh}}$ ^a	n.a.
	All pathways	NI- $hh_{\text{chem, c}}$ ^a	NI- $hh_{\text{chem, nc}}$	NI- hh_{chem} ^b	NI- E_{chem} ^b

Radiotoxicity (rad)	Ingestion (ing)	NI-hh _{rad,c,ing}	NI-hh _{rad,nc,ing}	NI-hh _{rad,ing} ^a	n.a.
	Inhalation (inh)	NI-hh _{rad,c,inh}	NI-hh _{rad,nc,inh}	NI-hh _{rad,inh} ^a	n.a.
	All pathway ^a	NI-hh _{rad,c} ^a	NI-hh _{rad,nc} ^a	NI-hh _{rad} ^b	NI-E _{rad} ^b
All toxicities	All pathways	NI-hh _c ^b	NI-hh _{nc} ^b	NI-hh ^c	NI-E ^c

^a first level of aggregation (on effects or on pathways)

^b second level of aggregation (on a category of toxicity)

^cthird level of aggregation (all inclusive)

The proposed method does not aim to assess risk realistically, but rather to offer a comparison tool able to put on a same scale different impact categories initially not comparable. It relies on oversimplification of environmental transfer processes, exposure and effect characterizations (for example, use of HC₅₀ rather than HC₁₀ as usually in ERA). The numerical values of the calculated indicators have no meaning in themselves, they do not indicate a level of detriment or damage. They are only the product of a standardised rigorous approach applicable to any set of substances in a perfectly similar way, authorizing comparisons of noxiousness of various inventories of multiple substances (or source-term) or for a given inventory of multiple substances, the time evolution of the indicators.

2.4 Data sources

Toxicity data were collected from acknowledged sources for both human and ecosystems, ensuring consistence by using as long as possible common supports. For chemicals, the databases associated with the version 2.0 of the tool USETOX were then consulted for humans (ED₅₀; Fantke *et al*, 2017) and ecosystems (EC₅₀; Hugonnet *et al*, 2014). For radionuclides, the FREDERICA database (Coppelstone *et al*, 2008) and the derived dose-effect relationships (Garnier-Laplace *et al*, 2010) underpinned the reasoning for

ecosystems. For humans, data were retrieved from the UNSCEAR report (UNSCEAR, 1982) and the paper from Edwards and Lloyd (1998).

Data regarding the parameterisation of transfer in the environment were found in the ERICA Tool (Brown *et al*, 2008) specifically for wildlife and in some IAEA reports (IAEA, 2001, 2010) for humans. Additional information and details are provided as supplemental data.

3. APPLICATION AND DISCUSSION

The proposed approach was applied to a well characterised source-term (High Level radioactive Waste - HLW), including stable and radioactive substances, and its evolutionary composition at the short-, medium- and long-term (0, 10^3 and 10^4 years). Due to radioactive decay, the radiological toxicity of radionuclides initially present decreases with time, by generating new substances that may themselves be radiologically or chemically toxic.

According to Table 1, basic, intermediate and final noxiousness indicators were calculated both for human health and ecosystems. The scarcity in (eco)toxicity data is a well-known weakness of risk assessment methods since it concerns the vast majority of chemical substances with the exception mainly of some metals (*e.g.* Pb, Hg, Ni). Due to the anticipated lack of data, especially for stable substances, the application focused on the need for demonstration on the basis of freshwater ecosystems for which the greatest number of substances are characterised in terms of toxicity. For the human health aspect, the already identified limitations were not considered at this stage of method development, but will have to be solved for an operational application.

3.1. *Intrinsic vs. In Situ Noxiousness*

The objective was to evaluate comparatively the global noxiousness of the time dependent compositions of the HLW (each of the three was considered as a different waste for the study) . We defined an intrinsic noxiousness, which is a property of a given composition of the waste itself (inner waste toxicity), apart from any consideration of dispersion in the environment. We defined additionally the *in situ* noxiousness, to integrate the filter effect of environmental dispersion. This was done at first by introducing in the calculation the liquid-solid partition coefficient. Both components of noxiousness allow a more complete characterisation of the waste under investigation.

3.2. *Use of Noxiousness Indicators in isolation*

3.2.1. *Ecological indicators*

The ecological noxiousness indicators were calculated for any substance which ecotoxicity was sufficiently documented. The resulting total intrinsic ecological NI decreases by about two orders of magnitude over a million years (Figure 1). Radiotoxicity is the major contributor over the full time period, the total chemotoxicity being less by two to four orders of magnitude. Taking into account the chemotoxicity of radionuclides and their stable decay products does not significantly alter the total chemotoxicity, almost entirely due to stable substances initially present. The contribution of stable substances generated by radioactive decay is not quantitatively significant. It significantly increases over time, by two orders of magnitude over the studied period. To illustrate the additional information possibly retrieved from such an approach, we focused on the hierarchization of toxics, which may support management decision. We

limited arbitrarily the toxic classification to the top ten substances in each category, radiotoxic and chemotoxic substances. They have been prioritized with regard to their contribution to the intrinsic ecological noxiousness of HLW from their noxiousness indicators as a function of time. Initially from far the most harmful for wildlife (Figure 2A), radionuclides lose ranking places over time and their order or even their nature is modified (e.g. ^{243}Am rises from ninth to second place, disappearance of ^{137}Cs), due to radioactive decay. After 1000 years, four metals (Al, Ni, Zn and Cr) integrate the first half of the ranking (Figure 2B), without change in their ranking. After 10 000 years, the top ten places are equitably occupied by radioactive (always the most harmful) and stable substances (Figure 2C), with order modification only for radionuclides. Intrinsic noxiousness indicators range almost over two orders of magnitude, while they varied initially over five orders of magnitude.

