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

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9

10 Abstract

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

substances. Combining ecotoxicity data and a simplified model of exposure of fauna and

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

24 According to acknowledged practices in LCIA and corresponding available data, we suggested implementing to human health an approach similar to that applied to 25 26 ecosystems. We produced eighb asic indicators combining effects categories (cancer and non cancer), exposure pathways (ingestion and inhlation) and substances (chemicals and 27 radionuclides). The principle of additivity supporting the whole proposed approach 28 29 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 30 noxiousness. 31 Applied to the time evolution of a High Level radioactive Waste (HLW), the method 32 confirmed over 1 million years the dominance of the radiotoxicity in the noxiousness of 33 34 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 35 progressively up. Finally, the HLW global noxiousness, integrating human health and 36 37 ecological aspects, was assessed through time at three stages and showed a temporal decrease as expected from the dominance of the radiotoxicity. 38 39

40 Capsule: The ecological noxiousness of chemicals and radionuclides can be expressed
41 by a single indicator, potentially tranposable 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 environment, and may impact simultaneously human and ecological health. This reality 47 generates for any living organism complex and uncertain exposure situations for which 48 49 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 50 assessment, ignoring other risks (pyscho-social, politic, etc.). The awareness of this 51 52 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 53 changing to integrate this new dimension. This can only be done progressively, as it 54 supposes to concily concepts and methods specific to each domain involved as for 55 example within the TRIAD approach, the procedure recommended by the ISO for site-56 57 specific ecological risk assessment of soil contamination (ISO, 2015). At present, it is possible to deal operationally with the complex question of the 58 assessment of a global impact of pollutant mixtures. Effect models exist that would make 59 60 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 61 approach we propose here should be seen as a proof of concept for a framework to 62 characterize complex source terms for the environment through a unique noxiousness 63 indicator. This indicator aims to integrate in an harmonized way human health and 64 ecological aspects for two categories of toxicity, chemotoxicity and radiotoxicity. The 65 final objective is to obtain a unique operational indicator of the noxiousness of these 66

67 complex source terms that would be a first-rate help for the management and68 communication of the covered risks.

The Life Cycle Impact Asessessment (LCIA) approach provides a source of inspiration 69 70 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 71 72 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 73 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 common toxic unit, this concept opens the door to ambitious integrative approaches. We 76 77 took a first step in that direction in 2007 (Garnier-Laplace et al, 2009), looking for the 78 assessment 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 79 approach (Beaumelle *et al*, 2017), with an investigation of the combined use of SSDs and 80 81 additivity models. On the strength of these developments, we propose an operational method that produces additive indicators for both chemotoxicity and radiotoxicity, 82 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 people and wildlife can be simultaneously exposed. We present first the construction of 85 86 this indicator. We defined a toxicity factor Ftox as a simplification of the Effect Factor used in Life Cycle Impact Assessment, which relies for a given substance on its 87 hazardous concentration for 50% of the species. This Ftox parameter is then combined 88 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 pathway, each toxicity endpoint. Basic NIs can be summed in different ways to produce 91 intermediate indicators, relevant for a category of toxicity, an exposure pathway or a 92 93 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 94 Level radioactive Waste, a mixture of radionuclides and stable metals ideal to test our 95 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.

The framework presented in this paper to address the risk assessment of mixtures of 99 stable and radioactive substances for both man and the environment was conceived to 100 meet an operational need for comparison between complex source terms, i.e. in a purely 101 102 comparative perspective between related sources of toxicity. This main goal allowed simplification in the processes taken into account; it required robust but overconservative 103 104 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 selected scenarios, our method as presented here should be seen as a starting point to 106 develop a more consistent and homogeneous approach, within a collaborative process 107 108 associating experts from each domain.

109 2. MATERIAL AND METHODS

110 The method described hereafter was initially developed for ecosystems. Its extension to111 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 though to be the required 114 associated concepts and basics to invite beyond this feasability exercise the most suitable 115 audience to contribute to its improvement.

The detailed mathematical formulations of the factors and indicators presented below, aswell 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).

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

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 $\Delta PAF_i/\Delta C_i$, also equal to the ratio 0.5/HC_{50i}, where HC_{50i} is the

127 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

129 ΔPAF is expressed as $0.5\Sigma_i(/\Delta C_i/HC_{50i})$. 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 μ Gy/h into an HC₅₀ 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 simplifying the EF expression. Focusing on comparisons, any proportionality factor 137 independent of the substance is useless and therefore removed. As such, we define a new 138 139 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 exposures. The corresponding Ftox takes the form of the inverse of the $ED50_{i,e,v}^{whole life}$. 142 This is the life dose inducing a 50% increase of the probability to develop the pathology e143 144 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 145 146 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 147 148 ED₅₀ 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 149 radioactive or stable substance) are, must also be solved. Some homogenization work 150 151 would also be probably necessary to obtain consistent datasets regardless the exposure pathway (ingestion, inhalation) and the pathology (cancer, non-cancer). 152

153 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 HC₅₀. 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).

