

PDF hosted at the Radboud Repository of the Radboud University Nijmegen

The following full text is an author's version which may differ from the publisher's version.

For additional information about this publication click this link.

<http://hdl.handle.net/2066/36405>

Please be advised that this information was generated on 2021-02-25 and may be subject to change.



Front page for deliverables

<i>Project no.</i>	003956
<i>Project acronym</i>	NOMIRACLE
<i>Project title</i>	Novel Methods for Integrated Risk Assessment of Cumulative Stressors in Europe
<i>Instrument</i>	IP
<i>Thematic Priority</i>	1.1.6.3, 'Global Change and Ecosystems' Topic VII.1.1.a, 'Development of risk assessment methodologies'

Deliverable reference number and title:

D.2.4.7 Report on the estimation of the spatial variance in environmental concentrations without making use of a spatially resolved model

Due date of deliverable: May 1, 2007 *Actual submission date:* 09.08.07

Start date of project: November 01, 2004 *Duration:* 5 years

Organisation name of lead contractor for this milestone: RU

Revision [draft, 1, 2, ...]: final

Project co-funded by the European Commission within the Sixth Framework Programme (2002-2006)		
Dissemination Level		
PU	Public	PU
PP	Restricted to other programme participants (including the Commission Services)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	

Authors and their organization: Anne Hollander; Mara Hauck; Mark A.J. Huijbregts; Ad M.J. Ragas; Dik Van de Meent, Radboud University Nijmegen				
Deliverable no: D.2.4.5	Nature: R	Dissemination level: PP	Date of delivery: 07.08.07	
Status: Final			Date of publishing:	
Reviewed by (name and period): Alberto Pistocchi (February 2007)				

Table of Contents

Table of Contents	3
Summary	4
1 Introduction.....	5
2 Methods	8
2.1 Model setup.....	8
2.1 Chemical properties	10
2.2 Model output and analysis	11
3 Results.....	13
3.1 Air concentrations	13
3.2 Soil concentrations.....	15
3.3 Water concentrations	17
4 Discussion.....	19
References.....	26

Summary

The environmental fate of chemicals is controlled by two main factors: the properties of the chemical and the characteristics of the environment. The classical box approach in multimedia mass balance models assumes that chemical properties largely determine the fate of substances in the environment, neglecting the potential role of the local environmental conditions. In this study, the relative influence of substance properties and of environmental characteristics on the variation in environmental concentrations of chemicals was estimated for nine different emission/receiving compartment scenarios and on three different spatial scales (50x50 km, 100x100 km, 250x250 km). Unit emissions were assumed. Depending on emission scenario, compartment and model resolutions, the range in predicted environmental concentrations spreads from 2 up to 10 orders of magnitude. The assumption that the variation in the fate of chemicals in the environment is, beside emission intensity, mainly dependent on substance properties, is correct. But from our study it appears that, regarding local emissions and concentrations, in some scenarios, environmental differences should not be ignored when estimating concentration variations. Particularly in the soil and water compartments, the spatial variation in environmental characteristics can have a substantial impact on the concentration variations.

1 Introduction

The environmental fate of chemicals is controlled by two main factors: the properties of the chemical (e.g. vapor pressure, water solubility, degradation half-lives) and the characteristics of the environment (e.g. temperature, soil organic carbon content, OH-radical concentration). At equal release rates, concentrations may diverge for different chemicals and for different environments (1). Multimedia mass balance models (box models) are often used to predict the environmental fate of chemicals for regulatory purposes, e.g. in comparative risk assessments. The classical box model (2-3) approach implicitly assumes that chemical properties largely determine the fate of substances in the environment. In these models, chemical-to-chemical differences in model outcomes are met by the requirement of physico-chemical input data of the studied compounds. However, the possible role of the environmental characteristics are disregarded by limiting the analyses to modeling ‘typical’ or ‘average’ conditions only. By focusing strongly on the chemical properties, the possible importance of the environmental characteristics may be underestimated. Does it suffice for environmental risk assessments of chemicals to take account of the substance that is released, neglecting the potential role of the local environmental conditions? How well do we know the relative contribution of substance properties and environmental characteristics to the fate of chemicals?

The traditional way to account for spatial variation in environmental characteristics is the use of spatially resolved models. In the last few years, many of such spatial multimedia models have been developed (e.g. IMPACT-2002 (4); Evn-BETR (5); BETR-Global (6); G-CIEMS (7)). Unfortunately, spatially explicit model outputs can only be derived at the rate of an increased model complexity and input data demand, whereas ‘low-complexity’ and ‘easy-to-use’ models are still desirable, particularly for screening assessments of large numbers of chemicals.

Both spatial and non-spatial multimedia fate models thus have their specific advantages and drawbacks, and the appropriate model to use depends on the specific purpose of the application under consideration (4, 8). Many modelers struggle with the question of how to handle spatial environmental differences in their modeling studies. However, to determine whether a spatially resolved model is necessary, one should have an idea of the spatial variation of the input data and of the influence of that spatial variation on the model results. Several authors have explored the influence of the spatial variation in single landscape characteristics on the fate of chemicals. Sweetman et al. (9), for example, examined the role of soil organic carbon in the global cycling of POPs, and Wania and McLachlan (10) estimated the influence of forest characteristics on the overall fate of semi-volatile organic chemicals. Furthermore, sensitivity analyses have been performed on both physico-chemical and landscape characteristics in multimedia mass balance models (11, 12). However, none of these studies have systematically addressed the relative importance of the spatial variation in the total set of environmental characteristics and the differences in substance properties. Furthermore, the spatial resolution of a model influences the spatial variation in both input data and modeling results. In the modeling studies mentioned above, only one spatial scale was considered. The influence of the choice of the modeling scale on the predicted spatial variation in concentrations has thus not been evaluated yet.

The objective of this study is to systematically estimate the influence of the spatial variation in environmental characteristics in Europe on the variation in environmental concentrations of chemicals and to compare it with the variation caused by differences in chemical characteristics. The study was performed for 200 organic chemicals, covering the domain of plausible chemical properties, and for a set of 19 environmental parameters. Calculations were performed for three emission scenarios, i.e. towards the air, water and soil compartments, respectively. The results were analyzed for the receiving compartments air,

water and soil. Emission intensities were assumed to be invariant for the purpose of this study.

To check the influence of spatial resolution on the variability in concentrations, calculations

were carried out for three spatial scales, i.e. 50x50 km, 100x100 km and 250x250 km.

