Special Issue Honouring Helias A. Udo de Haes: LCA Methodology

Continent-specific Intake Fractions and Characterization Factors for Toxic Emissions: Does it make a Difference?

David Rochat[1], Manuele Margni [2,1] and Olivier Jolliet [3,1]*

[1] Life Cycle Systems, GECOS, Institute of Environmental Science and Technology, Ecole Polytechnique Fédérale de Lausanne, Section 2, 1015 Lausanne EPFL, Switzerland
[2] CIRAIG, Ecole Polytechnique de Montreal, C.P. 6079, Succursale Centre Ville, Montreal (Qc) Canada, H3C 3A7
[3] Center for Risk Science and Communication, Department of Environmental Health Sciences, School of Public Health, University of Michigan, Ann Arbor, MI 48109, USA

* **Corresponding author** (ojolliet@umich.edu)

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Abstract

Goal and Scope. This paper aims to develop continental characterization factors for the human toxicity impacts of emissions released to air in different continents and to analyze under which conditions this spatial distinction makes a significant difference compared to generic characterization factors.

Methods. The IMPACT 2002 multimedia and multipathways model has been parameterized to define 6 continental box-models, each of them nested in a world box in order to capture impacts of emissions leaving the initial continent. Applying the model to a test set of 31 heterogeneous chemicals emitted to air, intake fractions and human toxicity characterization factors were calculated for each continent and compared.

Results and Discussion. For a given chemical, characterization factors can vary of typically a factor 5 to 10 between continents (max 10^2), mainly as a function of population density for inhalation and as a function of the total agriculture production per km^2 for ingestion. This is significant but still limited compared to the variation between substances, of 10^6 in intake fraction and of 10^{12} in cumulative risks.

Conclusion. The variation amplitude is limited for persistent chemicals and decreases with the fraction of the chemical advected out of the continent. Moreover, the ranking between continents remains almost the same for all chemicals. Therefore generic characterization factor for air emissions calculated at continental level, such as the one proposed by the common life cycle assessment method, are in most cases suitable for comparative purposes in any other continent. However, continent specific characterization factors are required if one is interested in evaluating absolute values or in comparing impact between scenarios with emissions in very different continents. For this purpose, a simplified but accurate correlation is determined to extrapolate continent specific intake fractions and characterization factors of a wide range of substances for Oceania, Africa, South America, North America and Asia, starting from the results of Europe as a base continent.

Recommandation and Perspectives. Further research should focus on linking the different continental boxes to obtain a global spatial model including major climatic phenomenon such as the air transport by jet stream. The level of spatial resolution, however, has to be carefully selected to capture significant differences, but at the same time to avoid unnecessarily requirement efforts for data gathering and calculation capabilities.

Keywords: Continent-specific variation; human toxicity; intake fraction; life cycle assessment (LCA); toxic emissions

Introduction

The development of generic characterization factors (*CF*) in life cycle impact assessment (LCIA) is historically motivated by the lack of spatial and temporal information when determining the environmental interventions per functional unit. These generic characterization factors are well adapted to evaluate global impacts, such as global warming and ozone layer depletion, but face criticisms when assessing all those impact categories that are not global in nature such as acidification, eutrophication, toxicity, etc. From a scientific point of view, one of the major problems is the inability to adequately model impacts due to a common disregard of the spatial differences in the fate and exposure and in the effect of environmental stressors (Udo de Haes et al. 2002). From a practical point of view, accounting for spatial differentiation in LCA remains complicated by the lack of spatial distinction maintained in most emissions and resource consumption inventory databases. However, there is an increasing demand on impact assessment methodologies reflecting regional concerns and being adapted to the local conditions for such impact that are not global in nature. It is not surprising having practitioners being reluctant applying characterization factors developed for a European context to assess impacts of toxic emissions related to another continent. This paper therefore aims to develop characterization factors for toxic air emissions in different continents and to analyze under which conditions this spatial distinction makes a significant difference compared to generic characterization factors. In addition, adapting LCIA methods to developing countries is one of the most important needs and objectives of the Life Cycle Initiative (Jolliet et al. 2004, Stewart and Jolliet 2004).

Several publications have quantified the variability linked to spatial inhomogeneity in multimedia modeling at national or regional scale (Klepper and den Hollander 1999, McKone et al. 2001, MacLeod et al. 2001, MacLeod et al. 2004, Prevedouros et al. 2004, Pennington et al. 2005, Wegener Sleeswijk 2005). Disregarding the release location, results demonstrated likely variations of up to two or three orders of magnitude in the chemical concentrations and human intake fractions, particularly for emissions to water. The variability linked to the release location could even increase up to 6 orders of magnitude (MacLeod et al. 2004, Pennington et al. 2005). Based on such findings, MacLeod et al.

(2004) provided 4 chemical specific regressions to extrapolate exposure estimates from the population density and the food production intensity variables. These correlations are however substance specific and cannot be used for extrapolation purposes across a wide range of substances.

All these works were focusing on a spatial differentiation with reference to zones of about 5 to 10 hundreds square kilometers. Characterization factors for human toxicity, HDF, at continental level have mainly been published for US (Hertwich et al. 2001), for Europe (Goedkoop et al. 2000, Huijbregts et al. 2000, Jolliet et al. 2003) and for Japan (Itsubo 2003). Little information is published for other continents and the existing ones cannot be compared on a consistent basis. Huijbregts and co-authors (2003) investigated the uncertainty in fate and exposure factors of different generic continent-specific environments, using a consistent model. They find out this could be moderately high, between a factor 2 to 10. They also propose correlations relating Australia and US to European factors, but without accounting for the specific chemical properties and parameters that determine if impact is mostly local or global. In addition, the authors claimed for further research to investigate whether the systematic differences found between the different evaluative environments are of direct relevance for LCA purposes.

We therefore aim to calculate differentiated intake fractions (*iF*) and human toxicity characterization factors for different continents using a consistent model to answer the following questions:

- How to model *iF* and characterization factors for various continents, taking into account the specific chemical properties?
- What is the data availability and variability at world level for calculating *iF*?
- How far is the variability of *iF* and *CFs* between continents significant? How does it compare to the variations between chemicals?
- What are the environmental and geographical key parameters affecting iF and its variation across continents?
- How to derive a general relationship to extrapolate continent-specific *iFs* and *CFs* of a wide range of specific chemicals, starting from the modeled intake fraction of a base continent.

We will first introduce the methodology in section 1, presenting the selected model, its structure and the data used to parameterize the different continents. In section 2, we will present results for a test set of 31 chemicals and analyze the continental variability in intake fractions as a function of chemical properties. We then propose a simplified but accurate method to extrapolate continent-specific *iFs* and *CFs* based both on chemical specific properties and on continent specific properties such as population densities. Results are focused on an air emission scenario, as air emissions are the most likely to involve both local impacts and long range transportation. In the conclusion (section 3) we will finally discuss the question contained in this paper title: does it make a difference and under which conditions?

1 Method

1.1 Framework and selection of the model

Characterization factors for toxicological impacts, *CF* [Impact/Mass] are based on models that account for chemical fate in the environment – *F* [time], human exposure – *E* [time^{-1}], and differences in toxicological response, as defined by the effect factor – *EF* [Impact/Mass]. This can be expressed in the following simplified equation (Guinee et al. 1996, Jolliet 1996, Goedkoop et al. 1999, Huijbregts et al. 2000, Hertwich et al. 2001, Udo de Haes et al. 2002):

$$CF_i^{me} = F_i^{mn} \cdot E_i^{nr} \cdot EF_i^{re} = iF_i^{mr} \cdot EF_i^{re} \quad (1)$$

The intake fraction, *iF* [dimensionless] combines fate and exposure factors describing the fraction of an emission that is ultimately taken in by a population (Bennett et al. 2002b). Subscript *i* describes a given chemical, superscripts *m* the emission compartment, *n* the environmental compartment, *r* the route of exposure and *e* the effect type (e.g. cancer or non-cancer). As effect factors in LCA are usually assumed to be additive, linear and independent of the time and location of exposure, the characterization factor is assumed linearly proportional to *iF*.

