Spatially explicit multimedia fate models for pollutants in Europe: State of the art and perspectives

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A B S T R A C T

A review by Hollander et al. (in preparation), discusses the relative potentials, advantages and shortcomings of spatial and non spatial models of chemical fate, highlighting that spatially explicit models may be needed for specific purposes. The present paper reviews the state of the art in spatially explicit chemical fate and transport modeling in Europe. We summarize the three main approaches currently adopted in spatially explicit modeling, namely (1) multiple box models, (2) numerical solutions of simultaneous advection–dispersion equations (ADE) in air, soil and water, and (3) the development of meta-models. As all three approaches experience limitations, we describe in further detail geographic information system (GIS)-based modeling as an alternative approach allowing a simple, yet spatially explicit description of chemical fate.

We review the input data needed, and the options available for their retrieval at the European scale. We also discuss the importance of, and limitations in model evaluation.

We observe that the high uncertainty in chemical emissions and physico-chemical behavior in the environment make realistic simulations difficult to obtain. Therefore we envisage a shift in model use from process simulation to hypothesis testing, in which explaining the discrepancies between observed and computed chemical concentrations in the environment takes importance over prediction *per se*. This shift may take advantage of using simple models in GIS with residual uses of complex models for detailed studies. It also calls for tighter joint interpretation of models and spatially distributed monitoring datasets, and more refined spatial representation of environmental drivers such as landscape and climate variables, and better emission estimates. In summary, we conclude that the problem is not "how to compute" (i.e. emphasis on numerical methods, spatial/temporal discretization, quantitative uncertainty and sensitivity analysis...) but "what to compute" (i.e. emphasis on spatial distribution of emissions, and the depiction of appropriate spatial patterns of environmental drivers).

1. Introduction

In the last years, increasing interest has been raised by spatially explicit multimedia chemical fate and transport models for the assessment of chemical concentrations. In this contribution we do not advocate systematic use of spatially explicit models: a discussion of the circumstances under which it is worth using spatially explicit models as an alternative to simpler and more traditional non spatial models in chemical fate evaluation can be found in Hollander et al. (this issue).

We rather discuss here which spatially explicit models to use, depending on the purposes of the study, once it is clear that a spatial model is needed.

It is worth mentioning that models used at a large scale and fine resolution (e.g. a catchment, an urban area, a contaminated site) are usually implemented to provide answers to very specific questions grounding on a wealth of information, and particularly field surveys and local, *ad hoc* measurements, and are usually requested to target a much finer accuracy. Throughout the paper, we limit instead our discussion to models applied for assessments at a small scale and coarse resolution, e.g. a country or a continent, which are conceptually comparable to "evaluative" models adopted in risk assessment (e.g. EC, 2003, 2004; Brandes et al., 1996). Although these models ideally aim at simulating chemicals at a given location, they normally target trends and orders of magnitude rather than point-wise quantities; the latter may be in fact affected by local factors (e.g., the stack height of an atmospheric emission or the mixing volume at the outlet of a water emission, or the specific location of a sampling point used for model evaluation), which are normally excluded from models at coarser resolution. Therefore, criteria to assess whether a large scale model matches or does not match observations are quantitatively very different from the ones used for small scale models (for instance, a factor 10 and an explained variance of 50% are already reasonable performances at a small scale, while for a large scale model a factor higher than 1.5 and explained variance less than 70% may be unacceptable), and this derives from a qualitatively different expectation from the model results.

It must be also pointed out that, as spatial distributions of contaminants can be reconstructed directly from monitoring data, through geostatistical estimation (e.g. Wackernagel, 1997), or other more recent techniques such as self-organizing maps (Kohonen, 1990; see also Pistocchi et al., submitted for publication), it is not uncommon to refer to such reconstructions as "models" as well. The discussion might be broadened to a large extent, but this is beyond our scope. In this paper we only focus on "causal" models, i.e. models predicting chemical concentrations given emissions (direct mode) or emissions given an observed concentration (inverse mode).

2. Spatially explicit models between realism and simplicity

2.1. Early models

Early spatially explicit models were developed in the 1990s and early 2000s, as rather coarse multiple linear box models each representing a region (Wania and Mackay, 1995; MacLeod et al., 2001), and were more conceptual schemes of possible mechanisms of environmental distribution of chemicals, than tools for the prediction of concentrations at a specific location. In this sense, they are rather similar to conceptual schemes such as the CliMoChem (Wegmann, 2004) and ChemRange (Scheringer et al., 2004a,b,c) models, aiming at describing not even a broad region as a box, but a more abstract entity such as a latitudinal zone on earth. Such models describe what Mackay (2001) calls "evaluative environments" and can only account for the chemical transfer between large regions, and for coarse-scale variability of landscape and climate.

2.2. More recent models

More recent models have attempted a more realistic description of contamination with reference to specific zones of more limited extension (Suzuki et al., 2004; Pennington et al., 2005; Prevedouros et al., 2004; MacLeod et al., 2005). In those cases, the region of interest was described through a more refined grid of cells with resolution of a few hundreds of kilometers, for each of which a mass balance was computed, considering local emissions, local removal mechanisms and transport to and from neighboring cells. This is done in practice by writing a system of linear equations in number equal to the grid cells, which are solved in a straightforward way for steady state (e.g. Pennington et al., 2005) or transient conditions (e.g. Suzuki et al., 2004). Parameters describing landscape and climate were assigned considering the actual geography of each cell, achieving thus some realism. However, these models cannot be regarded as "realistic" chemical fate simulators, for a number of reasons. In the first place, a common approach in the setup of these models is to specify landscape and climate parameters, hence inter- and intra-media transfer rates, as representative values constant in time, which hampers the possibility of capturing mechanisms of transfer strongly dependent on time variations of environmental processes which may be very relevant (Lammel, 2004). Adopting constant values for environmental removal rates may make of little use developing models more complex than steady state calculations (see von Waldow et al., 2008).