2 Methods

2.1 Model setup

Concentration predictions were performed with a nested multimedia mass balance model, of which the local scale was parameterized successively for the different regions of Europe, each with its own characteristic set of environmental conditions. A nested model was used in order to include the exchange of chemicals between the local scale cell and the surrounding areas. To this end, a local scale cell was implemented in the generic level III multimedia mass balance model BasinBox (13), which is based on SimpleBox 3.0 (14). The default model settings were applied, except that the upstream, midstream and downstream areas were aggregated. The local scale cell consisted of an air, soil, water, sediment and vegetation compartment. Figure 1 presents a schematic setup of the model study.

To parameterize the local scale cells, a dataset was created containing realistic property combinations of 19 environmental parameters in Europe. The environmental parameters taken into account and their references are given in Table 1, and the histograms of the different parameter values are given in the Supporting Information. These parameters correspond to the input requirements for the spatially distributed GIS-based model MAPPE (15). They were collected from global or continental scale data sets available from different sources, or developed based on previous analyses (16, 17). Details about individual parameters were discussed in (17).

Maps with single environmental properties were rasterized into grid cells within which the environmental property values were averaged. In order to study the influence of grid cell size on the predicted concentrations, grids of three different sizes were used: 50x50 km, 100x100 km, 250x250 km. From the raster maps, a property-combination table was created, containing all environmental properties for each grid cell, and environmental concentrations were calculated for each cell with its own characteristic set of environmental properties. For

some parameters, the values were unknown for one or more cells. In those cases, the average parameter value of the other cells was applied. If more than one third of the parameter values for a cell was unknown, this cell was not taken into account in the calculations, since with so many averaged characteristics, the regional properties become too uncertain and are likely to represent an unrealistic situation. This was the case for 5% of the 250x250 km cells, 15% of the 100x100 km cells, and for 29% of the 50x50 km cells. With the exclusion of these cells, 172, 910 and 3209 unique environmental property combinations were found for European regions with an area of 250x250, 100x100 and 50x50 km, respectively.

Results were obtained for each individual local cell, for emissions to respectively air, soil and water with a unit emission, and relative concentrations were compared. Emission was assumed to take place to the local cell only.

Table 1. Environmental parameters taken into account in the calculations.

Environmental parameter	Unit	Reference
Surface water depth	m	(16)
Soil erosion intensity	mm.y ⁻¹	(30)
Evapo-transpiration intensity	in mm.y ⁻¹	(17)
Area fraction of pasture soil	-	(31)
Area fraction of cropland soil	-	(31)
Area fraction of natural soil	-	(31)
Area fraction of surface water	-	(16)
Atmospheric mixing height	m	(32)
Soil organic carbon content	-	(33)
OH-concentration in air	molec.cm ⁻³	(32)
Precipitation intensity*	mm.y ⁻¹	(17, 34)
Soil moisture content	-	(17)
Suspended matter concentration in water	mg.l ⁻¹	(17)
Atmospheric temperature	°C	(34)
Wind velocity at 10m	m.s ⁻¹	(34)
Aerosol deposition velocity	m.s ⁻¹	(17)
Leaf area index	m ² .m ⁻²	(17)
Aerosol surface**	m ² .m ⁻³	(36)
Water inflow in cell	in m ³ .s ⁻¹	(16)

* A conversion was made from rainfall and snowfall intensity data to one value for precipitation. For this

parameter, 10 mm of snowfall was set equivalent to 1 mm of rain. ** The aerosol surface was estimated from

PM10-concentrations. The average European PM10-concentration was set equal to the average aerosol surface default in BasinBox ($1.5 \cdot 10^{-4} \text{ m}^2 \cdot \text{m}^{-3}$; (37)) and the aerosol surfaces in the different grid cells were made proportional to that.

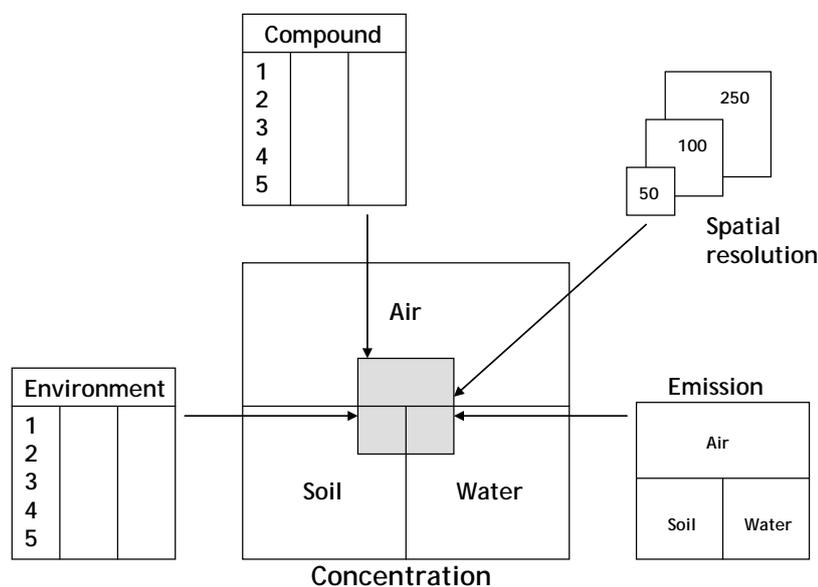


Figure 1. Schematic setup of the model study showing the different types of input parameters taken into account in the study. The gray block represents the local cell to which emissions take place and in which environmental concentrations are estimated.

2.1 Chemical properties

Calculations were performed for a group of 200 organic chemicals, showing a wide range in chemical partitioning properties and half-lives. The chemicals were randomly taken from a dataset of 356 chemicals of Huijbregts et al. (18). Due to the technical limitations of the spreadsheet model, we were not able to use this total dataset. The physico-chemical properties of the chemicals used in this study are given in Appendix 1 of the Supporting Information, and their air-water and soil-water partitioning properties are summarized in Figure 2.