Among the existing multimedia and multipathways exposure models (McKone 1993, Brandes et al. 1996, Huijbregts et al. 2000, MacLeod et al. 2001, Pennington et al. 2005), the authors selected IMPACT 2002 (Pennington et al. 2005) as it is well adapted for studying spatial differentiation. It consists of a common multimedia fate, a multipathways exposure model, and two effect modules for human health and ecotoxicity. IMPACT 2002 enables estimation of chemical concentrations in environmental media at a regional and a global scale. The human multiple pathways exposure module links chemical concentrations in environmental media (atmosphere, soil, surface water, and vegetation) calculated by the fate model to human intake though inhalation and ingestion. Ingestion pathways include drinking water consumption; incidental soil ingestion; and intake of contaminants in agricultural products (fruits, vegetables, grains,…) as well as in animal products, such as beef-, pig-, and poultry-meat, eggs, fish, and milk. Intake fractions are calculated from the contaminant concentration in food and livestock production levels at each location, the water extracted to serve a given population at each location, as well as the population distribution when considering inhalation. The agricultural vegetation module in the chemical fate model IMPACT 2002 distinguishes two major types of vegetation, as suggested by McKone (1993): exposed and unexposed produced. The first one being exposed to atmospheric deposition, similar to foliage in the fate module, and the second protected from such direct contact with the atmosphere like stems and comestibles roots.

Cumulative risk and potential impact per kg of emission are calculated by combining cumulative chemical intake with risk-based effect factors. However, a detailed study of human risks remain outside the main scope of the present study, which is mainly focused on fate and exposure.

1.2 Adapting IMPACT 2002 to other continents

Six continental models are developed by adapting the Western European model to all continents worldwide. A typical nested approach (Cowan et al. 1994) was adopted with a continental box nested in a world box to account for any intake that may occur as a result of contaminant advection outside of the initially considered continental region. This is in line with the broadness of the LCA approach, accounting for the overall impacts both within and outside the continent of emission. The geographical boundaries of the continental boxes are shown in **Fig. 1**.

Parameters affecting the fate, the exposure, and thus the human intake fraction were specifically collected for each continent. As a first approximation we decided to modify a selected number of parameters responsible for the highest variations between continents. Geographical data such as surface area, the share of land, fresh water and marine water, as well as the freshwater mean depth were calculated using Geographic Information System (GIS). Mean annual precipitation and runoff data (rainfall – evapo-transpiration) were taken from 0.5 x 0.5 degree grid data from the Global Run Off Data Centre (GRDC) (Global Runoff Data Centre 2004). Annual average air flow are calculated using the underlying wind velocity data of the model GEOS-CHEM (Bey et al. 2001) and the perpendicular cross-sectional areas, with air sub-divided according to a grid. Population data were taken from the CIA factbook (CIA 2004). In this simplified data gathering procedure, default values of IMPACT 2002 such as soil depth, pH, suspended particulate matter, OH concentration, etc. remained unchanged for all the continents (Pennington et al. 2005) as the impact of their variability is restricted at a continental scale. **Table 1** summarizes the collected data specific to each continent and describes the corresponding literature sources.

Human exposure via food is linked to the location where the food is produced. Food agricultural production statistics were taken from Faostat database (FAO 2004). Food production for each continent is given in **Table 2** and summed up to a world production. The model takes into account the

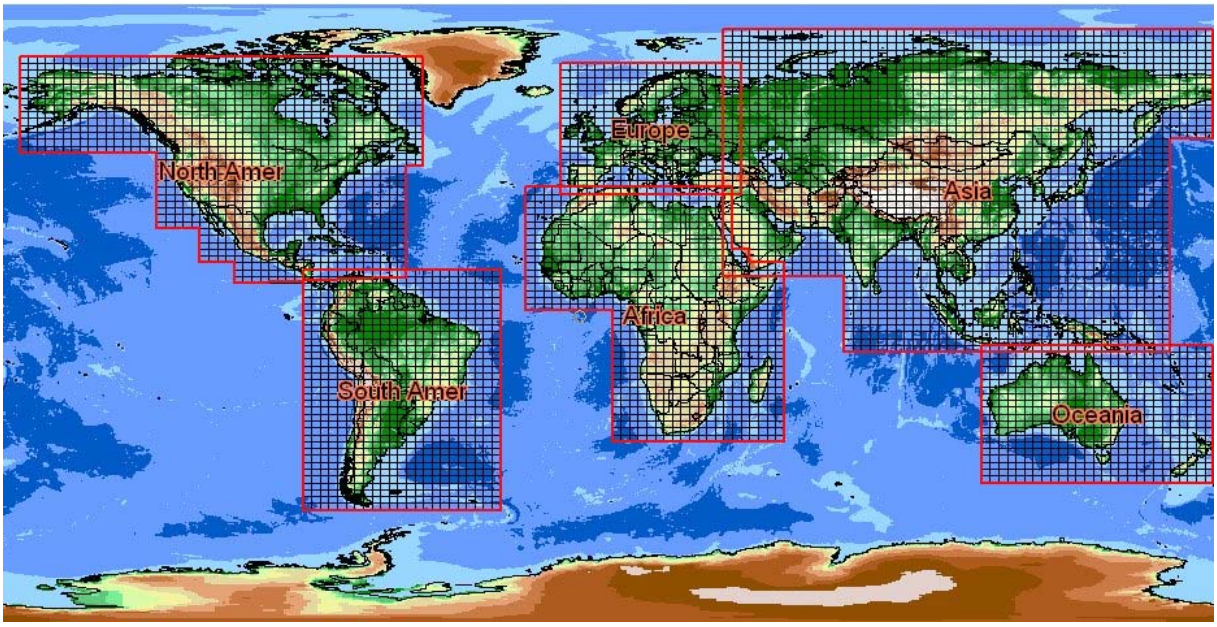

Fig. 1: Areas covered by the continental boxes

Table 1: Main geographical and environmental parameters

	Africa	Asia	Europe	N America	Oceania	S America	World	*Source*
Population	$7.96{\cdot}10^{+8}$	$3.76{\cdot}10^{+09}$	$6.51{\cdot}10^{+08}$	$4.89{\cdot}10^{+08}$	$3.10{\cdot}10^{+07}$	$3.47{\cdot}10^{+08}$	$6.07{\cdot}10^{+09}$	*CIA factbook*
Soil Area (m²)	$3.01{\cdot}10^{+13}$	$4.63{\cdot}10^{+13}$	$7.74{\cdot}10^{+12}$	$2.08{\cdot}10^{+13}$	$8.07{\cdot}10^{+12}$	$1.77{\cdot}10^{+13}$	$1.31{\cdot}10^{+14}$	*calculated with GIS*
Seawater Area (m²)	$1.95{\cdot}10^{+13}$	$5.51{\cdot}10^{+13}$	$6.49{\cdot}10^{+12}$	$2.54{\cdot}10^{+13}$	$2.05{\cdot}10^{+13}$	$2.56{\cdot}10^{+13}$	$3.65{\cdot}10^{+14}$	*calculated with GIS*
Freshwater Area (m²)	$6.83{\cdot}10^{+11}$	$1.05{\cdot}10^{+12}$	$1.50{\cdot}10^{+11}$	$1.29{\cdot}10^{+12}$	$7.30{\cdot}10^{+10}$	$3.01{\cdot}10^{+11}$	$3.54{\cdot}10^{+12}$	*calculated with GIS*
Freshwater mean depth (m)	46.00	13.00	15.00	20.00	3.00	8.00	23.5	*calculated with GIS*
Precipitation (m/hour)	$5.71{\cdot}10^{-05}$	$5.71{\cdot}10^{-05}$	$7.99{\cdot}10^{-05}$	$4.57{\cdot}10^{-05}$	$2.85{\cdot}10^{-05}$	$1.14{\cdot}10^{-04}$	$3.83{\cdot}10^{-05}$	*Faostat*
Mean runoff (m³/hour)	$5.15{\cdot}10^{+08}$	$1.68{\cdot}10^{+09}$	$2.29{\cdot}10^{+08}$	$6.34{\cdot}10^{+08}$	$7.37{\cdot}10^{+07}$	$1.35{\cdot}10^{+09}$	$4.48{\cdot}10^{+09}$	*GRDC*
Average air flow (m³/hour)	$3.85{\cdot}10^{+13}$	$5.70{\cdot}10^{+13}$	$2.04{\cdot}10^{+13}$	$2.62{\cdot}10^{+14}$	$6.04{\cdot}10^{+14}$	$5.69{\cdot}10^{+14}$		*Geoschem*
Average marine flow (m³/hour)	$1.97{\cdot}10^{+11}$	$1.00{\cdot}10^{+12}$	$1.67{\cdot}10^{+11}$	$4.18{\cdot}10^{+11}$	$4.01{\cdot}10^{+11}$	$4.05{\cdot}10^{+11}$		*Mariano surface velocity model*