3.2.2. Human health indicators

When looking for completeness to calculate ecological noxiousness indicators, the determination of human health indicators focused on a reduced number of substances. The objective was clearly to demonstrate feasibility of an approach that would produce consistent noxiousness indicators for humans and wildlife integrating chemo- and radiotoxicity. The substances were selected among those for which both types of indicators could be calculated.

Within the limit of the selected substances and under the assumptions made, the total intrinsic chemical noxiousness of HLW does not evolve for human health over the studied period of time (Figure 3). This indicator is mainly driven by cancer effects linked to ingestion of substances, (inhalation is of minor contribution - results not shown). This

noxiousness, initially up to four orders of magnitude lower than that of radiological harm, remains lower for about 10,000 years. Effects mainly responsible for radiotoxicity would be non-cancer effects, but differences between the two types of effect are much less marked than for chemotoxicity (results not shown). As expected, results in terms of *in situ* noxiousness would be similar, with a shift toward lowest values of indicators due to the self-time filter effect. These observations have no absolute value; they depend closely on the substances taken into consideration. This exercise has been done for illustrative purposes only.

3.3. Use of Noxiousness Indicators in combination

Indicators were calculated for human health and ecosystems for the same source term according to a harmonized approach, making them consistent. The next step would logically to combine them to express the total noxiousness of this source term and ultimately to compare several such terms. Multiple combinations are possible. The simplicity of reading and interpretation guided the choice presented here. Both indicators are reported on a single graph to allow categorization and immediate comparison of the source terms. Each source-term is positioned according to its combination of indicators: the human health indicator is plotted on the x-axis, the ecological indicator on the y-axis, even though their values are not necessarily bound. Classes of noxiousness are defined as the bands delimited by two successive pairs of identical values on the x and y axes (*i.e.* band defined by $x_1=y_1=a$ and $x_2=y_2=a+1$ increment). Two source terms in the same class (*i.e.* same band) have a comparable total intrinsic noxiousness. Two source terms in different bands have different noxiousness the one in the right-most band being considered more harmful. This representation offers the advantage of a great readability,

but supposes the implicit assumption of the equivalence between noxiousness for humans and ecosystems for the same value of their respective indicators. This is questionable and should be considered on a case-by-case basis, depending on the objective and context of the noxiousness characterization. This issue could be solved by weighting one or the other of the indicators. Then the question arises as to which weight values to apply...

Indicators were retrieved from the sets of results acquired for substances common to both previous steps. We defined 14 classes of noxiousness covering the range of values of total intrinsic noxiousness indicators for humans and ecosystems (**Figure 4**).

The decrease over time of the total intrinsic noxiousness indicators for the selection of stable and radioactive substances from the HLW, already discussed, moves the waste noxiousness from category XIII in the short term to category XI then VIII in the medium and long terms respectively.

4. CONCLUSIONS

The proposed method was applied to the time changing composition of a High Level radioactive Waste. Such a material includes both radionuclides and toxic metals, but is today regulated only with regard to its radiotoxicity. Within the acknowledged limitations of the approach, the global Noxiousness Indicator we calculated confirmed as expected the dominance of this radiotoxicity through time over a period of 1 million years, for both people and the environment. The use in combination of human health and ecological indicators allowed to obtain a clear and synthetic view of the temporal evolution of the global noxiousness of the waste, which could be a powerful communication and decision support. Lower levels of integration are also rich in insights, notably in terms of

comparison between substances. This method is however largely perfectible, for example due to some “dead ends”, data gaps, over simplification or huge uncertainties.

The concept of a single indicator of noxiousness for both human health and ecosystems, explicitly embedding radio- and chemotoxicity, is attractive and the related approach we presented appears promising. It illustrates the feasibility of full integration into a consistent approach related to human and ecosystem health. The reverse of the method is for chemicals at worst the well known lack of toxicity data, at best their heterogeneity. Effect endpoints, species, and other characteristics of lab tests devoted to EC_x determination are extremely variable. Methods to obtain metadata from these basic information are no longer homogeneous. The next step in the development of such indicators would necessarily require significant work to homogenize all the underlying data. Additional consideration should be paid to bioavailability that conditions the substance toxicity. The treatment of uncertainties has to be added. Solving these aspects will produce truly consistent indicators, which use in combination will need an extra step of normalization and weighing. This finalization requires the involvement of all stakeholders, especially when combining human and environmental health indicators.

5. SUPPLEMENTAL DATA

The Supplemental Data are available on the Wiley Online Library at DOI:10.1002/etc.xxx.

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405

406 **FIGURE CAPTIONS**

407 Fig.1 temporal evolution of indicators of intrinsic ecological noxiousness of HLW,
408 depending on the stable or radioactive nature of the substances and their toxicity

409 Fig.2 temporal evolution of the ranking of the ten radionuclides and stable substances
410 within HLW the most inherently harmful to wildlife as a function of time

411 Fig.3 temporal evolution of indicators of intrinsic and *in situ* noxiousness for human
412 health of a selection of substances within HLW, depending on their toxicity

413 Fig.4 positioning of the selection of stable and radioactive substances from the HLW at
414 short, medium and long term in the abacus of total intrinsic noxiousness

415 Figure 1

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418 Figure 2

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422 Figure 3

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425 Figure 4

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427