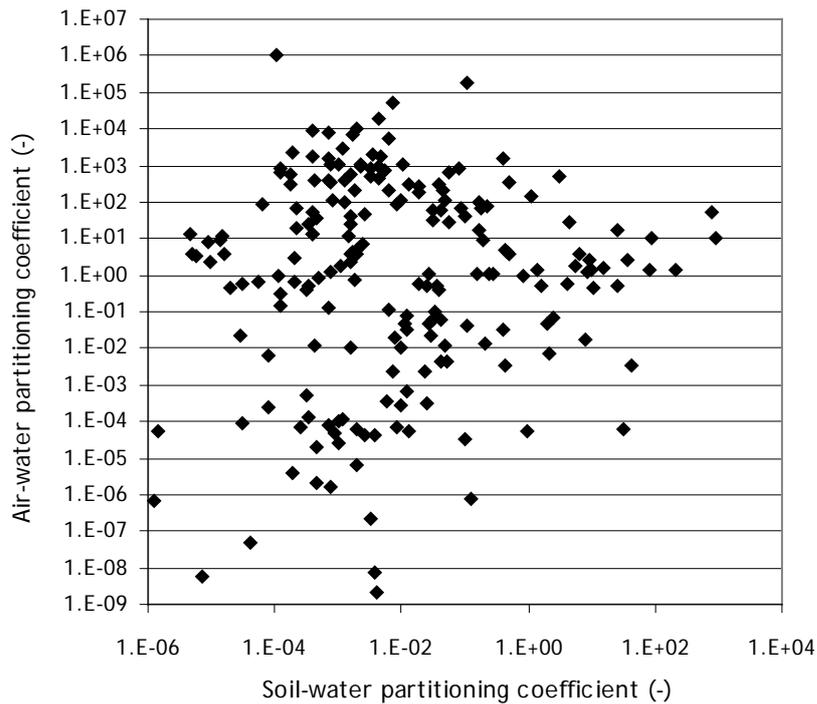


Figure 2: Air-water and soil-water partitioning properties of the 200 organic compounds used for the model calculations.

2.2 Model output and analysis

Environmental concentrations were predicted in three compartments: air, water and soil. For each scenario, concentration ranges (CR) between the different compounds, defined as the ratio between the 95th and 5th concentration percentiles, were calculated for each grid cell separately. The concentration ranges between the grid cells were obtained for each individual compound. Additionally, the concentration range of the total set of concentrations was calculated per scenario (CR_{total}). From this, the average concentration ranges caused by compound differences ($CR_{chemical}$) and the average concentration ranges caused by environmental differences (CR_{area}) were derived. Their 95% confidence levels were also calculated. For each scenario, the CRs were plotted into separate graphs for the 50x50, 100x100 and 250x250 km spatial scales. From

these graphs, the relative influences of compound variability and environmental variability on the total variation in environmental concentrations were analyzed.

3 Results

3.1 Air concentrations

Air compartment

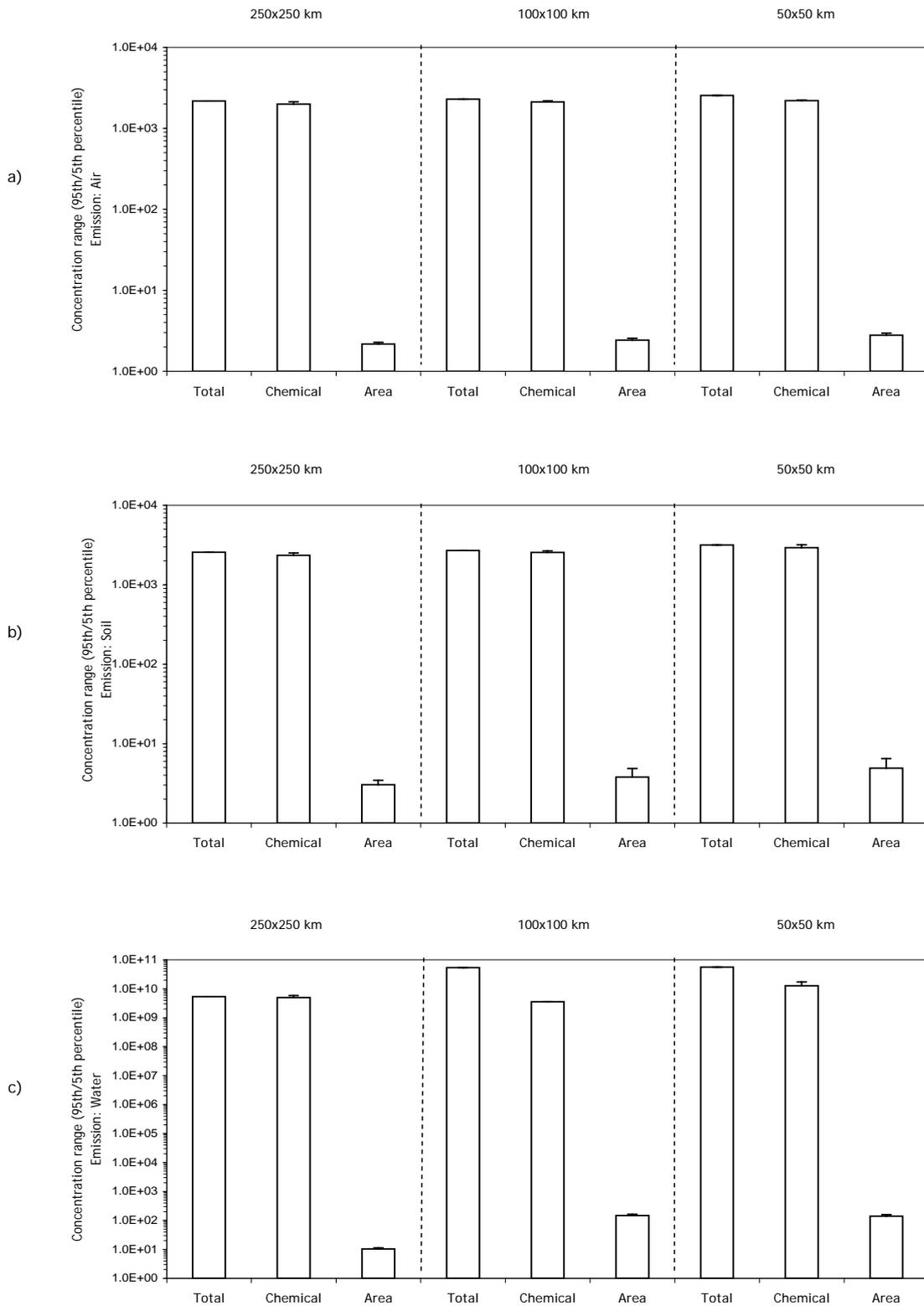


Figure 3. The average concentration ranges (CR) and their 95% confidence levels for the environmental concentrations in the air compartment in Europe, for three different emission scenarios. Concentration ranges are given for the total concentration variation (CR_{total}), the concentration variation caused by the variation in substance properties ($CR_{chemical}$) and the concentration variation caused by differences in environmental characteristics (CR_{area}). 1a) Emission compartment: air, 1b) Emission compartment: soil, 1c) Emission compartment: water.

