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

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9

10 **Abstract**

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

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

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

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

39

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

42

43 **Keywords:** noxiousness, integration, human health, ecosystem, chemotoxicity,
44 radiotoxicity

45 **1. INTRODUCTION**

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

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

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

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

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

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

109 **2. MATERIAL AND METHODS**

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

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

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

118 *2.1 Elementary bricks: Fate and Effect Factors*

119 Most of the method was described in a previous work (Garnier-Laplace *et al*, 2009).
120 Directly inspired from the LCIA methodology, it combines a fate-analysis step and an
121 effect-analysis step respectively described by a fate factor (FF) and an effect factor (EF)
122 (Pennington *et al*, 2006). These factors allow to express for each substance one-by-one
123 the change in exposure from a given release and the change in effect per unit change of
124 exposure. In summary, FF is defined per substance as the equilibrium ratio between the
125 concentration within the receiving compartment and the concentration in the donor. EF is
126 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
127 Hazardous Concentration of the substance i affecting 50% of species at their 50% effect,
128 as recommended to compare impact categories (Pennington *et al*, 2006). The variation in
129 ΔPAF is expressed as $0.5\sum_i(\Delta\text{C}_i/\text{HC}_{50i})$. Theories, assumptions and options related to the
130 ΔPAF , EF, their relationship and their use in a multi-stressor context have been presented
131 and discussed elsewhere (e.g. Van de Meent *et al*, 2005 ; Garnier-Laplace *et al*, 2009).
132 The adaptation of the method to radionuclides needs to convert radiotoxicity data
133 expressed in $\mu\text{Gy/h}$ into an HC_{50} expressed in mol/L, as for stable substances. The
134 conversion process described by Garnier-Laplace *et al* (2009) is replicated.

135 *2.2. The toxicity factor Ftox*

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

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

153 *2.3. The Noxiousness Indicator NI*

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

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

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

180 effects categories ...) or even the global noxiousness. Basically, each indicator NI-
 181 $hh_{\text{toxicity,effect,pathway}}$ is calculated for each substance, stable or radioactive. They are then
 182 summed for all substances in a toxicity category (chemo- or radiotoxicity) for a given
 183 exposure pathway and a given effect category to produce the eight basic indicators (i.e.
 184 $NI-hh_{\text{chem,c,ing}}$ indicator related to the cancer effect linked of the ingestion of all the
 185 chemical substances). The first level of integration produces by summing over effects
 186 (cancer and non-cancer) four intermediate indicators that express the noxiousness related
 187 to an exposure pathway for a given category of toxicity (i.e. $NI-hh_{\text{chem,ing}}$ indicator related
 188 to the toxicity by ingestion of all chemicals), and by summing over exposure pathways
 189 four other indicators that express the noxiousness related to an effect category for the
 190 same category of toxicity (i.e. $NI-hh_{\text{chem,c}}$ indicator related to cancer due to all chemicals).
 191 The second level of integration produces two indicators, one per category of toxicity (i.e.
 192 $NI-hh_{\text{chem}}$ the indicator related to chemotoxicity including all effects and pathways). The
 193 third and last level of integration gives the final single expected indicator by summing the
 194 two previous ones.

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

197 Table 1. Indicators proposed to characterize the noxiousness of complex source terms for
 198 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	$NI-hh_{\text{chem,c}}^a$	$NI-hh_{\text{chem,nc}}$	$NI-hh_{\text{chem}}^b$	$NI-E_{\text{chem}}^b$

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

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

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

201 ^c third level of aggregation (all inclusive)

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

212 2.4 Data sources

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

219 ecosystems. For humans, data were retrieved from the UNSCEAR report (UNSCEAR,
220 1982) and the paper from Edwards and Lloyd (1998).

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

225 **3. APPLICATION AND DISCUSSION**

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

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

241 *3.1. Intrinsic vs. In Situ Noxiousness*

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

250 *3.2. Use of Noxiousness Indicators in isolation*

251 *3.2.1. Ecological indicators*

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

263 limited arbitrarily the toxic classification to the top ten substances in each category,
264 radiotoxic and chemotoxic substances. They have been prioritized with regard to their
265 contribution to the intrinsic ecological noxiousness of HLW from their noxiousness
266 indicators as a function of time. Initially from far the most harmful for wildlife (Figure
267 2A), radionuclides lose ranking places over time and their order or even their nature is
268 modified (e.g. ^{243}Am rises from ninth to second place, disappearance of ^{137}Cs), due to
269 radioactive decay. After 1000 years, four metals (Al, Ni, Zn and Cr) integrate the first
270 half of the ranking (Figure2B), without change in their ranking. After 10 000 years, the
271 top ten places are equitably occupied by radioactive (always the most harmful) and stable
272 substances (Figure2C), with order modification only for radionuclides. Intrinsic
273 noxiousness indicators range almost over two orders of magnitude, while they varied
274 initially over five orders of magnitude.