Another remarkable simplification that makes these models comparable with single box models is the high degree of simplification in describing the spatial variation of landscape and climate parameters, usually by providing an average value over a very large region. Moreover, intra-media transporting currents (such as the water discharge between two cells, or wind flows) are not always easy to estimate. In particular, while transport in the inland stream network is fundamentally unidirectional and one-dimensional, and advection is clearly dominant over dispersion, transport in oceans needs always (at least) a two-dimensional description, and the atmosphere may well require a three-dimensional description. Things are complicated by the importance that dispersion takes on advection in these cases. The representative transporting currents estimated in the above quoted models are likely to reproduce the correct order of magnitude of advection and dispersion removal rates, but cannot be used to describe spatial patterns arising from these mechanisms, such as contamination plumes from a source of emissions (see, for instance, the comparison between box models and atmospheric transport models presented in Hollander et al., 2008).

For these reasons, a similar performance of spatial and non spatial models is often highlighted (e.g. in the comparison of models for HCB and PAHs by Armitage et al., 2007).

2.3. Complex transport models

To describe more complex spatial patterns of chemical transport, the full advection–dispersion equation (ADE; e.g. Pistocchi, 2008a) should be solved, considering a two- (for oceans) or three-dimensional (for the atmosphere) field of transporting currents. For soils, the ADE is often solved in one (vertical) dimension. ADE models of very high complexity have been developed, such as DEHM-POP (Hansen et al., 2004), or MSCE-POP (Gusev et al., 2005). Lammel et al., 2007, propose the use of such more complex models, with specific reference to the MCTM model, as tools to identify the mechanics of pollutant transport; the authors highlight that traditional box models may underestimate long range transport and overestimate sinks of chemicals in the environment, and should be therefore calibrated with benchmarks provided by such more complex models.

Complex models based on the ADE bring together what is considered as the most realistic method for each environmental compartment considered. Typically, these models have grown by adding soil and ocean compartments to an original atmospheric model, which makes them biased towards the atmospheric phase of environmental fate. The spatial patterns of contamination described by ADE-based models over large regions have a more realistic appearance, although their results have seldom been experimentally evaluated for chemicals other than conventional pollutants, due to the very limited monitoring data available.

Reliance on these models is based primarily on the reasonable performance of the ADE solutions in each medium separately, which has been tested traditionally at fine scale for soils (a plot or an experimental vertical profile), and at regional and even global scale for oceans and, most of all, atmosphere.

Despite their high degree of sophistication, these models have a number of weaknesses. First of all, notwithstanding their elegant mathematical formulation, they are not fully based on fundamental physical relationships; at steps, a number of empirical correctors are added which are in practice calibration parameters impossible to evaluate in the absence of experimental data. These correctors are particularly used at the interface between two media, e.g. in the description of volatilization fluxes, often represented through a "series of resistances" analogy (e.g. Schwarzenbach et al., 1993) the parameters of which are only derived empirically. Understanding the limited physics behind even rigorous solutions to the ADE in the context of environmental modeling has stimulated criticism towards claims of their being "physics-based", in favor of a more modest but philosophically conscious definition of "mediating models" (see e.g. Morton, 1993, and Beven, 1996, for a more thorough discussion).

Another aspect not to be neglected is that all numerical solutions to the ADE carry some unavoidable artifacts, among which maybe the most important one is numerical dispersion (e.g. Zheng and Bennett, 1995). This phenomenon depends on the resolution of the numerical grid of models in relationship to the celerity of transfer of perturbations, and may be of the same magnitude as physical dispersion in the environment. A demonstration of the equivalence between grid resolution and assumed "physical" dispersion, and its positive exploitation in modeling, is provided by Sukop (2001), with reference to soil contaminant modeling.

Table 1
Meaning of the terms R, D and the type of distance, for Eq. (4) applied to different media.

Environmental compartment	Dominant dilution mechanism	ξ	w	Dilution function, D	Decay function, R
Stream network	Advection	Curvilinear abscissa on the stream network	Stream velocity (input map)	$\alpha(\xi)$	$e^{-k\frac{\xi}{w}}$
Atmosphere	Dispersion	Euclidean distance	Suggested constant value: 3 m/s	$\alpha_0\xi^b$	$e^{-k\frac{\xi}{w}}$
Soil, ocean	Mixing at the source	Euclidean distance	–	$\alpha_0\bar{\xi}$	$\frac{\delta(\xi)}{\bar{\xi}k+w}$

See text for the meaning of the symbols.

It is worth mentioning, finally, that these models rely on an assigned field of transporting currents, which is in itself generally derived from a hydrologic, oceanographic or meteorological model, in turn affected by different errors, which can be seldom evaluated. As a conclusion, although in principle much more accurate and capable of detailed description of processes, sophisticated models produce as output spatial distributions of contaminants, which are *a priori* only one possible (although real-looking and consistent) realization of the environmental dispersion process, unless accurate model calibration is conducted. Model equations cannot be separated by the set of model parameters assigned. The fact that more than one combination of model equations and parameters yield equally satisfactory results, sometimes called model equifinality (e.g. Beven, 2006), further contributes to weakening our belief in even very comprehensive and advanced models.

On the other hand, these models are overwhelmingly data demanding, computation intensive, and require much longer time to run compared to simpler models, which makes them extremely expensive and not practical for routine assessments or preliminary studies. Although modern computers and parallel, high performance computing do not pose, in principle, strong computational limitations, practical constraints when applying numerical solutions of the ADE in environmental modeling still limit their spatial resolution or coverage: as numerical solutions of the ADE are obtained by dividing the computational domain into an array of discrete elements for each of which an equation is solved algebraically, describing a domain of approximately 5000 km × 5000 km such as continental Europe with a resolution of 1 km would imply a system of equations of order 2.5×10^7.