Figure 3 shows that the CR_{total} ranges from about 3 to 9 orders of magnitude in the air compartment, being smallest if emissions occur to air and largest if emissions occur to water. If emissions take place to air or soil, the CR_{total} hardly varies with model resolution. If emissions occur to water, the CR_{total} increases with a factor 10 between the 250x250 km and the 50x50 km scale, which is mainly caused by an increasing CR_{area} . In all scenarios, on all spatial scales, the CR_{total} is mainly caused by the variation in substance properties. In the air and the soil emission scenario, the $CR_{chemical}$ accounts for more than 98% of the CR_{total} . In the water emission scenario, the variation in concentrations caused by differences between compounds is even several orders of magnitude larger than the variation caused by spatial environmental characteristics.

The 95% confidence levels are relatively high for the CR_{area} in the emission scenario towards soil. This implies that for a small number of compounds, the variation between the different grid cells is relatively large. This phenomenon occurs strongest on the 50x50 km model resolution.

3.2 Soil concentrations

Soil compartment

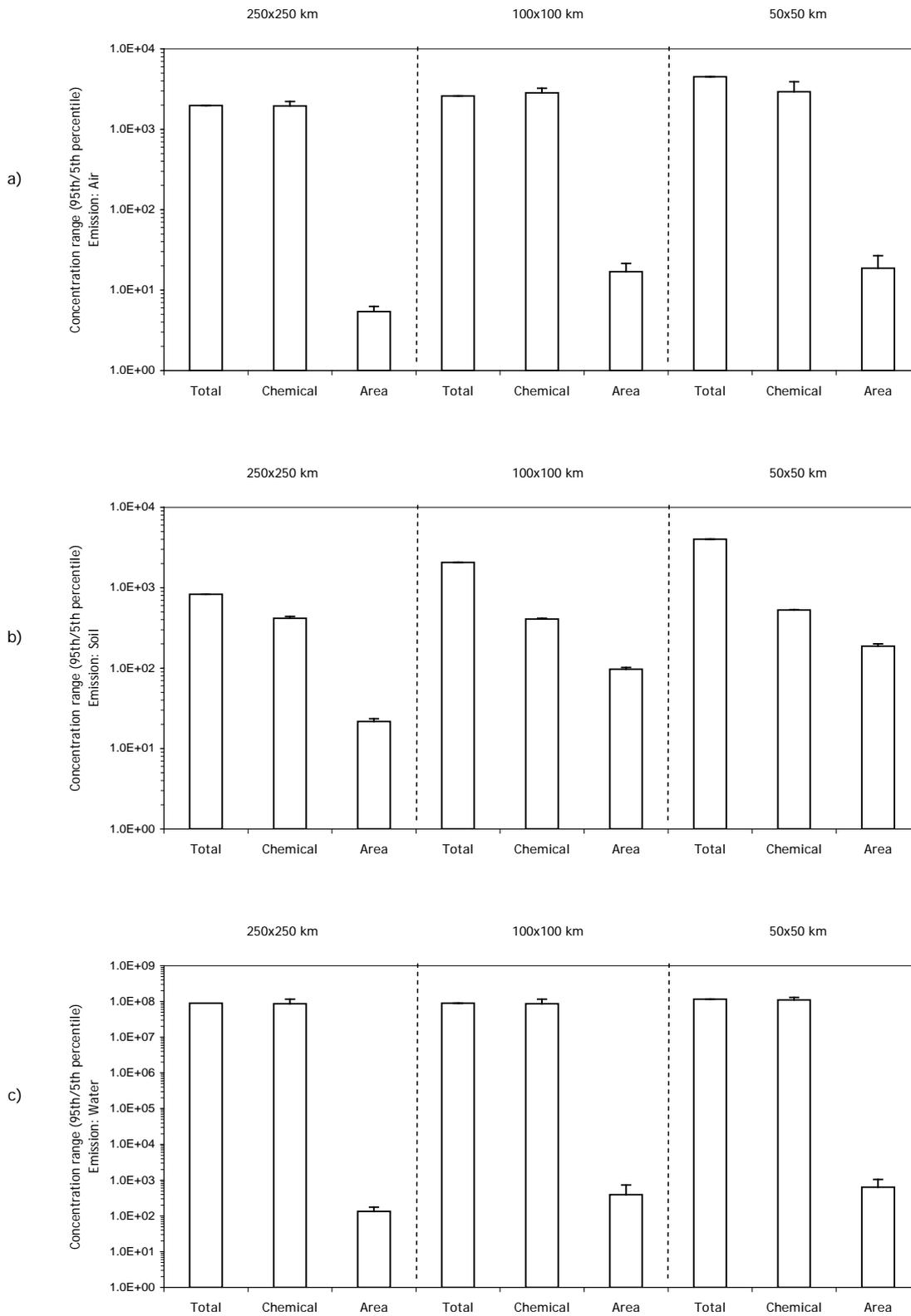


Figure 4. The average concentration ranges (CR) and their 95% confidence levels for the environmental concentrations in the soil compartment in Europe, for three different emission scenarios. Concentration ranges are given for the total concentration variation (CR_{total}), the concentration variation caused by the variation in substance properties ($CR_{chemical}$) and the concentration variation caused by differences in environmental characteristics (CR_{area}). 1a) Emission compartment: air, 1b) Emission compartment: soil, 1c) Emission compartment: water.