275 3.2.2. *Human health indicators*

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

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

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

294 *3.3. Use of Noxiousness Indicators in combination*

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

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

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

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

321 **4. CONCLUSIONS**

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

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

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

347 **5. SUPPLEMENTAL DATA**

348 The Supplemental Data are available on the Wiley Online Library at

349 DOI:10.1002/etc.xxx.

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405

406 **FIGURE CAPTIONS**

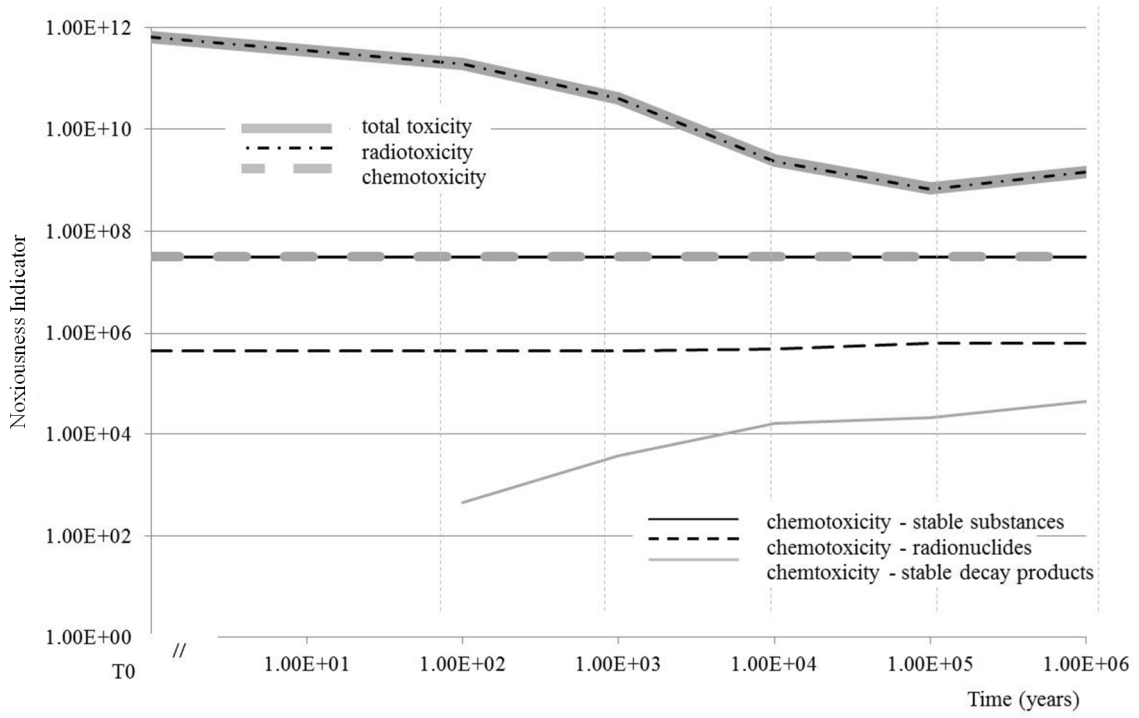
407 Fig.1 temporal evolution of indicators of intrinsic ecological noxiousness of HLW,
408 depending on the stable or radioactive nature of the substances and their toxicity