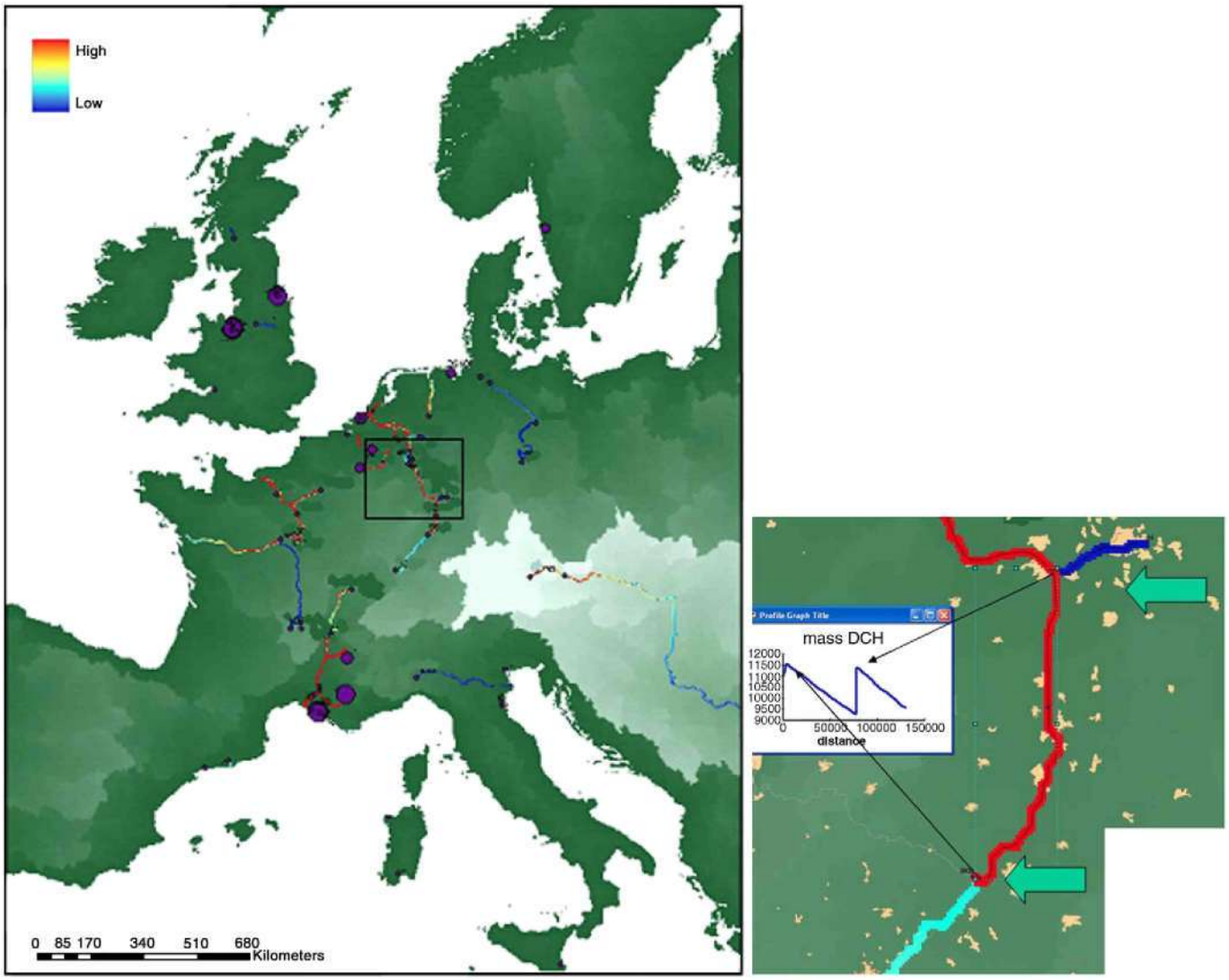

Fig. 1. Example of maps of chemical mass (concentration times water flow) transported in river waters across Europe from a generic distribution of point sources (points represent emissions of dichloroethane (DCH) from the EPER emission inventory: www.eper.ec.europa.eu). Left: Whole Europe. Right: Detail highlighting how mass profiles along a river stretch show a saw-like sequence of exponentials with peaks at each emission source. Concentrations are estimated with Eq. (1) following the algorithms presented in Pistocchi, 2005. In right panel, arrows represent dichloroethane emissions to water.