Figure 4 shows that the CR_{total} ranges from about 800 to $1 \cdot 10^8$ in the soil compartment, being smallest if emissions occur towards soil and largest if emissions occur to water. If emissions take place to air, the CR_{total} increases with model resolution from 2000 on the 250x250 km scale, to 3000 on the 50x50 km scale. In this emission scenario, the $CR_{chemical}$ is about a factor 100-300 larger than the CR_{area} . The more detailed the model resolution, the stronger the relative influence of environmental characteristics. This is also the case, even more clearly, if emissions occur to soil. In this compartment $CR_{chemical}$ varies between a factor 400 to 500 between the largest and the smallest model scale, while the CR_{area} shows an increase from about 20 at the 250x250 km scale to 200 at the 50x50 km scale. If emissions occur to water, the $CR_{chemical}$ also dominates the CR_{total} ; $CR_{chemical}$ in this scenario is always at least five orders of magnitude larger than CR_{area} . However, in this emission scenario, environmental conditions also result in concentration differences of more than a factor 600 on the 50x50 km scale. The 95% confidence levels are relatively high for the CV_{area} on the 100x100 km and 50x50 km scales if emissions occur to water

3.3 Water concentrations

Water compartment

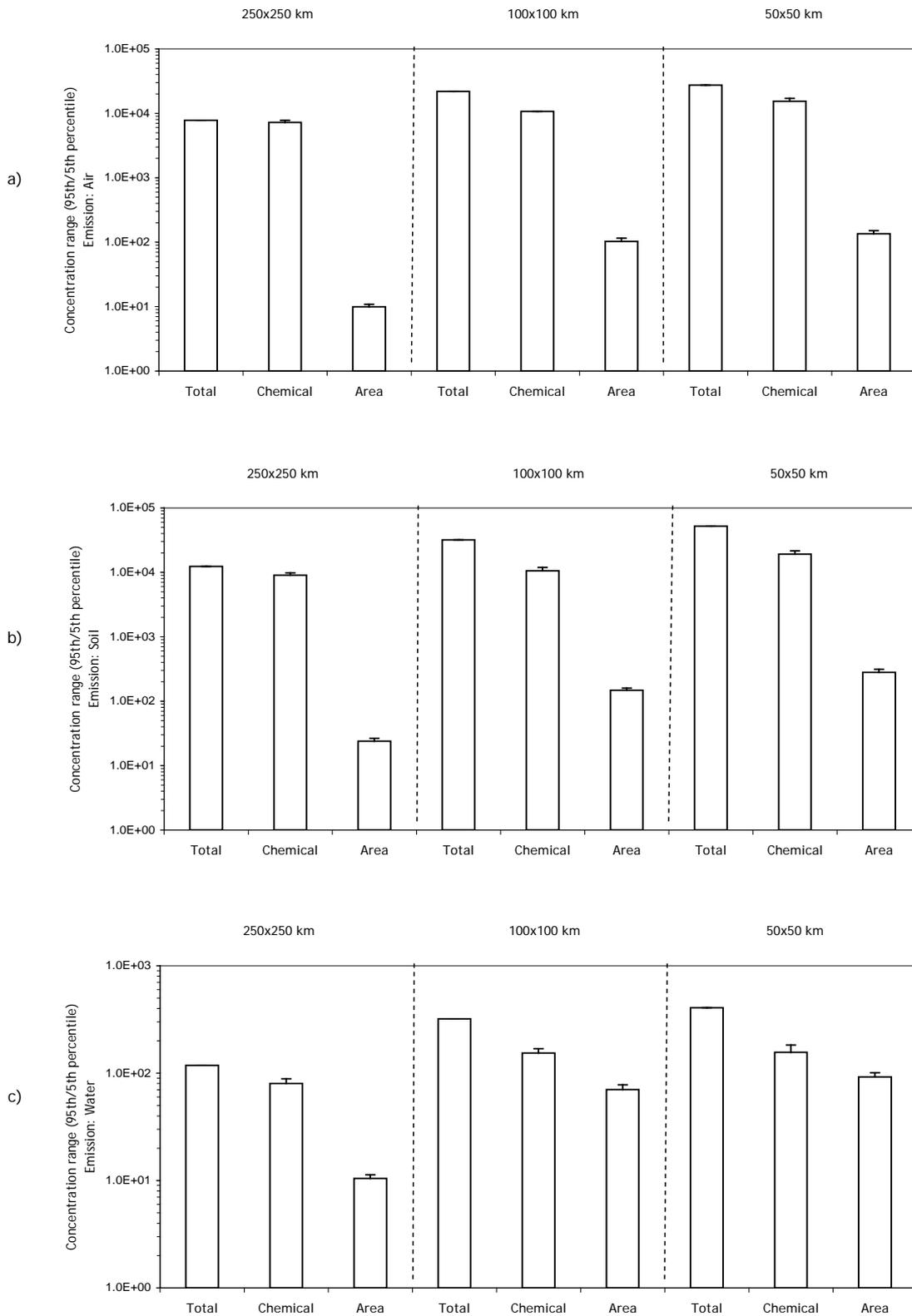


Figure 5. The average concentration ranges (CR) and their 95% confidence levels for the environmental concentrations in the water compartment in Europe, for three different emission scenarios. Concentration ranges are given for the total concentration variation (CR_{total}), the concentration variation caused by the variation in substance properties ($CR_{chemical}$) and the concentration variation caused by differences in environmental characteristics (CR_{area}). 1a) Emission compartment: air, 1b) Emission compartment: soil, 1c) Emission compartment: water.

Figure 5 shows that the CR_{total} ranges from about 120 to $5 \cdot 10^4$ in the water compartment, being smallest if emissions occur to water and largest if emissions occur to soil. If emissions take place to air, the CR_{total} increases with a factor 3.5 between the 250x250 km and 50x50 km scale, from $8 \cdot 10^3$ to $3 \cdot 10^4$. In this scenario, CR_{area} has at maximum (on the 50x50 km scale) a value of 135. The $CR_{chemical}$ is always at least 100 times larger than the CR_{area} . In case of emissions to soil, with respect to the other emission scenarios, the CR_{area} is relatively large compared to $CR_{chemical}$, and on a more detailed resolution, this relative importance increases. The CR_{total} increases with a factor of 2.6 from the 250x250 km scale to the 100x100 km scale, while on the 50x50 km scale, the CR_{total} is a factor of 1.6 larger than on the 100x100 km scale. CR_{area} almost fully accounts for this increase, since this factor increases with a factor 11 between the 250x250 km and 50x50 km scales. In this scenario, CR_{area} is at maximum a factor 280. For water emissions, the $CR_{chemical}$ is respectively 7.7, 2.2, and 1.7 times larger on the 250x250 km, 100x100 km and 50x50 km scales compared to the CR_{area} , so the relative influence of CR_{area} on the total variation in concentrations is large with respect to the other scenarios. Values for the CR_{total} range from 118 to 400.

4 Discussion

Environmental concentration variations

The results of this study show the relative influence of substance properties and of environmental characteristics on the variation in environmental concentrations of chemicals for different scenarios and on different spatial scales. In the present study, emissions were assumed to take place to the local cell only, and only local environmental concentrations were considered. The influence of the local emissions on the area outside the cell, and the influence of regional emissions on the local cell have not been studied. Furthermore, unit emissions were assumed. Within the framework of the current modeling setup, only concentration variations in the local cell were considered.