Table 2: Production data in kg/year (source: FAOstat database)

Production in (kg/year)	Africa	Asia	Europe	N America	Oceania	S America	World
Unexposed produce	$3.21 \cdot 10^{+11}$	$1.13 \cdot 10^{+12}$	$3.64 \cdot 10^{+11}$	$3.00 \cdot 10^{+11}$	$5.60 \cdot 10^{+10}$	$5.41 \cdot 10^{+11}$	$2.71 \cdot 10^{+12}$
Exposed produce	$1.84 \cdot 10^{+11}$	$1.46 \cdot 10^{+12}$	$5.51 \cdot 10^{+11}$	$5.28 \cdot 10^{+11}$	$4.41 \cdot 10^{+10}$	$1.53 \cdot 10^{+11}$	$2.92E^{+12}$
Fresh water fish	$2.39 \cdot 10^{+09}$	$2.38 \cdot 10^{+10}$	$8.83 \cdot 10^{+08}$	$5.81 \cdot 10^{+08}$	$1.88 \cdot 10^{+07}$	$5.48 \cdot 10^{+08}$	$2.83 \cdot 10^{+10}$
Pigs	$6.98 \cdot 10^{+08}$	$4.84 \cdot 10^{+10}$	$2.50 \cdot 10^{+10}$	$1.16 \cdot 10^{+10}$	$4.73 \cdot 10^{+08}$	$3.01 \cdot 10^{+09}$	$8.91 \cdot 10^{+10}$
Beef	$4.26 \cdot 10^{+09}$	$1.37 \cdot 10^{+10}$	$1.16 \cdot 10^{+10}$	$1.55 \cdot 10^{+10}$	$2.58 \cdot 10^{+09}$	$1.18 \cdot 10^{+10}$	$5.95 \cdot 10^{+10}$
Broilers	$3.10 \cdot 10^{+09}$	$2.31 \cdot 10^{+10}$	$1.17 \cdot 10^{+10}$	$2.03 \cdot 10^{+10}$	$7.67 \cdot 10^{+08}$	$9.69 \cdot 10^{+09}$	$6.87 \cdot 10^{+10}$
Goat and Sheep meat	$1.80 \cdot 10^{+09}$	$6.03 \cdot 10^{+09}$	$1.48 \cdot 10^{+09}$	$2.08 \cdot 10^{+08}$	$1.23 \cdot 10^{+09}$	$3.32 \cdot 10^{+08}$	$1.11 \cdot 10^{+10}$
Eggs	$1.98 \cdot 10^{+09}$	$3.32 \cdot 10^{+10}$	$9.35 \cdot 10^{+09}$	$7.59 \cdot 10^{+09}$	$2.02 \cdot 10^{+08}$	$2.87 \cdot 10^{+09}$	$5.52 \cdot 10^{+10}$
Dairy products (Cow milk)	$2.50 \cdot 10^{+10}$	$1.69 \cdot 10^{+11}$	$2.10 \cdot 10^{+11}$	$9.74 \cdot 10^{+10}$	$2.35 \cdot 10^{+10}$	$4.49 \cdot 10^{+10}$	$5.70 \cdot 10^{+11}$
Sea fish	$6.07 \cdot 10^{+09}$	$2.77 \cdot 10^{+10}$	$1.05 \cdot 10^{+10}$	$6.83 \cdot 10^{+09}$	$5.75 \cdot 10^{+09}$	$1.74 \cdot 10^{+10}$	$7.42 \cdot 10^{+10}$

export of food and the fraction of produced food used to feed animals or for industrial use.

Figs. 2 and **3** show important differences in population density and food production per km² of more than one order of magnitude. These variations in exposure parameters are likely to be reflected in significant variations between continent-specific intake fractions.

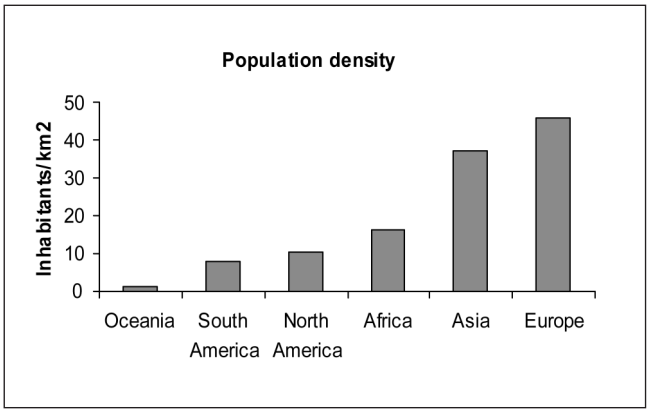

Fig. 2: Population density (inhabitants/km²) in the 6 continents

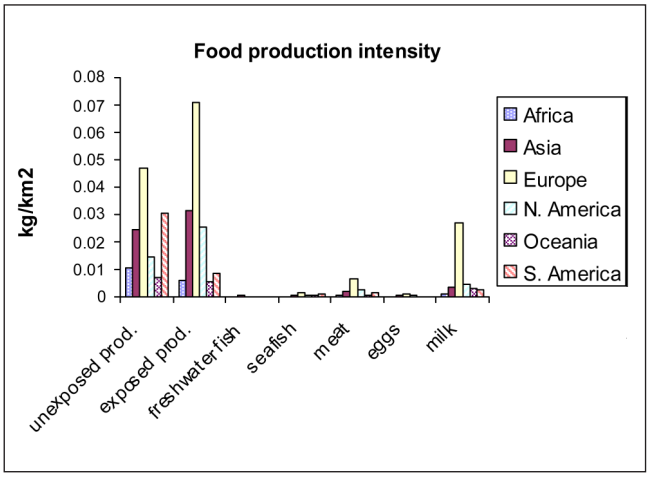

Fig. 3: Food production intensity (kg/km²) for human consumption

2 Results and Discussion

2.1 Comparison of intake fractions (*iF*)

A set of representative organic, non-dissociating chemicals was used for this comparison, covering plausible differences in partitioning behavior, dominant human exposure pathways, overall environmental persistence, and long-range transport characteristics. Chemical properties were assumed to reflect variations under average conditions for a broad range of chemicals (Margni 2003). This dataset was also used within the OMNIITOX project (Molander et al. 2004). The model was run for a constant emission at the rate of 1 kg/hour in air in different continents, leading to calculation of the Intake Fraction, that is independent of the emission rate. First, the intake fraction for the entire set of substances is presented. Then, detailed results are illustrated and discussed for four specific substances selected on the basis of their widely different chemical properties covering the combinations of high and low octanol-water partitioning coefficient, K_{OW}, and high and low persistence in air (physical-chemical properties of the representative chemicals are given in the **Supporting Information**, online only, see DOI: http://dx.doi.org/10.1065/lca2006.05.XXX). The cumulative risk is finally calculated for each chemical as the intake fraction is multiplied by the dose-response slope, which is assumed equal for all continents.

Fig. 4 shows the variation in intake fraction between continents for an emission to air. It enables to discuss how far these differences are important compared to the variability between substances. Intake fractions vary significantly up to about 10^2 between continents depending on the considered substance. This is, however, still limited compared to the typical variation of 10^6 between substances for ingestion. Interestingly, **Fig. 4a** shows that the variation between continents is very small for high intake fraction by inhalation. This corresponds to highly persistent substance in air, thus a more or less uniform concentration increase worldwide whatever the location of emission.