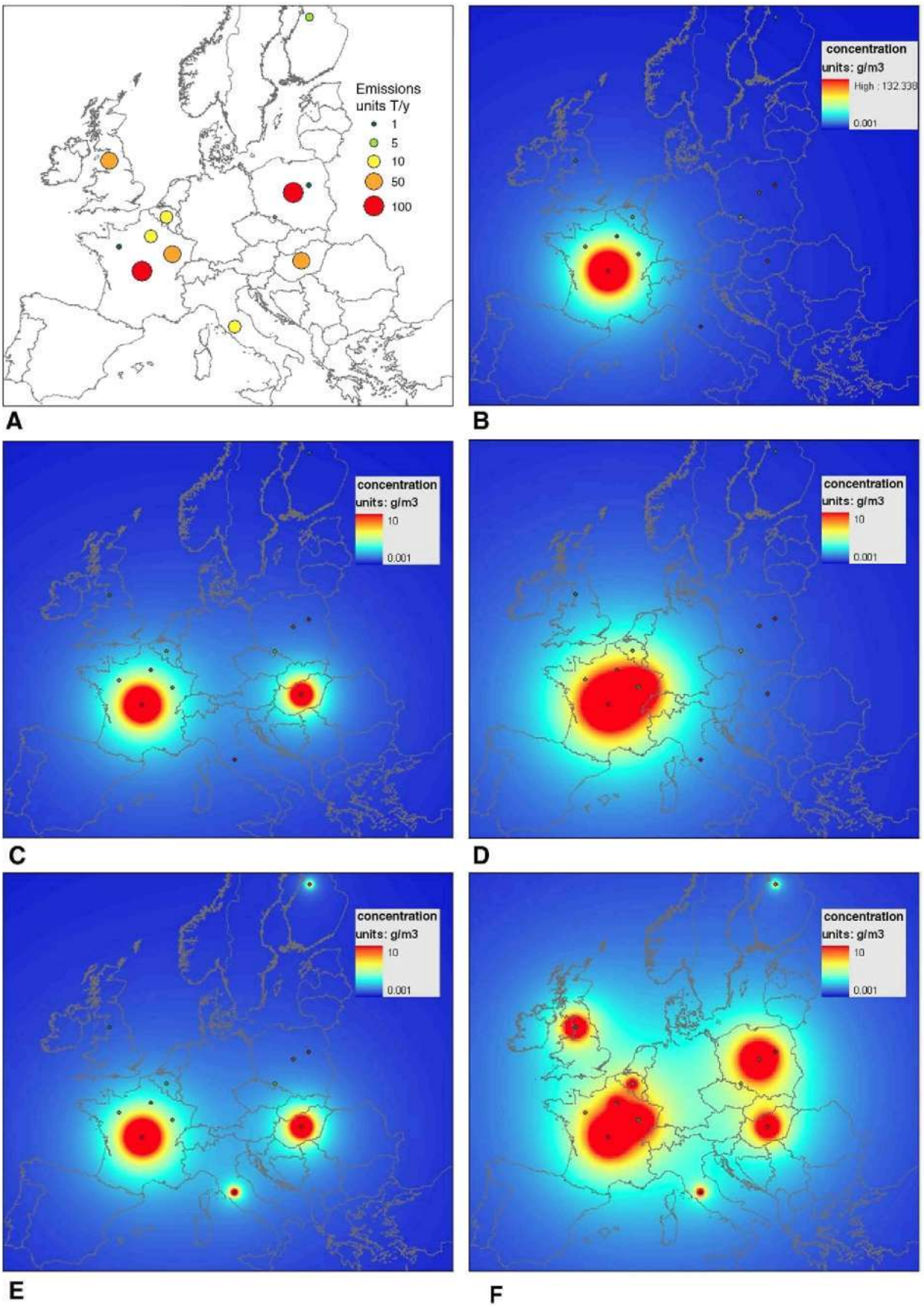

Fig. 2. Example of maps of concentrations across Europe of a generic pollutant due to punctual emissions of different intensities, as represented in A. Maps present cumulative concentrations due to emissions in a single point (B), two remote points (C), two close points (D), four distant points (E) and many points distributed across Europe (F).

Table 2
Examples of landscape and climate parameters used in chemical fate modeling.

Compartment	Parameter	Methods	Examples	Typical resolution	Links
Air	Atmospheric boundary layer height	Reanalysis of meteorological simulations	ECMWF ERA40;	2.5°	http://www.ecmwf.int/
	Air temperature, precipitation, wind speed	Interpolation of climatological stations; reanalysis of meteorological simulations	ECMWF ERA40; CRU Climatology	2.5° 10′	http://www.cru.uea.ac.uk/cru/data/
	Aerosol concentration and deposition flux	Remote sensing of aerosol optical depth; specialized atmospheric models	MODIS products; ICAROS products of LANDSAT TM and SPOT; MERIS products; TM5, REMSAD, CMAQ models	30 m^{-3} km	http://modis.gsfc.nasa.gov/ http://envisat.esa.int/instruments/meris/ http://www.phys.uu.nl/~tm5/ http://remsad.saintl.com/ http://www.cmaq-model.org/
	OH-radical concentration	Specialized atmospheric models	TM5 model (see Pistocchi et al., 2006)	1°	
Water	Runoff	Calibrated models	GRDC composite runoff fields (Fekete et al., 2000)	30′	http://www.bafg.de/GRDC/Home/homepage__node.html
	Hydraulic geometry	Morphological analysis of digital elevation models (DEM)	See Pistocchi and Pennington, 2006	Depending on the digital elevation model	
	Suspended particulate matter (rivers)	Empirical models, sediment rating curves	See review in Pistocchi, 2008c	Depending on the digital elevation model	
	Suspended particulate matter (seawater)	Remote sensing	See Pistocchi et al., 2006		http://oceancolor.gsfc.nasa.gov/SeaWiFS/
	Ocean mixing depth	Models Interpolation of climatological observations	E.g. GETM model for Europe Monterey–Levitus dataset		http://getm.eu/ http://www.cdc.noaa.gov/data/gridded/data.nodc.woa94.html
	Sea temperature	Models Remote sensing	E.g. GETM (See Pistocchi et al., 2006)	1°	http://getm.eu/ http://oceancolor.gsfc.nasa.gov/SeaWiFS/
	Sea currents	Models Interpolation of drifter data	E.g. GETM (See Pistocchi et al., 2006)	25 km	http://getm.eu/
Soil	Soil texture, bulk density, porosity, organic carbon	Soil surveys and national/international compilations	European Soil Database	1 km	http://eusoils.jrc.ec.europa.eu/
	Soil moisture	Simplified soil water balance models	See Pistocchi et al., 2008	1 km	
	Erosion rates	Erosion models	See review in Pistocchi, 2008c	1 km	
Vegetation	Leaf area index	Remote sensing	FAPAR derived products; see Pistocchi et al., 2006	1 km	http://fapar.jrc.ec.europa.eu

But the main disappointment with the use of such models lies often in the excessive detail of their description of chemical fate, compared with the actual knowledge of both chemical emissions and the fundamental physico-chemical properties of substances in the environment. Indeed, predicted environmental concentrations are highly correlated in space to emissions, which may explain a very large portion of the variance (see e.g. the discussion in Hollander et al., this issue). Unfortunately, however, emissions are seldom known in orders of magnitude, and almost never in spatial patterns for most chemicals. Therefore, the use of these models should be limited to detailed studies on pollutants.