In general, concentration differences are predicted to be larger if a smaller local scale is applied in the model calculations, and the relative influence of environmental characteristics on the total variation increases on a more detailed spatial scale. The environmental concentration variations predicted on a 50x50 km scale can be up to a factor 5 larger than the spatial variations on a 250x250 km scale. The relative importance of the variation in substance properties compared to the variation in environmental characteristics generally increases with increasing model resolution. At maximum, the concentration range between the different areas differs a factor 14 between the 250x250 and 50x50 km scale. This can be explained by the fact that at a coarser spatial resolution, the spatial variation in environmental characteristics is partly lost due to the averaging of environmental parameters over a larger area. From these results, it can be concluded that the model resolution can have an impact on the environmental concentration variations that are derived from box model calculations, particularly in the soil and water compartments. Hertwich and McKone (20) did not find any effect of (pollutant-specific) spatial scales on the potential dose of chemicals. On the contrary, other studies from

related branches of environmental modeling show that the model resolution significantly impacts the variability in model output. Zoras et al. (21) concluded that in meteorological models, the model resolution significantly impacts the variability of model output. In hydrological modeling, several studies have been carried out exploring the appropriate input data resolution of the models (22-24). For crop growth monitoring systems, a grid size of 50x50 km was mentioned as an appropriate resolution (25), while in ocean modeling, a resolution of 0.5° or 0.25° is critically important for a realistic simulation of saline water flows from the Atlantic to the Arctic regions (26). From our study, it can be concluded that for the air compartment, the differences in the spatial variations are negligible between the 250x250 km and 50x50 km scales when unit emissions are assumed. This means that for the current model setting, little additional information is derived for air from models with a resolution finer than 250x250 km. In the soil and water compartments, the spatial variation increases between the 100x100 and 50x50 km scales, implying that the choice of model resolution can influence the predictions for these compartments in multimedia mass balance models.

It is also found that, except in the air compartment, the CR_{area} is always larger in the primary compartments, to which the emission occurs, meaning that environmental characteristics are more dominant in the primary compartments. Transport processes from the emission compartment towards the other compartments are mainly driven by the physical-chemical properties of the compound, and therefore concentration differences in the secondary and tertiary compartments are mainly caused by substance differences. Only when air is the primary compartment, the environmental characteristics are less influential due to the mobility of this medium and its rapid mixing. Spatial environmental differences are being undone in that case.

Air concentrations

In all scenarios in which concentrations in air were considered, substance properties account almost fully for the variation in environmental concentrations. Predicted concentration ranges in the air are largest if emissions occur to water, due to large differences in the volatilization rates of the different compounds. The absolute concentrations in the air in this scenario are low to extremely low, implying that only a very small fraction of the emitted substance leaves the water compartment. Since volatilization rates are also partly determined by environmental circumstances, like wind speed, air temperature and suspended matter concentrations, this also results in a somewhat larger impact of environmental characteristics on the total concentration variation than in the air emission scenario.

Another phenomenon in the air compartment is that the 95% confidence levels are relatively high for the CR_{area} in the soil emission scenario, particularly on the 50x50 km scale. This means that for a small number of compounds, the concentration variation in the air between the different grid cells is relatively large. This is the case for those chemicals for which deposition and/or revolatilization are relatively important transport processes. The spatial variation in deposition is large, for example, for chemicals with a low Henry's law constant, due to differences in precipitation intensity throughout Europe. These outcomes are in accordance with those of Hertwich and McKone (20), who found that the characteristic travel distance of these chemicals is predicted substantially different in model situations with and without rain. One should take into account that continuous rain is assumed in our level III model study, which may cause an overestimation of the substance removal from the atmosphere through wet deposition (27).

Soil concentrations

In the soil compartment, environmental characteristics play a more important role in determining chemical fate with respect to substance properties than in the air compartment,

particularly if emissions occur to soil. In that case, the variation in environmental characteristics such as the organic carbon content of soils, which can be the main factor according to Hertwich et al. (11), soil moisture content, and erosion intensity, notably influences the variation in environmental concentrations. Due to the relatively large importance of the variation in environmental characteristics, total concentration variations increase with increasing model resolution in this scenario. Total concentration ranges in the soil compartment are largest if emissions occur to water. In that scenario, processes in the air, water and soil compartment play a role, since the only transport route of chemicals from water to soil is via the air. This means that almost all physico-chemical properties and environmental characteristics are involved in the fate calculations, resulting in a wide spread of predicted concentrations. Moreover, from the air concentration scenario with emissions to water had already become clear that the different compounds show a wide range in volatilization intensities from water, which is also reflected in the predicted soil concentrations if emissions go to water.

Water concentrations

In the water compartment, both substance properties and environmental parameters play a role, with the largest relative influence of the environmental characteristics in the water emission scenario. In this scenario, a large fraction of the emitted substance stays in the water compartment, which means that the fate of chemicals is dominated by the emission-intensity. At unit emissions, no large differences in concentrations appear between different compounds. In this scenario, the combination of the parameters ‘area fraction of water’, ‘water depth’ and ‘water inflow into the grid cell’ substantially determines the spatial variation in environmental concentrations. These three parameters account for the retention time of water in a cell, and their spatial variation on the 250x250 km is much smaller than on the 100x100 and 50x50 km scales, due to the averaging of these parameters. These results, indicating that differences in

environmental characteristics may have a substantial impact on the water concentrations, is in line with the one drawn by Pennington et al. (4), who argued that when predicting concentrations with a non-spatial multimedia fate model, it is necessary to take into account spatial differences in residence times in the water compartment, such as large lakes. When emissions occur to soil, the main transport route of chemicals to water is by runoff. In this process, both differences in substance properties (soil and pore water concentrations) and environmental characteristics (runoff intensities, soil and pore water concentrations, water characteristics) are of influence, resulting in relatively large concentration ranges. The transport route from soil to water via volatilization and atmospheric deposition is of marginal importance.

Model study setup

For this study, we created a European dataset containing as much environmental characteristics as were available at the time, but unfortunately, it was not possible to get spatial information on all environmental parameters that are input in multimedia fate models. Parameter values that were lacking were, for example, the porosity of soils and the fraction of soil water leaching to deeper ground water layers. However, according to Hertwich et al. (11), the parameters that are considered to be the most influential on the predicted concentrations, which are the organic carbon content of soils, the fraction suspended solids in surface water and the fraction of land area in a grid cell, were readily taken up in our dataset.