The ranking of the continents is almost the same for every substance. The magnitude of variation between continents is related to population density and to differences in the intensity of exposed food production as shown by the following detailed analysis on the four selected substances.

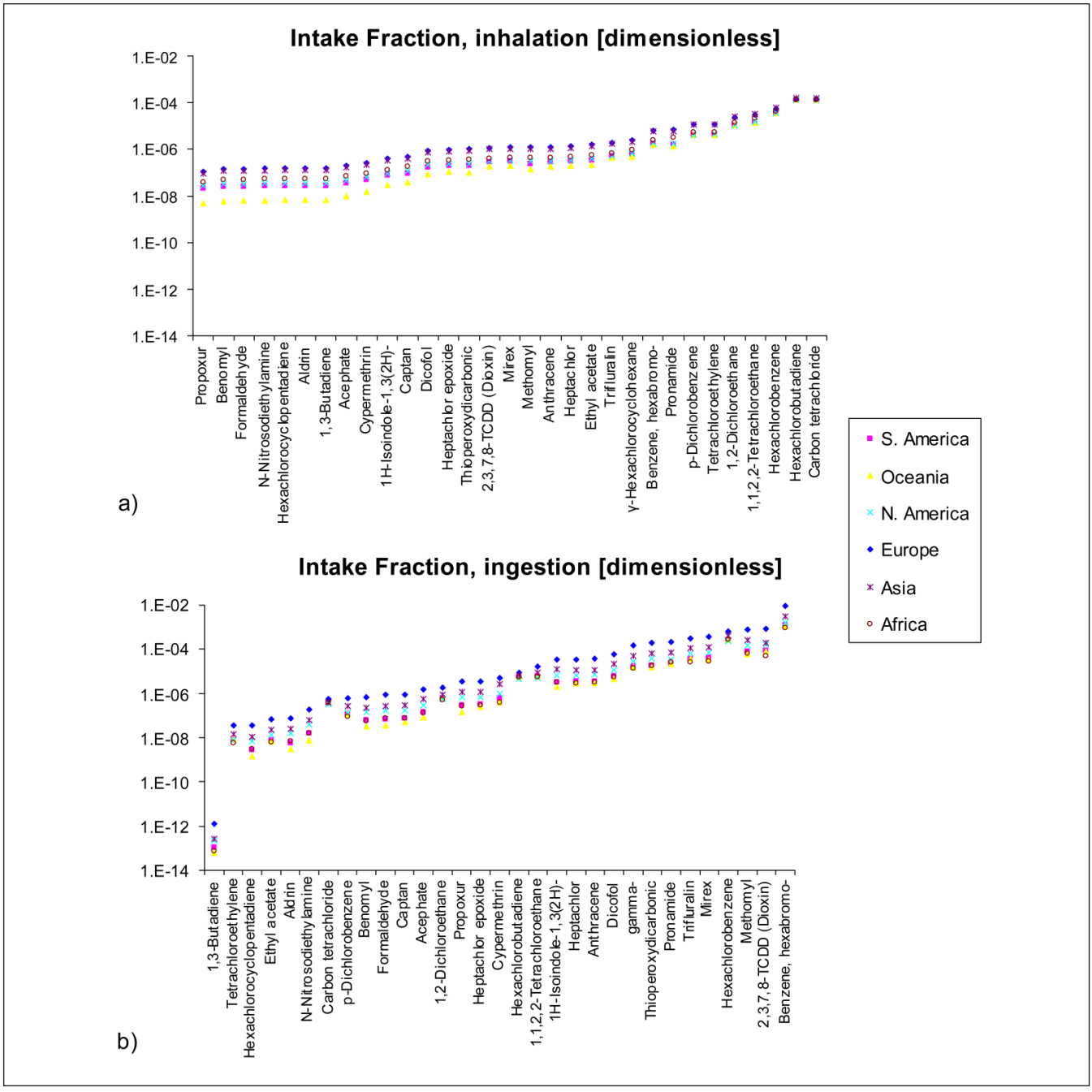

Fig. 4: Intake fraction variability for a dataset of 31 chemicals released to the air compartment of 6 different continental models (South America, Oceania, North America, Europe, Asia and Africa), each nested in a world box. Inhalation a) and ingestion b) exposure route are ordered by increasing *iF*

2.2 Detailed *iF* analysis for four substances

Because of a relatively small K_{OW}, ***tetrachloroethylene*** (**Fig. 5a**) does not bio-accumulate significantly, which explains the small intake fraction for this substance in agreement with the observations by Bennett et al. (2002a). The exposure is dominated by inhalation because of a relatively high Henry's Law constant. Moreover, its relatively fast degradation rate in air competes with the air advection rates for large continents, such as Africa, Asia and Europe, implying that the intake is dominated by the continent of emission and do not exceeds a fraction of 1 per 100,000: 1kg emitted causes a population intake of up to 10 mg. For less densely populated continents, such as Oceania and South America, exposure occurs mainly at the global level and is one order of magnitude smaller.

Similarly to tetrachloroethylene, ***carbon tetrachloride*** (**Fig. 5b**) shows a low K_{OW} and does not bio-accumulate. However, its partition to air (high Henry's Law constant) and persistence in the same medium is significantly higher (more than 1 order of magnitude) than for tetrachloroethylene. This leads

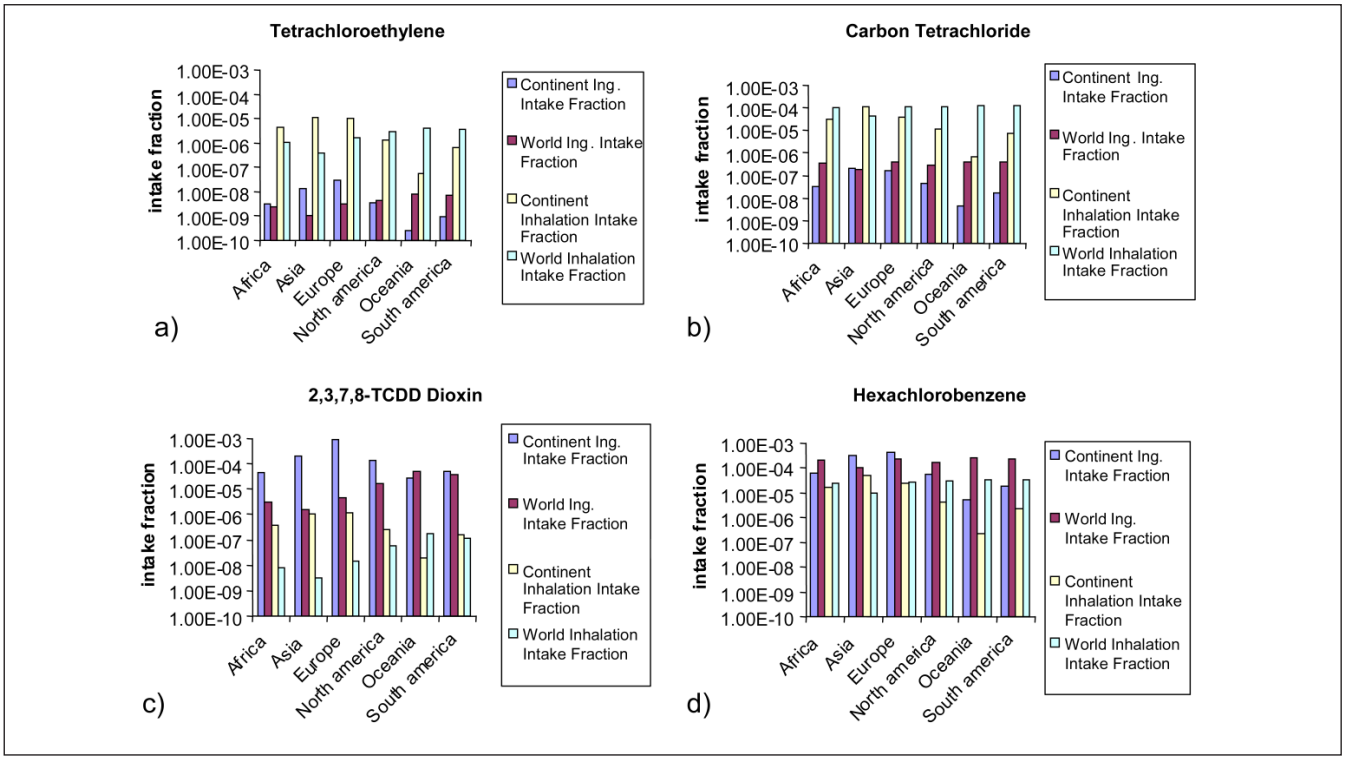

Fig. 5: Intake fraction for an emission to air of tetrachloroethylene a), carbon tetrachloride b), 2,3,7,8-TCDD Dioxin c) and hexachlorobenzene d) detailed per emission continent and exposure pathways (ingestion and oral)

to a higher intake fraction by inhalation that is rather uniform worldwide. The continent-specific impacts are therefore proportional to the population and higher for Asia, which accounts for half of the world population.