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 then been applied to human health in a similar however a bit more complex way. In 165 166 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 167 pathways. Indeed the ED₅₀ values for people required by the method are usually reported 168 169 for a given exposure pathway. A look in some relevant databases showed that data are 170 easily available mainly for ingestion or inhalation. We ackownledged that other exposure 171 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 172 could be satisfied by the immediate availability of ingestion and inhalation data. 173 Moreover reducing the application to these two exposures pathways allowed a certain 174 175 homogeneity in the data required for human and ecosystem health by using a same internationaly recognised source. That means finally that eight basic indicators with the 176 177 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 178 summing some or all the individual indicators (all substances per effect category, all 179

180 effects categories ...) or even the global noxiousness. Basically, each indicator NI-181 hh_{toxicity,effect,pathway} is calculated for each substance, stable or radioactive. They are then summed for all subtances in a toxicity category (chemo- or radiotoxicity) for a given 182 183 exposure pathway and a given effect category to produce the eight basic indicators (i.e. NI-hhchem,c,ing indicator related to the cancer effect linked ot the ingestion of all the 184 185 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 to an exposure pathway for a given category of toxicity (i.e.NI-hh_{chem, ing} indicator related 187 188 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 189 190 same category of toxicity (i.e. NI-hh_{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. NI-hh_{chem} the indicator related to chemotoxicity including all effects and pathways). The 192 third and last level of integration gives the final single expected indicator by summing the 193 194 two previous ones.

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

197 Table 1. Indicators proposed to characterize the noxiousness of complex source terms for198 human health (hh) and ecosystems (E), depending on the level of aggregation

		Human health			Ecosystem
Toxicity	Exposure pathway	Cancer (c)	Non-cancer (nc)	Both	All effects
Chemotoxicity	Ingestion (ing)	NI-hhchem,c,ing	NI-hhchem,nc,ing	NI-hh _{chem,ing} ^a	n.a.
(chem)					

NI-hhchem,c,inh

NI-hh_{chem.c}^a

NI-hh_{chem.nc.inh}

NI-hh_{chem,nc}

NI-hhchem.inh^a

NI-hhchem^b

n.a.

NI-E_{chem}^b

Inhalation (inh)

All pathways

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)

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

201 ^cthird level of aggregation (all inclusive)

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

212 *2.4 Data sources*

Toxicity data were collected from aknowledged 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 (Copplestone *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.

225 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, 10³ and 10⁴ 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 232 calculated both for human health and ecosystems. The scarcity in (eco)toxicity data is a 233 234 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 number of substances are characterised in terms of toxicity. For the human health aspect, 238 the already identified limitations were not considered at this stage of method 239 240 development, but will have to be solved for an operational application.

241 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

- 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*

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

263 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 264 contribution to the intrinsic ecological noxiousness of HLW from their noxiousness 265 266 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 267 modified (e.g. 243 Am rises from ninth to second place, disappareance of 137 Cs), due to 268 radioactive decay. After 1000 years, four metals (Al, Ni, Zn and Cr) integrate the first 269 half of the ranking (Figure2B), without change in their ranking. After 10 000 years, the 270 271 top ten places are equitably occupied by radioactive (always the most harmful) and stable substances (Figure 2C), with order modification only for radionuclides. Intrinsic 272 273 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*

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

286 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 287 be non-cancer effects, but differences between the two types of effect are much less 288 289 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 290 291 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 292 293 purposes only.

294 *3.3. Use of Noxiousness Indicators in combination*

Indicators were calculated for human health and ecosystems for the same source term 295 296 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 297 298 ultimately to compare several such terms. Multiple combinations are possible. The 299 simplicity of reading and interpretation guided the choice presented here. Both indicators are reported on a single graph to allow categorization and immediate comparison of the 300 source terms. Each source-term is positioned according to its combination of indicators: 301 302 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 the bands delimited by two successive pairs of identical values on the x and y axes (i.e. 304 305 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 306 307 different bands have different noxiousness the one in the right-most band being considered more harmful. This representation offers the advantage of a great readibility, 308

309 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 310 should be considered on a case-by-case basis, depending on the objective and context of 311 312 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... 313 Indicators were retrieved from the sets of results acquired for substances common to both 314 315 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

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.

321 4. CONCLUSIONS

The proposed method was applied to the time changing composition of a High Level 322 radioactive Waste. Such a material includes both radionuclides and toxic metals, but is 323 324 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 indicators allowed to obtain a clear and synthetic view of the temporal evolution of the 328 global noxiousness of the waste, which could be a powerful communication and decision 329 support. Lower levels of integration are also rich in insights, notably in terms of 330

331 comparison between substances. This method is however largely perfectible, for example due to some "dead ends", data gaps, over simplification or huge uncertainties. 332 333 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 consistent approach related to human and ecosystem health. The reverse of the method is 336 337 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 indicators would necessarily require significant work to homogenize all the underlying 341 data. Additional consideration should be payed to bioavailability that conditions the 342 343 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 344 of normalization and weigthing. This finalization requires the involvment of all 345 346 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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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



418 Figure 2





422 Figure 3



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