2.4. The issue of model evaluation

In the literature, there are numerous examples of spatially explicit models used at local or regional scale, with different degrees of success in simulation and prediction. Any review of this material is beyond the scope of this work. It is interesting, on the other hand, to notice that not many models have been fully evaluated for prediction of chemical fate at the continental or global scale. A recent example by Lamon et al. (2009), investigating implications of climate change on the fate of pollutants, shows that the spatially explicit multimedia model BETR Global (MacLeod et al., 2005) predicts correct orders of magnitude, and captures general spatial trends, of air concentrations of PCB congeners 28 and 153. However, the assessment is referred to 16 measurement points, included in 9 out of 288 cells in the model domain in the Northern Hemisphere. The information used for validation represents the best available knowledge on these chemicals, and gives an idea of the current difficulties in obtaining well-tested model simulations. Given this difficulty of evaluation, it is not surprising that one of the main uses of spatially explicit models is in the development and testing of hypotheses on global transport mechanisms. Examples are provided by the study of the global fate of European PAHs by Sehili and Lammel (2007), using a complex ADE-based transport model; the study of Armitage et al. (2006), on PFOA, using a multiple box model; the assessment of persistence and long range transport potential of chemicals performed by Leip and Lammel (2004), highlighting the importance of exploring global variations in chemical concentration using realistic spatially explicit models. However, studies with similar aims have been conducted also with far simpler models (e.g. Scheringer et al., 2004a,b), and the advantage of using global transport models based on the ADE, with high computational burden and a higher number

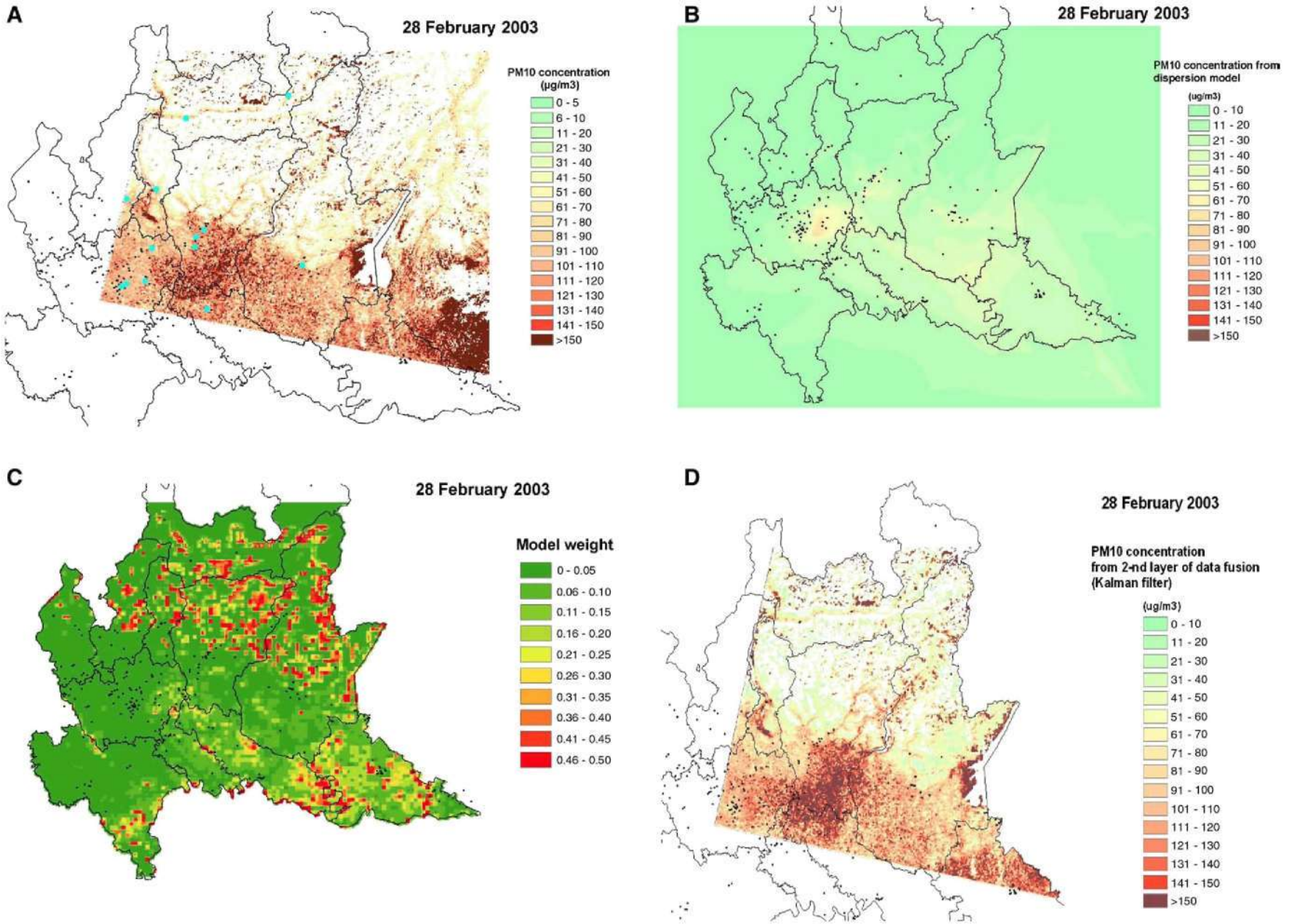

Fig. 3. Example of data assimilation between (A) remote sensing-derived PM10 concentrations in the ambient air in the region of Lombardy, N. Italy; (B) PM10 concentrations derived from the REMSAD atmospheric fate model for aerosol. The two data models are fused using a Kalman-filter based algorithm that optimizes the assimilation procedure assigning variable weights to the 2 data sources across the domain (C). The final, optimized result is given in map (D).

of parameters affecting the results (with clear consequences on sensitivity assessment and error tracking) is still to be clearly proven in the absence of full model validation. We will return on the issue of model evaluation later in this paper.