On the other hand, the next two substances, **dioxine** and **hexachlorobenzene** have relatively low Henry Law constants and high K_{OW}, which means that pollutants tend to leave the air compartment and bioconcentrate in the food chain (**Figs. 5c,d**). The differences in air degradation explain that dioxin (relatively short half life in air) mainly affects the continent where it is emitted. Its high bioconcentration factor in vegetable, milk and meat leads to very high intake fractions by ingestion of up to 1 per thousand, especially in Europe that shows the highest fraction of cultivated land, coupled with high agriculture production intensity (see Fig. 3). These values are in the same order of magnitude as experimentally based intake fraction for dioxin of 0.003 for Europe (Margni et al. 2004) and 0.002 for USA (Bennett et al. 2002a). Hexachlorobenzene has a more uniform impact worlwide than dioxin because of its extremely high persistence in all environmental compartments.

2.3 Extrapolation for different continents

As shown by MacLeod et al. (2004), the intake fraction mostly varies according to different substance specific linear regressions, as a function of the population density for inhalation and as a function of the food production rates for ingestion. However, in the context of continent-specific variation, it would be highly valuable to establish a more general relationship enabling extrapolating the intake fraction of a wide range of substances, starting from the modeled intake fraction of a base continent – Europe in the case of Impact 2002. Figs. 4 and 5 suggest that the variability of the intake fraction between continents decreases as a function of the residence time in air, ultimately leading to a constant worldwide concentration and intake fraction. In other words, the higher the fraction advected from the specific continent to the world, the lower the variation between continents. Following an analysis of the mass balance equations, we have therefore plotted the ratio of continental to European intake fractions by inhalation as a function of the fraction of the substance advected out of Europe (**Fig. 6a**). For local pollutant with little advection out of Europe, the iF_i^c / iF_i^{Europe} ratio is close to the ratio of the population densities ($P_{dens}^c / P_{dens}^{Europe}$). This ratio increases linearly with the advected fraction up to one for very persistent substances.

The Intake fraction for a substance *i* emitted to air in a continent *c* can therefore be approximated by the following relationship:

$$iF_{i\ inhalation}^{c} = iF_{i\ inhalation}^{Europe}\left(P_{dens}^{c} / P_{dens}^{Europe} + \beta_{inhalation}^{c}\left(1 - P_{dens}^{c} / P_{dens}^{Europe}\right) k_{a,adv\ i}^{Europe} / k_{a,tot\ i}^{Europe}\right) \quad (2)$$

Where the ratio of population densities and the slope are given in **Table 3**.

Interestingly, for all continents but for Asia, the slope $\beta_{inhalation}$ is close to 0.58, the R^2 higher than 0.94 and the 95% confidence interval on individual prediction lower than

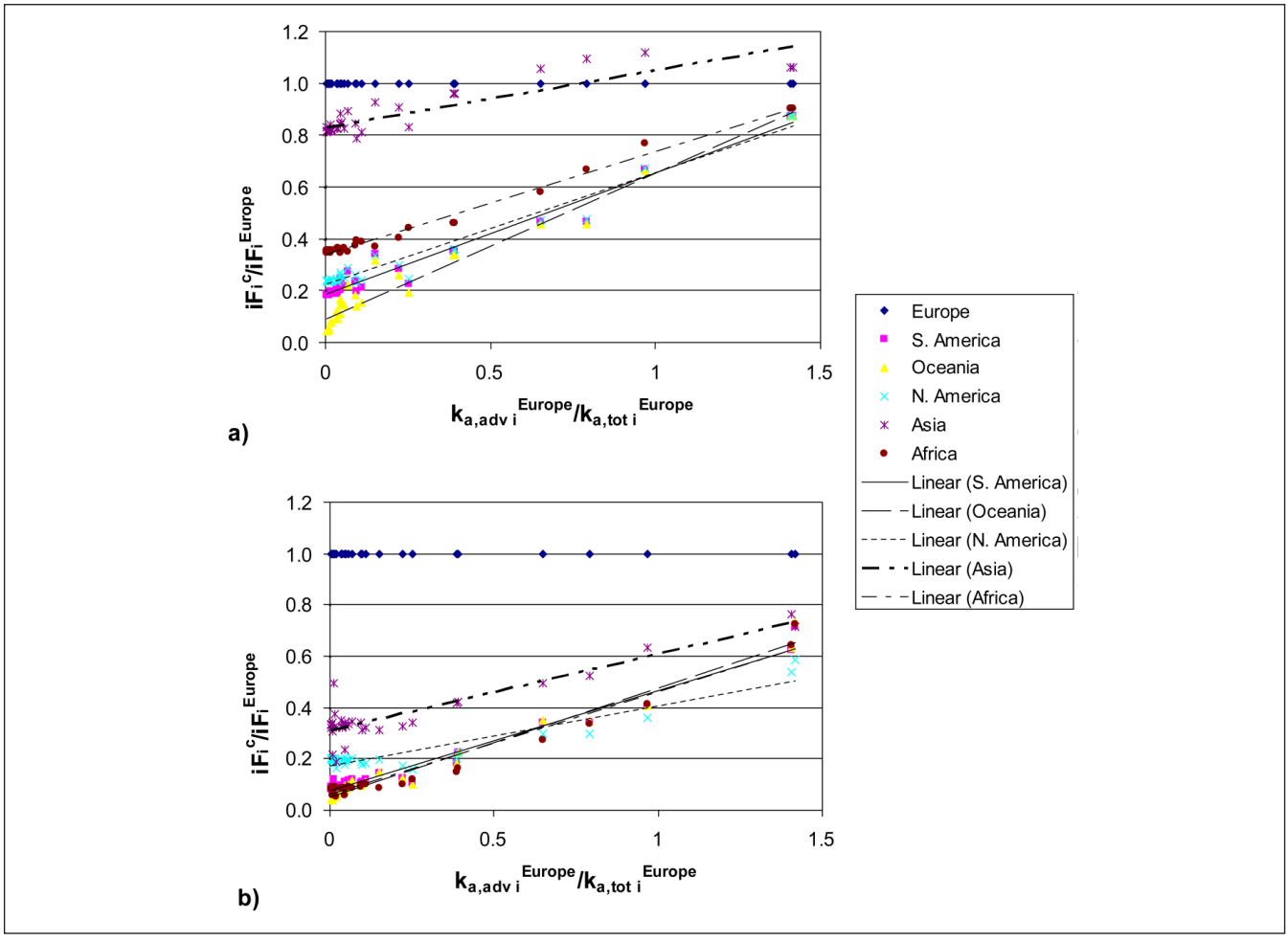

Fig. 6: Ratio of continental to European intake fractions (iF_i^c / iF_i^{Europe}) for an emission to air as a function of the fraction advected out of Europe ($k_{a,adv\,i}^{Europe} / k_{a,tot\,i}^{Europe}$), where the advection rate out of Europe is 0.00080 1/h and $k_{a,tot\,i}^{Europe}$ is the overall rate constant in air for substance i released in Europe. a) inhalation route b) ingestion route

15%. The higher variation with Asia (R^2=0.84) is linked to the fact that Asia represents in itself 62% of the world population. For very persistent substances, a significant part of the advection out of Asia is nevertheless taken in later by the Asian population itself. This feedback effect has been discussed in detail by Margni et al. (2004) and explains why the advected fraction can be higher than one for very persistent pollutants.