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

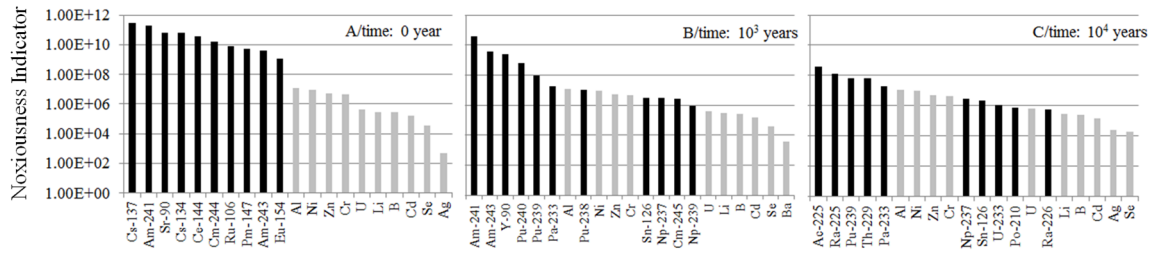
415 Figure 1



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418 Figure 2

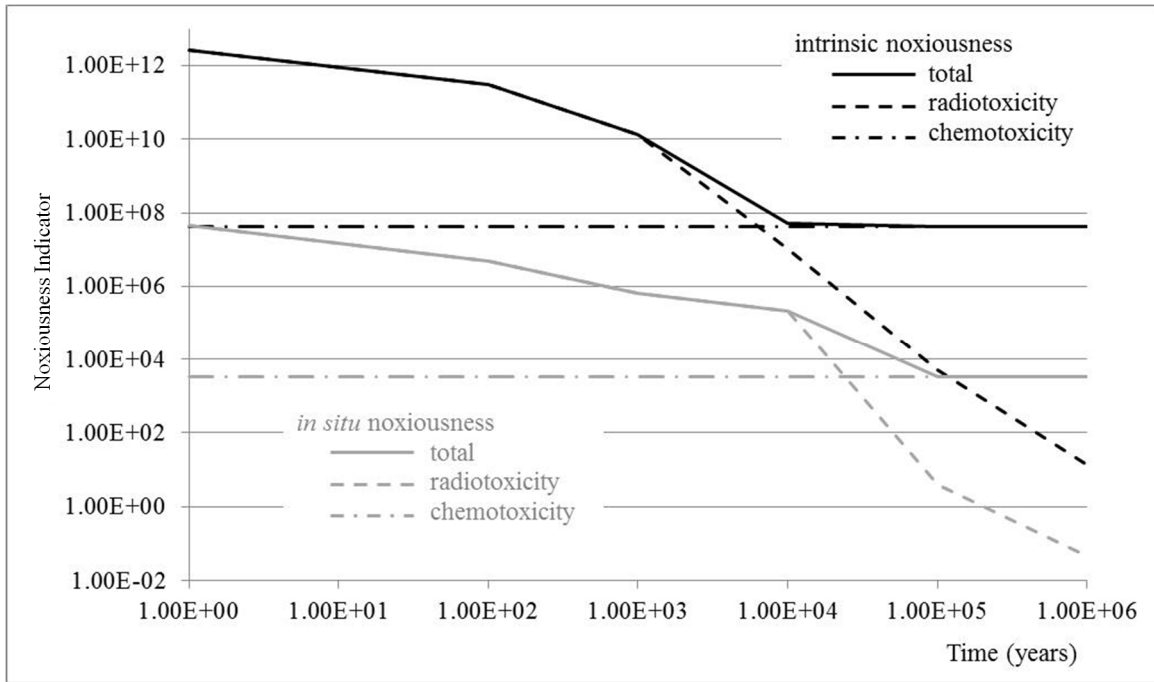
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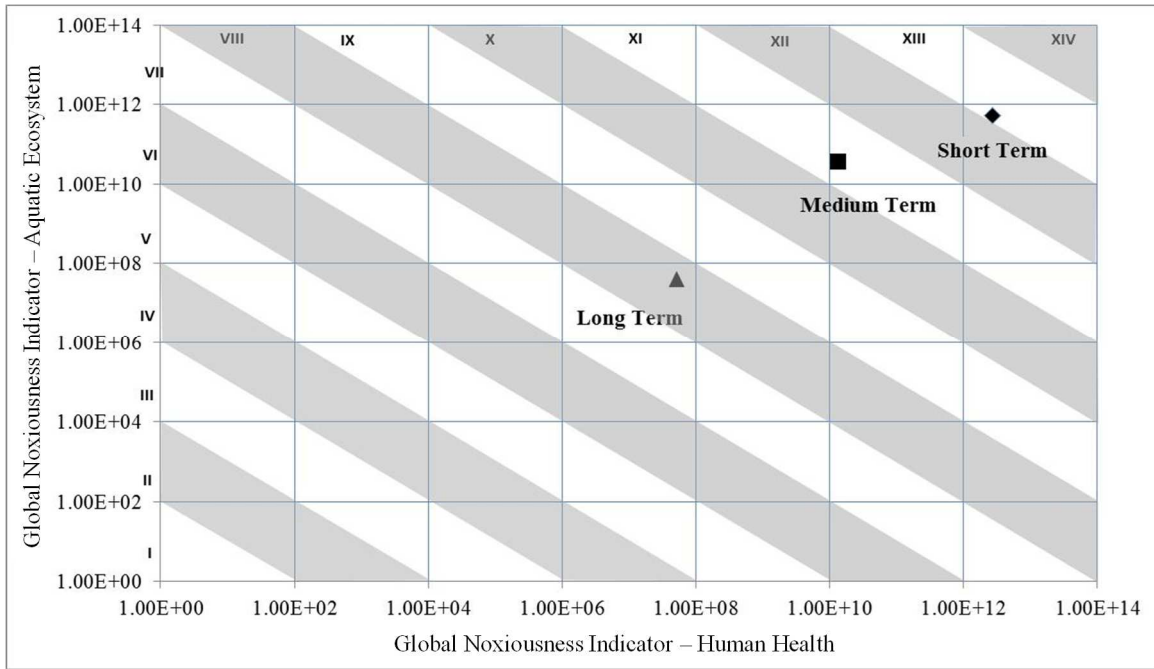
422 Figure 3



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425 Figure 4



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