On the combination of USEtox® and SimpleBox 4 Nano models for the derivatization of size-dependent characterization factors for engineered nanomaterials

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Abstract. Even if it has been claimed that Life Cycle Assessment is an essential tool to analyze, evaluate, understand and manage the environmental and health impacts of nanotechnology, few studies incorporate characterization factors (CFs) for human toxicity and freshwater ecotoxicity accounting for the impacts of engineered nanomaterials (ENMs) beyond their manufacturing stage. The objective of the present work consisted in identifying the correspondence between the information required and outputs provided by the USEtox® consensus model (which is not nanospecific) and the SimpleBox4Nano model (which accounts for nanospecific processes, e.g. aggregation, attachment and dissolution for Fate Factor derivatization) in order to assess the possibility of integrating the two to derive size-dependent CFs for the varying sizes of ENMs throughout their life cycle. The possibility to combine and integrate the two models appeared to be limited since there is no absolute correspondence between the two of them.

1 Introduction

Life cycle assessment (LCA) and its corresponding ISO framework [1, 2] are recognized as suitable tools to identify the potential environmental and human health impacts of nano-enabled applications (NEAs) or nano-enabled products (NEPs) along their complete life

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cycles covering manufacturing, use and end-of-life stages [3]. As such, a number of review articles have been published in the past decade that cover the application of LCA to nanotechnology such as the recent work by Salieri et al. [4].

The LCA methodology comprises four iterative steps: (i) goal and scope definition, (ii) inventory analysis, (iii) impact assessment, and (iv) interpretation. Life cycle impact assessment (LCIA) converts emissions into environmental damages through linked fate-exposure-effect models that require robust experimental data and a mechanistic understanding for each of these components. LCIA methods describe environmental impacts in terms of characterization factors (CFs). A CF is a substance-specific quantitative representation of the (relative) impact of a substance in the environment. CFs are based on models of cause-effect chains for a specific impact category.

USEtox® [5, 6] is a fate-effect model specifically made for LCA-applications as it calculates human and eco-toxicity CFs based on the properties of a substance and the environmental compartment of initial release. The model estimates CFs by multiplying three other aggregated parameters related to fate (fate factor, FF), exposure (exposure factor, XF), and toxicity (effect factor EF), respectively, of a specific chemical. USEtox® operates on four different spatial scales: indoor, urban, continental and global. The indoor and urban scales only have an air compartment, whereas the continental and global scales consist of five compartments: air, agricultural soil, natural soil, freshwater and sea water. USEtox® cannot be directly applied to the LCIAs of NEPs and NEAs since the fate modelling is not nanospecific. SimpleBox4Nano (SB4N) [7, 8, 9] is a fate model able to model the fate of engineered nanomaterials (ENMs) depending on their size. SB4N has three main compartments: regional, continental and global, but the inner nested compartment, regional, has not only air as a medium but also freshwater (including lake, lake sediment, freshwater and freshwater sediment), seawater (including surface sea, deep sea and marine sediment) and natural, agricultural soil and urban/industrial soil. From all these media transfers to the other compartments and media are possible. Furthermore, the global compartment is split into three sub-compartments: moderate, arctic and tropical. SB4N models the fate of (i) freely dispersed (pristine) nanoparticle, (ii) nanoparticle hetero-aggregated with natural colloid particles (<450 nm) and (iii) nanoparticle attached to larger natural particles (>450 nm) [9].

Ettrup et al. [10] adapted the USEtox® 2.0 consensus model to integrate the SB4N for the development of CF of TiO_2 nanoparticles to be incorporated in future LCA studies. Also focusing on TiO_2 nanoparticles, Salieri et al. [11] presented an integrative approach for USEtox® 2.0 model with SB4N and proposed CFs for the freshwater toxicity impact category. More recently, Temizel-Sekeryan and Hicks [12] have calculated freshwater ecotoxicity CFs for silver nanoparticles by combining the principles of colloidal science with the USEtox® model using data from published mesocosm conditions.

The objective of the present work was to compare the information requirements and output by the two models in order to identify possible limitations in their integration for the derivation of size-dependent CFs for the varying sizes of ENMs released throughout the life cycle of NEPs and NEAs.

2 Materials and Methods

SimpleBox4.0-Nano and USEtox® 2.12 versions have been used. The comparison of the two models comprised two steps: (i) definition of the USEtox® air, water and soil scenarios in which the main parameters for the Regional and Continental compartments in SB4N have been set to fit those of USEtox®' Urban and Continental compartments, respectively, and (ii) identification of rate constants that are common for the two models.

3 Results and discussion

3.1 Common constants for the two models

In this section, original USEtox® values were selected to be inserted in the USEtox® Air, Water & Soil scenario(s) defined in SB4N (SB4N - *Scenarios* sheet. Landscape settings). As shown in Table 1, only the *Area land* rate constant from the Urban compartment in USEtox® (USEtox®-DEF values) needs to be fed into the rate constants of the Regional scale of SB4N. As indicated in Table 2, *Area land*, *Area sea*, *Fraction fresh water*, *Fraction natural soil*, *Fraction agricultural soil* and *Depth fresh water* constants' s values in the Continental scale of SB4N need to be adjusted to fit with USEtox® (USEtox®-DEF values).