3. Strategies of simplification

This practical experience with models has fostered the development of alternative strategies seeking a balance between realism in the spatial distribution of chemicals on one side, and practicity and cost of use on the other. In the literature, there are some examples of models developed in an attempt to simplify the ones based on the ADE, at the same time keeping a relatively very fine spatial resolution. For instance, Bachmann (2006), developed a classic multiple box model with boxes of 1 km resolution for all processes on land in Europe, while keeping a coarser resolution in the atmosphere, described in turn through a trajectory-based approach. However, the very fine resolution on land does not correspond to knowledge detailed enough, e.g. concerning soil properties or the representation of hydraulic geometries determining water residence times. Moreover, the numerical complexity of calculations does not correspond to the realism in simulation one may expect, given the simplistic first-order representation of many environmental processes. Therefore, this type of approach promises to lead to little improvement with respect to classical multi-box models with coarser resolution, at the same time approaching the computational burdens of ADE-based models.

3.1. Multimedia models or cascades of media-specific models?

A first relevant step forward in spatially explicit multimedia chemical fate modeling has come instead from the consideration that, once a chemical is emitted to one environmental compartment, it can be transported to other compartments, but its feedback fluxes from other compartments to the one of emission are very rarely relevant in quantitative terms (Margni et al., 2004). Therefore, for most applications, it is sufficient to treat one compartment at a time (i.e. set up a single-medium chemical fate model), and treat transport to other media as "losses"; then these losses can be used as input to other single-medium models of different compartments. There are a number of models that can be applied to each single medium, or to a combination of two. Examples of spatially explicit chemical fate models that can be applied to model large regions for different single (or coupled) environmental media include GREAT-ER (Feijtel et al., 1997), a stochastic simulator based on a plug-flow equation model of rivers, that predicts concentrations of chemicals from wastewater and other sources in inland waters and has been used in the context of water management; SWAT (Neitsch et al., 2002), a model including catchment hydrology, soil processes, stream processes, with the resolution of a sub-catchment, used for the simulation of nutrients and pesticides over large domains; and LOTOS-EUROS (Schaap et al., 2008), an atmospheric transport model for continental Europe.

A cascade of off-the-shelf single-medium models combined together may be a practical option, but often a single one of these models is in itself already rather complex, and their combination may result in a still very complex model.

3.2. Meta-models

An increasingly followed strategy to simplify models is to build meta-models (e.g. Piñeros Garcet et al., 2006). A meta-model can be described as a model that allows estimating chemical concentrations as a function of physico-chemical properties, emission rates and a set of pre-calculated concentration fields corresponding to reference chemicals. Usually such model is obtained through some form of linear or non linear regression analysis (e.g. Tiktak et al., 2006), artificial neural networks (e.g. El Tabach et al., 2007) or other similar techniques. Pre-calculated concentration fields are generated by making emissions and physico-chemical properties vary over appropriate and representative ranges of values, usually following random sampling logics (Piñeros Garcet et al., 2006). An extensive meta-modeling activity has been developed in Europe for pesticides, using the PEARL model (Leistra et al., 2001) within the HAIR project (http://www.rivm.nl/rvs/risbeoor/Modellen/HAIR.jsp) and the MACRO model (Larsson and Jarvis, 1999) within the FOOTPRINT project (http://www.eu-footprint.org).

Once pre-calculated fields are available, the calculation of concentrations is extremely fast and generally very accurate with respect to the original model. Therefore, meta-modeling is a feasible technique to generalize complex numerical models to cases not yet analyzed. The main problem with this approach is that sometimes the pre-calculation of concentration fields is very time-consuming, and the benefits of pre-calculation can be observed only when frequent and varied routine application of the underlying model is foreseen. If this is not the case, developing pre-calculated fields may be less convenient than just running the underlying model in different case studies. Moreover, meta-models rely strictly on the underlying model assumptions and parameterization. It is not infrequent that an even well established model undergoes important modifications in time. In such cases, adaptations to the meta-model to reflect modifications simply require re-running the model.

A special case of meta-model is the construction of source–receptor relationships for advective–dispersive media such as the atmosphere or oceans. In such cases, the ADE for a conservative contaminant is always linear. Moreover, in such case the only relevant fate process is, by definition, dilution (advection and dispersion). Roemer et al. (2005), have developed a meta-model that they called ADEPT, which describes the concentration from emissions in each European country, distributed in space like population density, by running the LOTOS model (Builtjes et al., 2003) with appropriate assumptions. Their meta-model takes the form of maps of spatially explicit source–receptor relationships, i.e. concentrations at a given location (receptor) from emission at another (sources). Source–receptor model approaches have been extensively adopted in long range atmospheric modeling within global or continental decision support systems such as GAINS (http://gains.iiasa.ac.at/gains/).