Table 3: Parameters and statistical data related to the extrapolation curves in Eqs. 2 and 3

Inhalation (Eq.2)	**S. America**	**Oceania**	**N. America**	**Asia**	**Africa**
Intercept $P_{dens}{}^c/P_{dens}{}^{Europe}$	0.18	0.02	0.23	0.81	0.35
Slope $\beta^c_{inhalation}$	0.58	0.65	0.55	1.28	0.59
Standard error on slope	0.02	0.03	0.02	0.10	0.01
R^2	0.98	0.94	0.97	0.84	0.99
Standard error on individual prediction	0.04	0.07	0.04	0.24	0.03
95% confidence interval on prediction	0.08	0.15	0.08	0.49	0.06
Ingestion (Eq.3)					
Intercept $\beta^c_{ingestion}$	0.08	0.05	0.17	0.31	0.06
Standard error on intercept	0.01	0.01	0.01	0.01	0.01
Slope $\beta^c_{ingestion}$	0.39	0.42	0.23	0.30	0.40
Standard error on slope	0.01	0.01	0.01	0.02	0.01
R^2	0.97	0.97	0.88	0.84	0.96
Standard error on individual prediction	0.02	0.02	0.04	0.08	0.03
95% confidence interval on prediction	0.05	0.05	0.07	0.17	0.07

Similar figures and equations can be established for the ingestion pathway (**Fig. 6b**). As shown in the **Supporting Information** (online only, see DOI: http://dx.doi.org/10.1065/lca2006.05.xxx), the intercept is related to the amount of food produced per unit area in each continent. It is especially the exposed vegetation that dominates the intake in most cases, except for a few substances, for which milk and meat are significant. The relationship is, however, not as direct as with the population densities for inhalation, due to the variety of intake pathways and to the variation in vegetation volume across continents. The corresponding approximation for ingestion is therefore given by:

$$iF^{c}_{i\,ingestion} = iF^{Europe}_{i\,ingestion}\left(\alpha_{ingestion} + \beta_{ingestion}\, k^{Europe}_{a,adv\,i} \Big/ k^{Europe}_{a,tot\,i}\right) \qquad (3)$$

where the intercept value $\alpha_{ingestion}$ and the slope $\beta_{ingestion}$ are given in Table 3 for each continent.

The proposed correlation explains more than 84% of the variability and even more than 96% for Oceania, Africa and South America.

2.4 Overall cumulative risks

Once intake fractions are combined with effect factors as proposed by Crettaz and colleagues (Crettaz et al. 2002, Pennington et al. 2002), cumulative risks vary significantly of about 10^2 between continents depending on the considered substance (**Fig. 7**). This is, however, relatively low compared to the variation of 10^{12} between substances.

3 Conclusion

This project showed the feasibility to readily determine generic characterization factors for different geographical world regions, using publicly available data to parameterize multimedia and multipathways exposure. Results show that despite important variations in characterization factors relative to which continent the pollutant is emitted:

- this remains restricted to two orders of magnitude compared the variations of the entire set that achieves up to twelve orders of magnitude, and
- the ranking tends to remain constant supporting the choice to use of generic characterization factors, as suggested in common life cycle impact assessment methods, for LCA studies.

The main parameters affecting continent-specific variations are the population density for the inhalation route and the total agricultural production for the ingestion route of exposure confirming the findings of MacLeod et al. (2004). The study of four substances also showed that population density and agriculture cultivated areas may affect the magnitude and location of the impact, which may happen outside the continent in which the substance is emitted. The more persistent the substance is, the higher the impact outside its continent of emission and the less variation is observed between continents.

Generic characterization factors are not sufficiently precise to determine absolute values or to compare impacts from two scenarios whose major emissions takes place in different continents. In this case, the continent specific characterization factors should be considered. The main parameters affecting continent-specific variations are the population density for the inhalation route and the total agricultural production for the ingestion route of exposure. For this purpose we proposed a simplified method enabling extrapolating continent specific intake fractions of a wide range of substances, starting from the modeled intake fraction of a base continent as a function of the fraction of the chemical advected out of the region. Eqs. 2 and 3 enable to extrapolate the intake fraction for any continent, based on the European intake fraction, with more than 84% of the variability explained. The 95% confidence

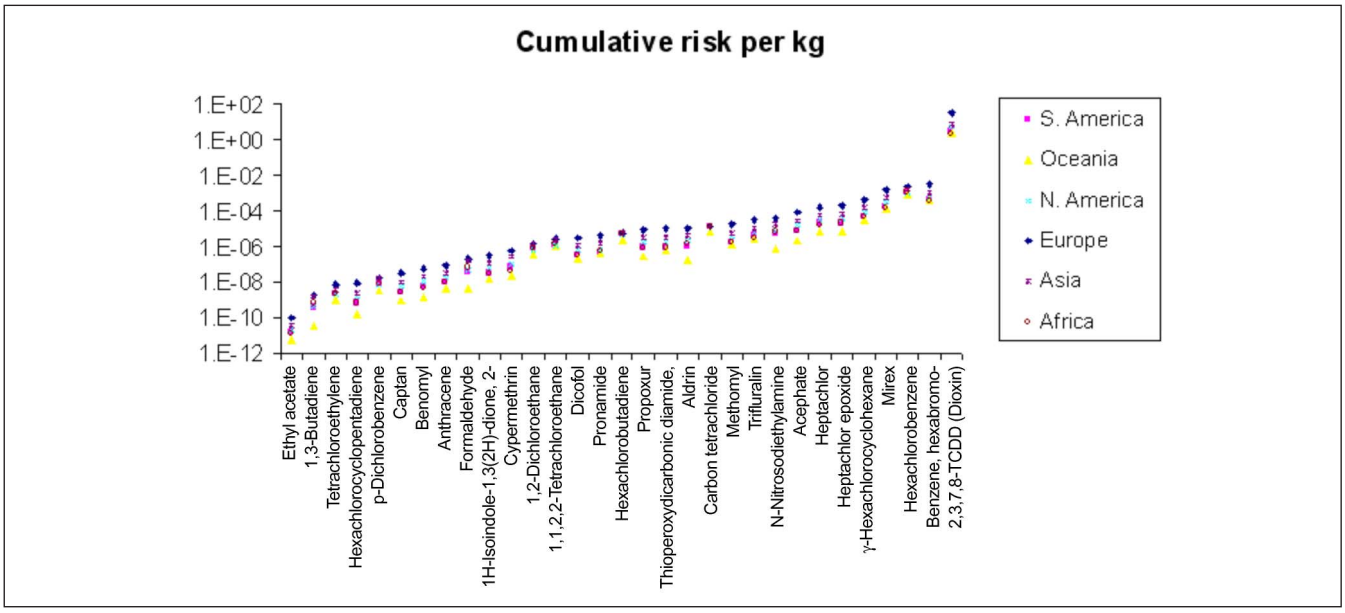

Fig. 7: Cumulative risk per kg of substance emitted for the OMNITOX dataset

interval of 5% to 50% are low compared to the overall variation in intake fraction of 6 to 10 orders of magnitude and the 12 orders of magnitude in cumulative risk. This simplified method could be readily adapted to extrapolate continent-specific *iF* for any other model. As these results and correlations refer to an air emission scenario, they need to be further extended to consider other media of release, resulting in potentially different spatial variabilities and correlations. It would also be highly interesting to test the proposed regression at national or regional, taking profit of the GLOBACK database (Wegener Sleeswijk 2005).

As impact in the world box can even be dominant for some chemical, further research should focus on linking the different continental boxes to obtain a global spatial model comparable to the European spatial model (Pennington et al. 2005) or to extend the world model proposed by Toose and colleagues (Toose et al. 2004) by adding exposure to the fate modeling. The level of spatial resolution has to be carefully selected: The aim is to capture significant differences, but at the same time to avoid unnecessarily requirement efforts for data gathering and calculation capabilities. Moreover, some major climatic phenomenon must be included in the modeling, such as considering an upper air level to include high altitude inter continental substance transport by jet stream.