Table 1. Constants of the Regional scale in the SB4N. Values from the Urban compartment of USEtox® (USEtox®-DEF values) with grey background need to be transferred.

Constant – Regional scale	**Variable Name**	**Units**	**SB4N Default scenario**	**USEtox® Air, Water & Soil Scenario**
Area land	AREAland.R	[km^2]	2,3E+05	**2,4E+02**
Area sea	AREAsea.R	[km^2]	1,0E+03	1,0E+03
Fraction lake water	FRAClake.R	-	2,5E-03	2,5E-03
Fraction fresh water	FRACfresh.R	-	2,8E-02	2,8E-02
Fraction natural soil	FRACnatsoil.R	-	2,7E-01	2,7E-01
Fraction agricultural soil	FRACagsoil.R	-	6,0E-01	6,0E-01
Fraction urban/industrial soil	FRACothersoil.R	-	1,0E-01	1,0E-01
Temperature	TEMP.R	[oC]	1,2E+01	1,2E+01
Wind speed	WINDspeed.R	m.s^{-1}	3,0E+00	3,0E+00
Average precipitation	RAINrate.R	mm.yr^{-1}	7,0E+02	7,0E+02
Depth lake water	DEPTHlake.R	m	1,0E+02	1,0E+02
Depth fresh water	DEPTHfreshwater.R	m	3,0E+00	3,0E+00
FRACTION discharge regional fresh water to continental scale	FRAC.w1R.w1C	-	0,0E+00	0,0E+00
Fraction run off	FRACrun.R	-	2,5E-01	2,5E-01
Fraction infiltration	FRACinf.R	-	2,5E-01	2,5E-01
Soil erosion	EROSION.R	mm.yr^{-1}	3,0E-02	3,0E-02

Table 2. Constants of the Continental Scale in SB4N. Values from the Continental compartment of USEtox® (USEtox®-DEF values) with grey background need to be transferred.

Constant – Continental scale	**Variable Name**	**Units**	**SB4N Default scenario**	**USEtox® Air, Water & Soil Scenario**
Area land	AREAland.C	[km^2]	3,7E+06	**9,0E+06**
Area sea	AREAsea.C	[km^2]	3,7E+06	**9,9E+05**
Fraction lake water	FRAClake.C	-	2,5E-03	2,5E-03
Fraction fresh water	FRACfresh.C	-	2,8E-02	**3,0E-02**
Fraction natural soil	FRACnatsoil.C	-	2,7E-01	**4,9E-01**
Fraction agricultural soil	FRACagsoil.C	-	6,0E-01	**4,9E-01**

Constant – Continental scale	Variable Name	Units	SB4N Default scenario	USEtox® Air, Water & Soil Scenario
Fraction urban/industrial soil	FRACothersoil.C	-	1,0E-01	1,0E-01
Temperature	TEMP.C	[oC]	1,2E+01	1,2E+01
Wind speed	WINDspeed.C	$m.s^{-1}$	3,0E+00	3,0E+00
Average precipitation	RAINrate.C	$mm.yr^{-1}$	7,0E+02	7,0E+02
Depth lake water	DEPTHlake.C	m	1,0E+02	1,0E+02
Depth fresh water	DEPTHfreshwater.C	m	3,0E+00	**2,5E+00**
FRACTION discharge continental fresh water to regional scale	FRAC.w1C.w1R	-	0,0E+00	0,0E+00
Fraction infiltration	FRACinf.C	-	2,5E-01	2,5E-01
Fraction run off	FRACrun.C	-	2,5E-01	2,5E-01
Soil erosion	EROSION.C	$mm.yr^{-1}$	3,0E-02	3,0E-02

3.2 Identification of mass balance rate constants that are common for the two models

USEtox® calculates the fate factors from 17 mass balance rate constants (k) [d^{-1}]. Mass balance rate constants are distributed as follows: (i) 4 for the continental environment, (ii) 11 for the intermedia transfer at continental scale, (iii) 1 for the urban environment and (iv) 1 for the intermedia transfer at urban scale (Tables 3-6). The Excel sheets and cells containing such constants in both USEtox® and SB4N have been indicated.

Table 3. Mass balance rate constants used by USEtox® to calculate the FF: Continental environment. Equivalence in SB4N. (N/A refers to Not Available, it has been indicated in italics)

Constant	Denomination (*Fate* sheet; USEtox®)	Excel cell identification (*Run* sheet; USEtox®)	Denomination in SB4N	Excel cell identification (*Engine* sheet, SB4N)	Excel cell identification (*All species output* sheet, SB4N)
k.aC.aU	TRANSFER air - urban scale	G25	N/A in SB4N		
k.aC.aG	TRANSFER air - *global* scale	G31	N/A in SB4N		
k.w1C.w2C	TRANSFER fresh water - coastal seawater	H28	TRANSFER rate continental fresh water - continental sea water (*Continental* sheet, SB4N)	BF67; BG68; BH69; BI70	Not used as a constant in the transport section (Output table 2)
k.w2C.w2G	*TRANSFER coastal seawater* - global *scale*	*I33*	*N/A in SB4N*		

In absence of an Urban compartment and as an alternative to k.aC.aU, SB4N uses k.aC.aR (TRANSFER rate continental air - regional air); Excel cells corresponding to AU9, AV10, AW11 and AX12 (*Engine* sheet, SB4N).