3.3. Analytic elements

Besides meta-modeling, in recent years attention has been focused on the possibility of describing even irregular, complex patterns using simple fundamental mathematical relationships in place of complex numerical methods. This path has been followed first in the domain of groundwater modeling, where a technique, called of the "analytic elements" (Strack, 1989; Haitjema, 1995), has been developed which consists of a superposition of simple analytical solutions to the ADE, corresponding to individual features such as sources/sinks, discontinuities in flow properties, boundaries, etc. Each solution is infinite in space, and appropriate consistency conditions are specified in order to solve the system of these solutions. In this way, realistic, complex flow patterns can be reproduced by superimposing simple models.

4. GIS-based modeling

With the development of computational geography (e.g. Openshaw and Abrahart, 2000) and the proposal of systematizing geographic information system (GIS) practice into a more structured "geographic information science" (Goodchild, 1992), GIS functions have started being studied not just as software functionalities, but rather as "methods" to be used for broad classes of geographic modeling problems. This has fostered the inclusion in GIS of generic operators enabling the construction of simulation models of environmental processes, without the need to code separate numerical models to be eventually coupled with GIS. Examples of this approach, also enabling time-dependent simulation, are provided by the GIS package PCRaster (Van Deursen, 1995; Karssenberg, 2002) which has been also used to

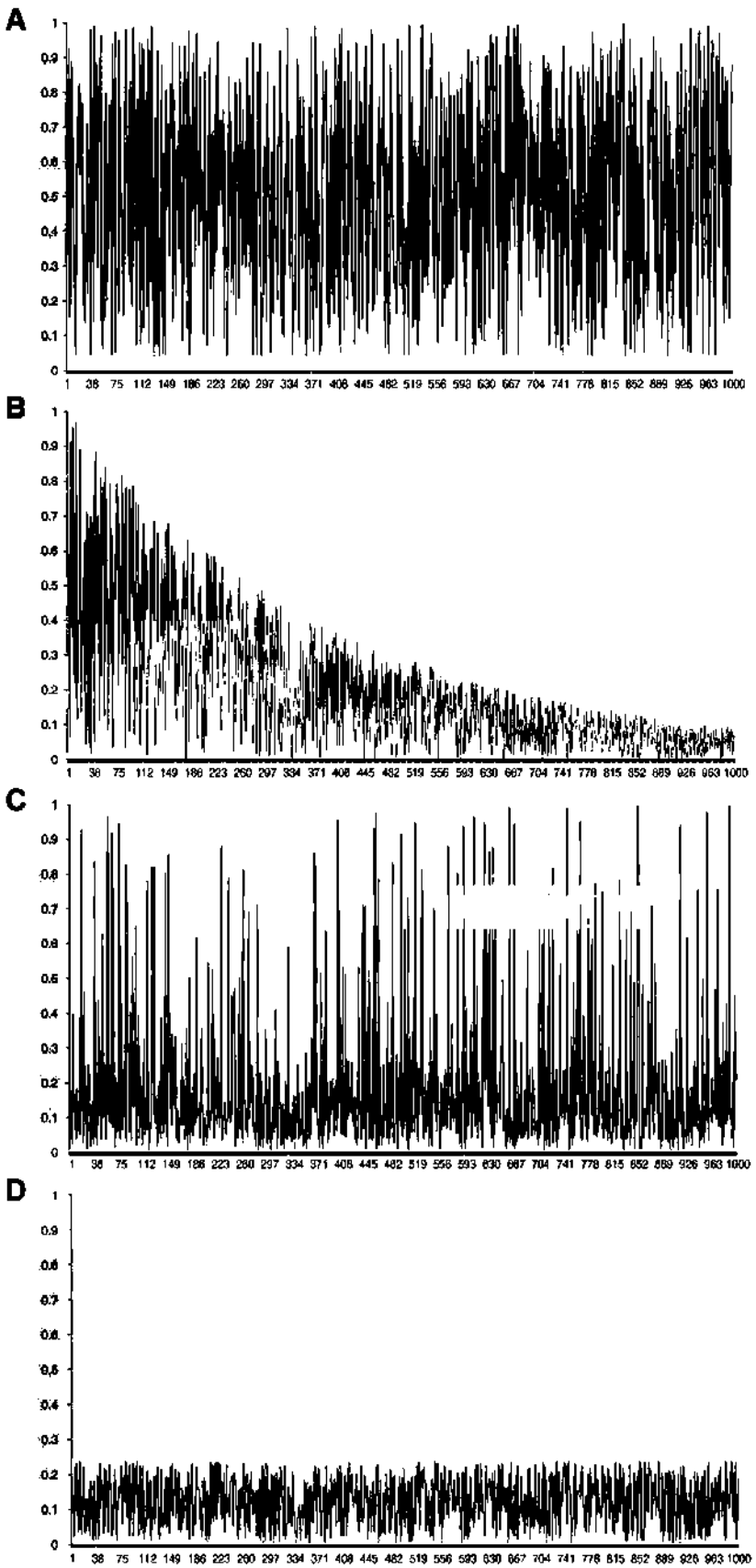

Fig. 4. Random emissions (A) and predicted mass in the presence of a strong exponential trend in degradation (B); with degradation rates within the same range of values as in (B) but randomly distributed in space, but with no trend (C); and with a constant degradation rate equal to the average in (B) (D). Numbers on the *x*-axis represent different, equally spaced locations along a gradient of temperature. The graphs are merely illustrative. See text for details.

build a pan-European hydrologic model (De Roo et al., 2000). Another example is the functionality provided by popular ESRI products ArcInfo© and, more recently, ArcGIS©, to perform simple groundwater contaminant modeling by combining flow path analysis and time-dependent dispersion (see Tauxe, 1994, for further details).