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Appendix: Supporting information

The appendix can be found in the online edition of this paper. You can access the online edition via the website <DOI: http://dx.doi.org/10.1065/lca2006.05.012>

Supporting Information (online only)

1 Physical-chemical Properties of the Set of Representative Organic, Non-dissociating Chemicals

Chemicals in **Table S1** were selected from approximately 500 non-dissociating organic chemicals using data adopted analogous to US EPA's draft WMPT tool data selection hierarchy (USEPA 1998). Data are from, in order of typically preference adopted, Mackay et al. data compilation handbooks (Mackay et al. 1995), Howard et al. data compilation handbooks (Howard 1991, Howard et al. 1991), Physprop experimental data (Syracuse Research Corporation), Epiwin experimental data (Howard et al. 2002), Physprop estimated data (Syracuse Research Corporation), Epiwin estimated data (Howard et al. 2002). Data gaps were additionally filled using the CalTox model (McKone et al. 2001) and the USES-LCA model (Howard et al. 2002) databases.

Table S1: Physical-chemical properties of the set of 31 representative organic, non-dissociating chemicals

Name	CAS	Molecular Mass (g/mole)	Henry's Constant (Pa m3 mol-1) or Kaw	Log Kow	air degradation half life (hours)	water degradation half life (hours)	sediment degradation half life (hours)	vegitation degradation half life (hours)	Soil degradation half life (hours)
Heptachlor epoxide	1024-57-3	389.32	1.50E+02	5.14E+00	3.30E+01	7.02E+03	9.60E+01	3.30E+01	7.02E+03
p-Dichlorobenzene	106-46-7	147.01	2.97E+02	3.47E+00	5.50E+02	1.70E+03	1.70E+04	5.50E+02	5.50E+03
1,3-Butadiene	106-99-0	54.09	2.57E+05	1.99E+00	5.00E+00	1.70E+02	1.70E+03	5.00E+00	5.50E+02
1,2-Dichloroethane	107-06-2	98.96	1.17E+02	1.44E+00	1.70E+03	1.70E+03	1.70E+04	1.70E+03	5.50E+03
Propoxur	114-26-1	209.24	4.50E-05	1.50E+00	5.00E+00	5.50E+02	1.70E+03	5.00E+00	5.50E+02
Dicofol	115-32-2	370.49	5.67E-05	5.02E+00	7.01E+01	8.99E+02	3.84E+02	7.01E+01	1.46E+03
Hexachlorobenzene	118-74-1	284.79	7.82E+01	5.50E+00	1.70E+04	5.50E+04	5.50E+04	1.70E+04	5.50E+04
Anthracene	120-12-7	178.20	4.28E+00	4.54E+00	5.50E+01	5.50E+02	1.70E+04	5.50E+01	5.50E+03
Tetrachloroethylene	127-18-4	165.83	1.74E+03	2.58E+00	5.50E+02	5.50E+02	5.50E+03	5.50E+02	1.70E+03
Captan	133-06-2	300.60	7.29E-01	2.30E+00	1.70E+01	1.70E+01	5.50E+02	1.70E+01	5.50E+02
1H-Isoindole-1,3(2H)-dione, 2- (trichloromethyl)thio -	133-07-3	296.56	3.86E-04	3.63E+00	2.69E+01	1.38E+04	1.38E+04	2.69E+01	1.38E+04
Thioperoxydicarbonic diamide, tetramethyl-	137-26-8	240.40	8.00E-03	1.73E+00	1.70E+02	1.70E+02	1.70E+03	1.70E+02	5.50E+02
Ethyl acetate	141-78-6	88.11	1.40E+01	6.90E-01	5.50E+01	5.50E+01	5.50E+02	5.50E+01	1.70E+02
Trifluralin	1582-09-8	335.50	2.67E+00	5.34E+00	1.70E+02	1.70E+03	5.50E+03	1.70E+02	1.70E+03
Methomyl	16752-77-5	162.20	1.87E-08	6.00E-01	5.50E+02	5.50E+03	5.50E+03	5.50E+02	5.50E+02
2,3,7,8-TCDD (Dioxin)	1746-01-6	322.00	2.47E+00	6.91E+00	1.70E+02	5.50E+02	5.50E+04	1.70E+02	1.70E+04
Benomyl	17804-35-2	290.30	1.93E-09	2.30E+00	5.00E+00	1.70E+02	5.50E+03	5.00E+00	1.70E+03
Mirex	2385-85-5	545.55	1.30E-01	5.28E+00	1.70E+02	1.70E+02	5.50E+04	1.70E+02	5.50E+04
Pronamide	23950-58-5	256.13	5.44E-01	3.51E+00	1.37E+03	9.79E+02	1.80E+02	1.37E+03	1.93E+03
Acephate	30560-19-1	183.20	5.06E-11	-1.00E+00	7.55E+00	1.26E+03	5.28E+01	7.55E+00	5.28E+01
Aldrin	309-00-2	364.93	1.09E+01	3.01E+00	5.00E+00	1.70E+04	5.50E+04	5.00E+00	1.70E+04
Formaldehyde	50-00-0	30.03	3.20E-02	3.50E-01	5.00E+00	5.50E+01	1.70E+02	5.00E+00	5.50E+01
Cypermethrin	52315-07-8	416.30	1.95E-05	6.60E+00	1.04E+01	1.20E+02	1.25E+03	1.04E+01	1.25E+03
N-Nitrosodiethylamine	55-18-5	102.14	1.75E-01	4.80E-01	5.00E+00	1.70E+01	5.50E+03	5.00E+00	1.70E+03
Carbon tetrachloride	56-23-5	153.82	3.25E+03	2.64E+00	1.70E+04	1.70E+03	1.70E+04	1.70E+04	5.50E+03
gamma-Hexachloro-cyclohexane	58-89-9	290.85	3.42E-01	3.70E+00	1.70E+02	1.70E+04	5.50E+04	1.70E+02	1.70E+04
Heptachlor	76-44-8	373.40	2.17E+01	5.27E+00	5.50E+01	5.50E+02	5.50E+03	5.50E+01	1.70E+03
Hexachlorocyclopentadiene	77-47-4	272.77	2.20E+03	5.11E+00	4.95E+00	8.65E+01	1.68E+03	4.95E+00	4.20E+02
1,1,2,2-Tetrachloroethane	79-34-5	167.85	2.57E+01	2.39E+00	1.70E+04	1.70E+03	1.70E+04	1.70E+04	5.50E+03
Hexachlorobutadiene	87-68-3	260.76	2.41E+03	4.70E+00	1.72E+04	1.70E+03	1.70E+03	1.72E+04	1.70E+03
Benzene, hexabromo-	87-82-1	551.49	2.85E+00	6.07E+00	2.24E+04	1.44E+03	5.76E+03	2.24E+04	1.44E+03

2 Main Exposure Pathways

It is mostly the exposed vegetal products that dominate the intake in most cases, except for a few substances, for which fish, milk and meat are significant (**Fig. S1**).