Table 4. Mass balance rate constants used by USEtox® to calculate the FF: Intermedia Transfer at Continental scale. Equivalence in SB4N. (N/A refers to Not Available, it has been indicated in italics)

Constant	**Denomination (*Fate* sheet; USEtox®)**	**Excel cell identification (*Run* sheet; USEtox®)**	**Denomination in SB4N**	**Excel cell identification (*Engine* sheet; SB4N)**
k.aC.w1C	TRANSFER air - fresh water	G27	Not Mentioned	AU63
k.aC.w2C	TRANSFER air - seawater	G28	Not Mentioned	AU67
k.aC.s1C	TRANSFER air - natural soil	G29	Not Mentioned	AU83
k.aC.s2C	TRANSFER air - agricultural soil	G30	Not Mentioned	AU87
k.w1C.aC	TRANSFER fresh water - air	H26	Not Mentioned	BF52
k.w2C.aC	TRANSFER seawater - air	I26	Not Mentioned	BJ52
k.s1C.aC	TRANSFER natural soil - air	J26	Not Mentioned	BZ52
k.s2C.aC	TRANSFER agricultural soil - air	K26	Not Mentioned	CD52
k.s1C.w1C	TRANSFER natural soil - fresh water	J27	TRANSFER rate natural soil – water (*Continental* sheet)	BZ63
k.s2C.w1C	TRANSFER agricultural soil - fresh water	K27	TRANSFER rate agricultural soil – water (*Continental* sheet)	CD63
k.w1C.s2C	*TRANSFER fresh water - agricultural soil*	*H30*	*N/A in SB4N*	

Table 5. Mass balance rate constants used by USEtox® to calculate the FF: Urban environment. Equivalence in SB4N. (N/A refers to Not Available, it has been indicated in italics)

Constant	**Denomination (*Fate* sheet; USEtox®)**	**Excel cell identification (*Run* sheet; USEtox®)**	**Denomination in SB4N**
k.aU.aC	*TRANSFER air - continental scale*	*F26*	*N/A in SB4N*

In absence of an Urban compartment and as an alternative to k.aU.aC, SB4N uses k.aR.aC (TRANSFER rate regional air – continental air); Excel cells corresponding to D52, E53, F54 and G55 (*Engine* sheet, SB4N).

Table 6. Mass balance rate constants used by USEtox® to calculate the FF: Intermedia Transfer at Urban scale. Equivalence in SB4N. (N/A refers to Not Available, it has been indicated in italics)

Constant	Denomination (*Fate* sheet; USEtox®)	Excel cell identification (*Run* sheet; USEtox®)	Denomination in SB4N
k.aU.s3U	*TRANSFER air - continental fresh water*	*F27*	*N/A in SB4N*

The absence of mass balance rate constants in SB4N related with the urban environment including k.aC.aU [TRANSFER air - urban scale]; k.aU.aC [TRANSFER air - continental scale] and k.aU.s3U [TRANSFER air - continental fresh water] can be anticipated, as the Urban compartment does not exist in SB4N; therefore, hereby proposed approach to assimilate it to the Regional compartment in SB4N. However, there are a number of additional constants used in USEtox® that are missing from SB4N, namely: k.aC.aG [TRANSFER air - *global* scale], k.w2C.w2G [TRANSFER coastal seawater - *global* scale] and k.w1C.s2C [TRANSFER fresh water - agricultural soil]. Additionally, k.w1C.w2C [TRANSFER fresh water - coastal seawater] is indeed included in SB4N though it does not appear in the *All species output* sheet in SB4N.

4 Conclusions

In order to derivate FF required for the calculation of CFs, the possibility of combining two different existing models has been evaluated. USEtox® is widely proposed for the LCIA phase (though still it is mostly proposed with limitations, e.g. for the effect factor) whereas SB4N is proposed for fate calculations for ENMs, especially as SB4N allows size-dependent calculations. Our approach to integrate the two models consisted of the identification and assimilation of common mass balance rate constants and aligning common constants that define the environmental compartments. However, this possibility has revealed to be limited since there is no absolute correspondence between the two models. Different approaches to integrate USEtox® and SB4N should be developed and tested in future studies.

Acknowledgement: The present work has received funding from (i) NANOSOLUTIONS (GAN 309329, FP7), (ii) NANOTEK+ (ER-2014/00031, Basque Government) and (iii) Grant IT1302-19 (Basque Government).

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