It is worth stressing that our study only refers to the variability in chemical and environmental properties; in many modeling applications, the influence of the spatial distribution and the absolute intensity of emissions can outweigh the influence of both environmental characteristics and substance properties on the concentration variations (4, 28).

In most multimedia fate models, it is assumed that chemical properties primarily account for the variation in environmental concentrations. This assumption was already

nuanced by Webster et al. (29), who concluded that the quality of results obtained from regional environmental fate models can be improved by the use of region-specific landscape parameters. Hertwich et al. (11) performed a sensitivity study on the variance in the potential dose to physico-chemical properties, exposure parameters, and environmental input parameters. From that study it was concluded that for most compounds, physico-chemical properties or exposure parameters largely account for the concentration variance. Consistently with the findings of Hertwich et al. (11), Fenner et al. (29a) concluded that the ranking of chemicals based on their overall persistency (P_{ov}) and long range transport potential (LRTP) is largely determined by the chemical properties. For compounds that are mainly present in the air compartment, our results are most similar to the ones of Hertwich et al. (11). For compounds that reside in the soil and water compartments, the present study shows that also environmental parameters can play a substantial role in the total concentration range, although chemical properties still account for the largest part of the concentration variation. In contrast to Hertwich et al., we did not consider each of the input parameters individually, but looked at the total set of environmental parameters in comparison with the total set of physico-chemical input parameters. Single environmental properties may have a relatively small impact on the variation in environmental concentrations, but from our study it appears that the complete set of environmental properties in some cases can have a substantial impact on the variation in environmental concentrations.

Implications for multimedia fate model studies

In this paper, a systematic insight was obtained in the relative influence of environmental characteristics and substance properties on the concentration variations of substances. The results of this study can provide information for which emission-receiving compartment scenarios, incorporating spatial differences in environmental parameters can be important. Depending on emission scenario, compartment and model resolutions, the range in predicted

environmental concentrations spreads from 2 up to 10 orders of magnitude. The assumption that the variation in the fate of chemicals in the environment is, beside emission intensity, mainly dependent on substance properties, is correct. But from our study it appears that, regarding local emissions and concentrations, in some scenarios, environmental differences should not be ignored when estimating concentration variations. Particularly in the soil and water compartments, the spatial variation in environmental characteristics can have a substantial impact on the concentration variations. In the air compartment, the influence of environmental characteristics may be ignored when unit emissions are assumed.

The results of this study also indicate the influence of model resolution on the spatial differences in concentrations. The averaging of environmental parameters over a larger area results in a more even concentration distribution across Europe. In environmental fate studies, the choice of model resolution can therefore have a slight influence on the information that is derived from the calculations. Particularly for soil and water concentrations, a resolution of 50x50 km provides more information on the concentration variations compared to larger scales. When one is interested in environmental concentration ranges occurring in an area, one should carefully choose an appropriate model resolution.

Acknowledgements

Dr. Harrie Hendriks is acknowledged for his useful discussions.

References

- (1) Webster, E.; Mackay, D. *Defining uncertainty and variability in environmental fate models*; CEMC report no. 200301; Trent University; Peterborough, 2003.
- (2) Mackay, D.; Paterson, S. Calculating fugacity. *Environ. Sci. Technol.* 1981, 15, 1006-1014.
- (3) Mackay, D.; Joy, M.; Paterson, S. A quantitative water, air, sediment interaction (QWASI) fugacity model for describing the fate of chemicals in lakes. *Chemosphere* 1982, 12, 981-997.
- (4) Pennington, D.W.; Margni, M.; Ammann, C.; Jolliet, O. Multimedia fate and human intake modelling: Spatial versus nonspatial insights for chemical emissions in Western Europe. *Environ. Sci. Technol.* 2005, 39, 1119-1128.
- (5) Prevedouros, K.; MacLeod, M.; Jones, K.C.; Sweetman, A. Modelling the fate of persistent organic pollutants in Europe: parameterisation of a gridded distribution model. *Environ. Pollut.* 2004, 128, 251-261.
- (6) MacLeod, M.; Riley, W.J.; McKone, T.E. Assessing the influence of climate variability on atmospheric concentrations of polychlorinated biphenyls using a global-scale mass balance model (BETR-Global). *Environ. Sci. Technol.* 2005, 39, 6749-6756.
- (7) Suzuki, N.; Murasawa, K.; Sakurai, T.; Nansai, K.; Matsushashi, K.; Moriguchi, Y.; Tanabe, K.; Nakasugi, O.; Morita, M. Geo-referenced multimedia environmental fate model (G-CIEMS): Model formulation and comparison to the generic model and monitoring approaches. *Environ. Sci. Technol.* 2005, 38, 5682-5693.
- (8) Cahill, T.M.; Mackay, D. A high-resolution model for estimating the environmental fate of multi-species chemicals: application to malathion and pentachlorophenol. *Chemosphere* 2003, 53, 571-581.

- (9) Sweetman, A.J.; Dalla Valle, M.; Prevedouros, K.; Jones, K.C. The role of soil organic carbon in the global cycling of persistent organic pollutants (POPs): interpreting and modelling field data. *Chemosphere* 2005, 60, 959-972.
- (10) Wania, F.; McLachlan, M.S. Estimating the influence of forests on the overall fate of semivolatile organic compounds using a multimedia fate model. *Environ. Sci. Technol.* 2001, 35, 582-590.
- (11) Hertwich, E.G.; McKone, T.E.; Pease, W.S. Parameter uncertainty and variability in evaluative fate and exposure models. *Risk Anal.* 1999, 19, 1193-1204.
- (12) Maddalena, R.L.; McKone, T.E.; Hshieh, D.P.H.; Geng, S. Influential input classification in probabilistic multimedia models. *Stoch. Env. Res. Risk A.* 2001, 15, 1-17.
- (13) Hollander, A.; Huijbregts, M.A.J.; Ragas, A.; Van de Meent, D. BasinBox: a generic multimedia fate model for predicting the fate of chemicals in river catchments. *Hydrobiologia* 2006, 565, 21-38.
- (14) Den Hollander, H.A.; Van Eijkeren, J.C.H.; Van de Meent, D. *SimpleBox 3.0: Multimedia mass balance model for evaluating the fate of chemicals in the environment*. Report number 601200003; National Institute for Public Health and the Environment (RIVM); Bilthoven, 2004.
- (15) Pistocchi, A.; Pennington, D.W. *Continental Scale Mapping of Chemical Fate using Spatially Explicit Multimedia Models*. In: *Proceedings of the 1st Open International NoMiracle Workshop, Verbania - Intra, Italy June 8-9, 2006*; Report EUR 22625 EN; Office for Official Publications of the European Communities; Luxembourg, 2006.
- (16) Pistocchi, A.; Pennington, D.W. European hydraulic geometries for continental scale environmental modelling. *J. Hydrol.* 2006, 329, 553- 567.
- (17) Pistocchi, A.; Vizcaino Martinez, M.P.; Pennington, D.W. *Analysis of Landscape and Climate Parameters for Continental Scale Assessment of the Fate of Pollutants*; Report

EUR 22624 EN; Office for Official Publications of the European Communities;
Luxembourg, 2006.