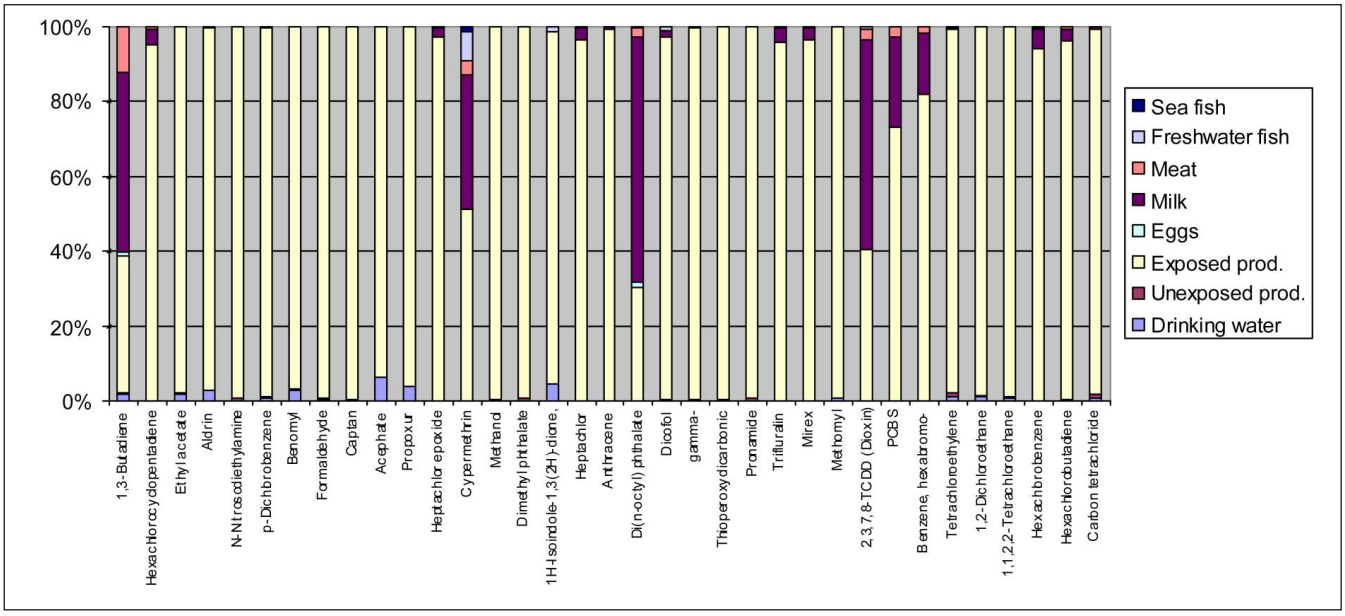

Fig. S1: Distribution of the intake by ingestion for the different intake pathways. Chemical are ordered by increasing ingestion intake fraction

3 Average Intake Fractions for Different Continents

Taking the European continent as a reference, **Fig. S2** plots the average reduction in intake fraction for other continents. For the inhalation route of exposure, the reduction in iF amounts up to a factor 5 and is indeed strongly correlated to the relative reduction in population density (Fig. S2a: R^2=0.99). For ingestion, reduction in iF of up to a factor 10 on average is strongly correlated to the total agriculture production per km² (Fig. S2b: R^2=0.97).

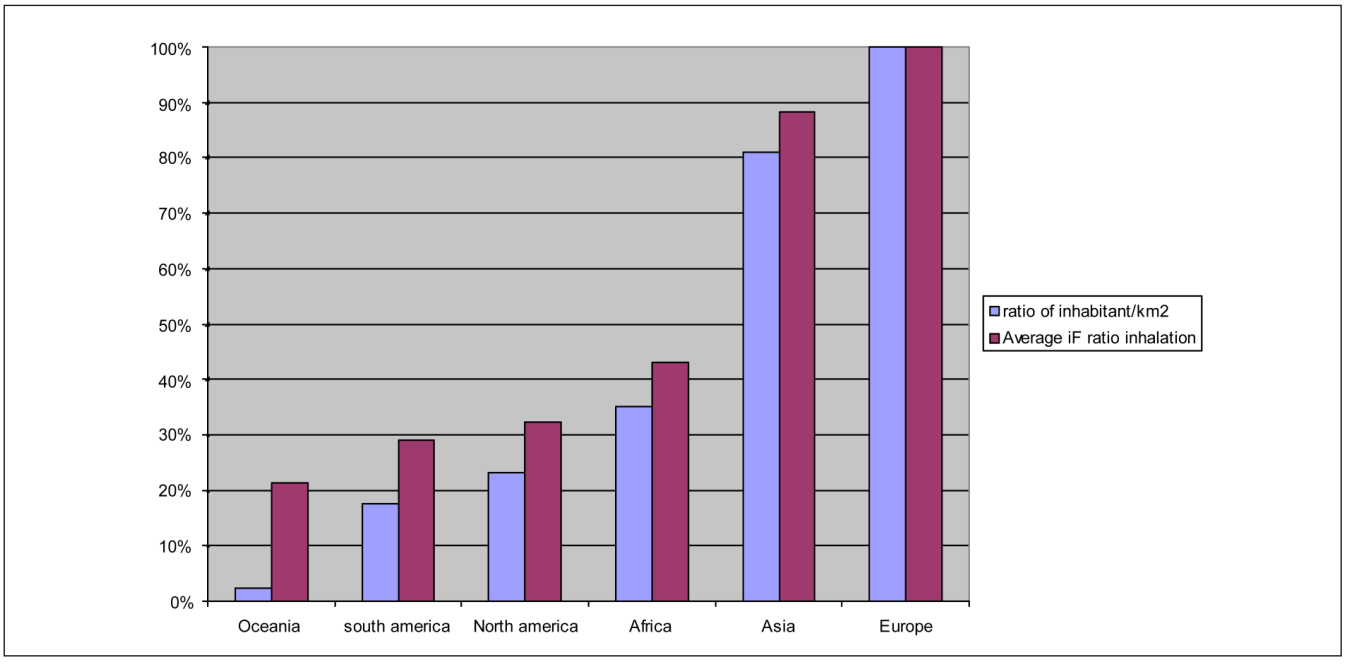

Fig. S2a: Average continental intake fraction by inhalation compared to population density. All data are normalized to Europe (100%)

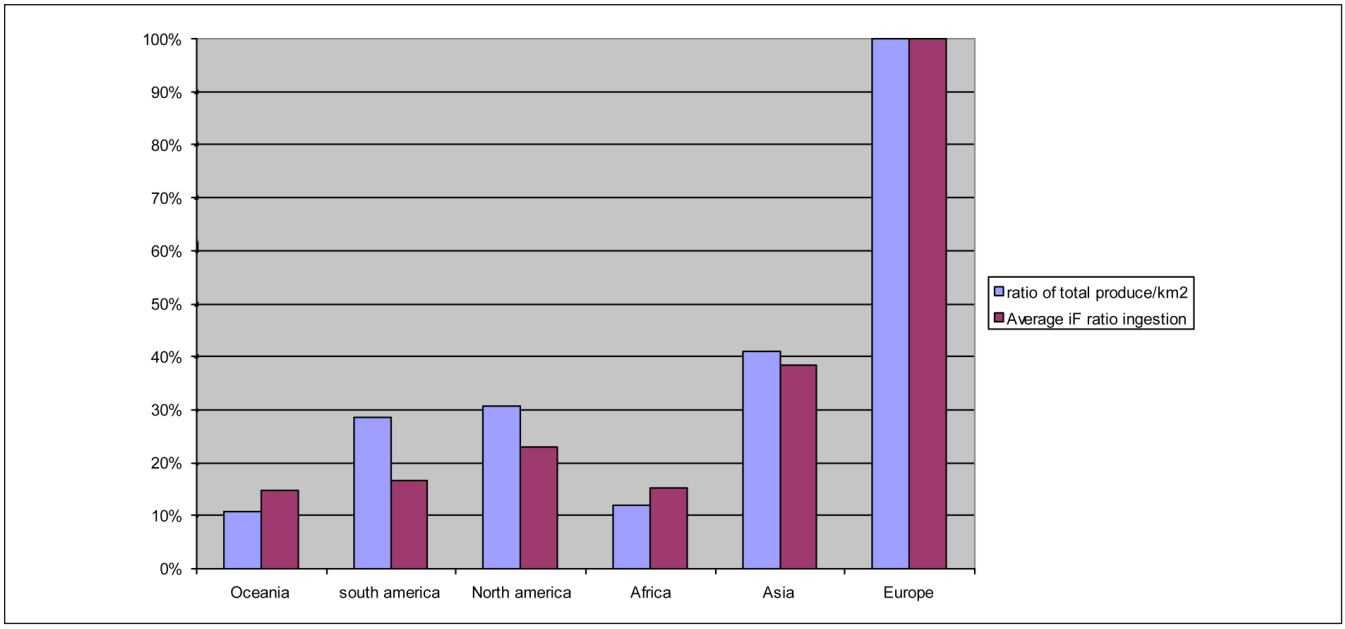

Fig. S2b: Average continental intake fraction by ingestion compared to the total agriculture production per km². All data are normalized to Europe (100%)

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