- (18) Huijbregts et al., dataset.
- (19)
- (20) Hertwich, E.G.; McKone, T.E. Pollutant-specific scale of multimedia models and its implications for the potential dose. *Environ. Sci. Technol.* 2001, 35, 142-148.
- (21) Zoras, S.; Triantafyllou, A.G.; Hurly, P.J. Grid sensitivity analysis for the calibration of a prognostic meteorological model in complex terrain by a screening experiment. *Environ. Modell. Softw.* 2007, 22, 33-39.
- (22) Shrestha, R.; Tachikawa, Y.; Takara, K. Input data resolution analysis for distributed hydrological modeling. *J. Hydrol.* 2006, 319, 36-50.
- (23) Kavvas, M.L.; Chen, Z.Q.; Tan, L.; Soong, S.-T.; Terakawa, A.; Yoshitani, J.; Fukami, K. A regional-scale land surface parameterization based on areally-averaged hydrological conservation equations. *Hydrol. Sci. J.* 1998, 43, 611–631.
- (24) Koren, V.I.; Finnerty, B.D.; Schaake, J.C.; Smith, M.B.; Seo, D.-J.; Duan, Q.-Y. Scale dependencies of hydrologic models to spatial variability of precipitation. *J. Hydrol.* 1999, 217, 285–302.
- (25) De Wit, A.J.W.; Boogaard, H.L.; Van Diepen, C.A. Spatial resolution of precipitation and radiation: the effect on regional crop yield forecasts. *Agr. Forest Meteorol.* 2005, 135, 156-168.
- (26) Oka, A.; Hasumi, H. Effects of model resolution on salt transport through Northern high-latitude passages and Atlantic meridional overturning circulation. *Ocean Model.* 2006, 13, 126-147.
- (27) Jolliet, O.; Hauschild, M. Modeling the influence of intermittent rain events on long-term fate and transport of organic air pollutants. *Environ. Sci. Technol.* 2005, 39, 4513-4522.

- (28) Sweetman, A.; Cousins, I.T.; Seth, R.; Jones, K.C.; Mackay, D. A dynamic level IV multimedia environmental model: Application to the fate of polychlorinated biphenyls in the United Kingdom over a 60-year period. *Environ. Toxicol. Chem.* 2002, 21, 930-940.
- (29) Webster, E.; Mackay, D.; Di Guardo, A.; Kane, D.; Woodfine, D. Regional differences in multimedia fate model outcome. *Chemosphere* 2004, 55, 1361-1376.
- (29a) Fenner, K.; Scheringer, M.; MacLeod, M.; Matthies, M.; McKone, T.; Stroebe, M.; Beyer, A.; Bonnell, M.; Le Gall, A.C.; Klasmeier, J.; Mackay, D.; Van de Meent, D.; Pennington, D.; Scharenberg, B.; Suzuki, N.; Wania, F. Comparing estimates of persistence and long-range transport potential among multimedia models. *Environ. Sci. Technol.* 2005, 39, 1932-1942.
- (30) Kirkby, M.J.; Jones, R.J.A.; Irvine, B.; Gobin, A.; Govers, G.; Cerdan, O.; Van Rompaey, A.J.J.; Le Bissonnais, Y.; Daroussin, J.; King, D.; Montanarella, L.; Grimm, M.; Vieillefont, V.; Puigdefabregas, J.; Boer, M.; Kosmas, C.; Yassoglou, N.; Tsara, M.; Mantel, S.; Van Lynden, G. *Pan-European Soil Erosion Risk Assessment: The PESERA Map, Version 1, October 2003*. European Soil Bureau; Research Report No.16, EUR 21176; Office for Official Publications of the European Communities; Luxembourg, 2004.
- (31) PELCOM. *Pan-European land use and land cover monitoring*. (<http://www.geoinformatie.nl/projects/pelcom/>). Visited at October 1, 2005.
- (32) Roemer, M.; Baart, A.; Libre, J.M. *ADEPT: development of an Atmospheric Deposition and Transport model for risk assessment*; TNO-report B&O-AR 2005-208; Apeldoorn, 2005.
- (33) Jones, R.J.A.; Hiederer, R.; Rusco, E.; Loveland, P.J.; Montanarella, L. *The map of organic carbon in topsoils in Europe. Version 1.2, September 2003*; Publication no.72 S.P.I.04.72. http://eussoils.jrc.it/ESDB_Archive/octop/octop_data.html; JRC; Ispra, 2004.

- (34) New, M.; Lister, D.; Hulme, M.; Makin, I. A high resolution data set of surface climate over global land areas. *Climate Res.* 2002, 21, 1-25.
- (35) MARS. *The crop growth monitoring system*. http://agrifish.jrc.it/marsstat-/Crop_Yield_Forecasting/cgms.htm. Visited at October 1st, 2005.
- (36) EMEP. *Modelled Air Concentration and Deposition Data*.
http://www.emep.int/Model_data/model_data.html; Visited at November 10, 2006.
- (37) Brandes, L.J.; Den Hollander, H.A.; Van de Meent, D. *SimpleBox 2.0: a nested multimedia fate model for evaluating the environmental fate of chemicals*; Report number 719101029; National Institute for Public Health and the Environment (RIVM); Bilthoven, 1996.
- (38) Stroebe, M.; Scheringer, M.; Held, H.; Hungerbühler, K. Inter-comparison of multimedia modeling approaches: modes of transport, measures of long-range transport potential and the spatial remote state. *Sci. Total Environ.* 2004, 321, 1-20.