We focus here on an approach, conceptually similar to the analytic elements, proposed in the field of chemical fate modeling, grounding on simple standard GIS-analytical operations, including local drainage delineation and map algebra, to describe the spatial patterns of contaminants (Pistocchi, 2005, 2008b). The basic idea of this approach is to replace the numerical solution to the ADE with a superposition of local analytical solutions: in most cases the spatial distribution of chemicals in the environment can be described as a reasonable first approximation using very simple conceptual schemes, and particularly the elementary flow schemes traditionally applied in compartmental analysis: besides the already mentioned box model, or "continuous stirred tank reactor" (CSTR), the plug-flow (PF) and Gaussian plume (GP) models usually represent well most of the typical environmental distributions.

4.1. The plug-flow model

For instance, a PF model is often the elective tool for simulation of river quality (e.g. Chapra, 1997). In this case, concentration of a chemical substance along the stream network downstream of an emission of intensity E is described by the analytical solution:

$$C(\xi) = \frac{E}{\alpha(\xi) w} e^{-\frac{k\xi}{w}} \tag{1}$$

where w is the stream velocity, x the curvilinear abscissa along the stream line Γ originating from the emission point, with origin $\xi = 0$ at the location of emission, $\alpha(\xi)$ is the stream cross sectional area (which can be represented as in Pistocchi and Pennington, 2006), and k is the first-order decay constant of the chemical.

When w and/or k are not constant but depend on ξ, the exponential in Eq. (1) needs to be replaced by $e^{-\int_0^{\xi} \frac{k}{w} d\xi}$. The formula can be computed practically as explained by Pistocchi (2005). The curvilinear abscissa ξ can always be converted to geographic coordinates (x,y) given the position of the stream network.

4.2. The Gaussian plume model

The GP model is used for screening level modeling of air emissions (e.g. Turner, 1994), groundwater pollution (e.g. Domenico and Schwarz, 1998), and ocean or lake dispersion problems (Csanady, 1973). In this case, concentration downstream or downwind of an emission, within a field of wind or water current of constant intensity w and direction parallel to the p-axis, is:

$$C(p,q) = \frac{E e^{-\left(\frac{q}{ap^b}\right)^2}}{\alpha_0 w \ (ap^b)} e^{-k\frac{p}{w}}; \tag{2}$$

α_0 is a parameter representing dilution near the source and is conceptually similar to a mixing depth/height, as discussed in Pistocchi and Galmarini (in press), while a and b represent empirical parameters of plume dispersion. It is clear that the Gaussian plume scheme cannot be used when variations in wind or current direction and intensity are relevant, as in the case of continental scale modeling. However, with reference to atmospheric dispersion in the European context, it has been shown that Eq. (2) can be replaced with the following:

$$C(p,q) = \frac{E}{\alpha_0 w \xi^b} e^{-k\frac{\xi}{w}} \tag{2'}$$

with $\xi = (p^2 + q^2)^{0.5}$, which proves valid over long distances, irrespective of wind, w (Pistocchi and Galmarini, in press). Also, those authors suggest acceptable values of parameters α_0, b and w generally applicable in Europe for screening level modeling.

4.3. The continuous stirred tank reactor

For the CSTR model, concentration is:

$$C(p,q) = \frac{E}{\alpha_0 \bar{\xi}} \left[\frac{\delta(\xi)}{\frac{\bar{\xi}}{\bar{w}} k + \frac{\bar{\xi}}{\bar{w}}} \right] \tag{3}$$

where $\bar{\xi}$ is the side length of the CSTR, α_0 represents the mixing height or depth of the CSTR, $\bar{w}$ is the average wind or water current speed within the CSTR, and $\delta(\xi)$ is the integral of the Dirac delta function over the CSTR, equal to 1 for $\xi \leq \bar{\xi}$ and 0 for $\xi > \bar{\xi}$. Eq. (3) can be used only to predict the concentration at the emission location, but not outside of it. This scheme can then be used when dilution outside of the emission location is so high that concentrations become negligible, as is the case of small emissions to the ocean compartment, or when k dominates over $\alpha_0 w$, i.e. advection is negligible.

4.4. Generalization for GIS-based modeling

Using these simple flow schemes allows the prediction of the local pattern of concentration by assuming *a priori* the mathematical shape of the patterns of concentration arising from a given source. The three simple models of Eqs. (1), (2′) and (3) can be used to compute concentrations everywhere in space arising from a single emission at a given location. Their characteristic is that they are just algebraic combinations of functions of spatially distributed parameters and an appropriate distance from emissions, in the form:

$$C = \frac{E}{D} R \tag{4}$$

where E is the emission value, and D and R are maps representing the "dilution function" D and the "removal function" R, and can be represented as maps as a function of an appropriate distance ξ from the source of emission. Table 1 summarizes the meaning of terms R, D and the type of distance ξ to be used in Eq. (4) for the cases discussed above.

For a spatial distribution of emissions, the linearity of the ADE considered here allows to simply superimpose the results from the different emissions. The most trivial case is for Eq. (3); in this case, one may consider a map of emissions E and maps of k, α_0 and $\bar{w}$ values, and the overall results is just the algebraic combination of these maps. For the case of Eq. (1), it is possible to use specific superimposition functions known as "flow accumulation" (Burrough and McDonnell, 1998; Pistocchi, 2005). An example of this type of calculation is shown in Fig. 1. For Eq. (2′), one calculation for each emission location is required, and then results for all emission points need to be summed together. This may become very impractical for a large number of emissions, but it is still very convenient for a limited number of sources (of the order of 10^3), compared with running complex numerical models (Vizcaino and Pistocchi, in preparation). An example of this calculation is shown in Fig. 2. Calculations conducted in this way are usually much quicker than the corresponding numerical models. Also, the model resolution is not as strongly limiting as in the case of numerical methods, because it is not related to the order of a system of equations to be solved, as in the case of models based on the ADE.

The use of these simplified models, implemented directly using GIS-analytical capabilities, allows obtaining reasonably realistic spatial distributions of chemical concentrations, through extremely simple calculations. This has been shown with reference to the

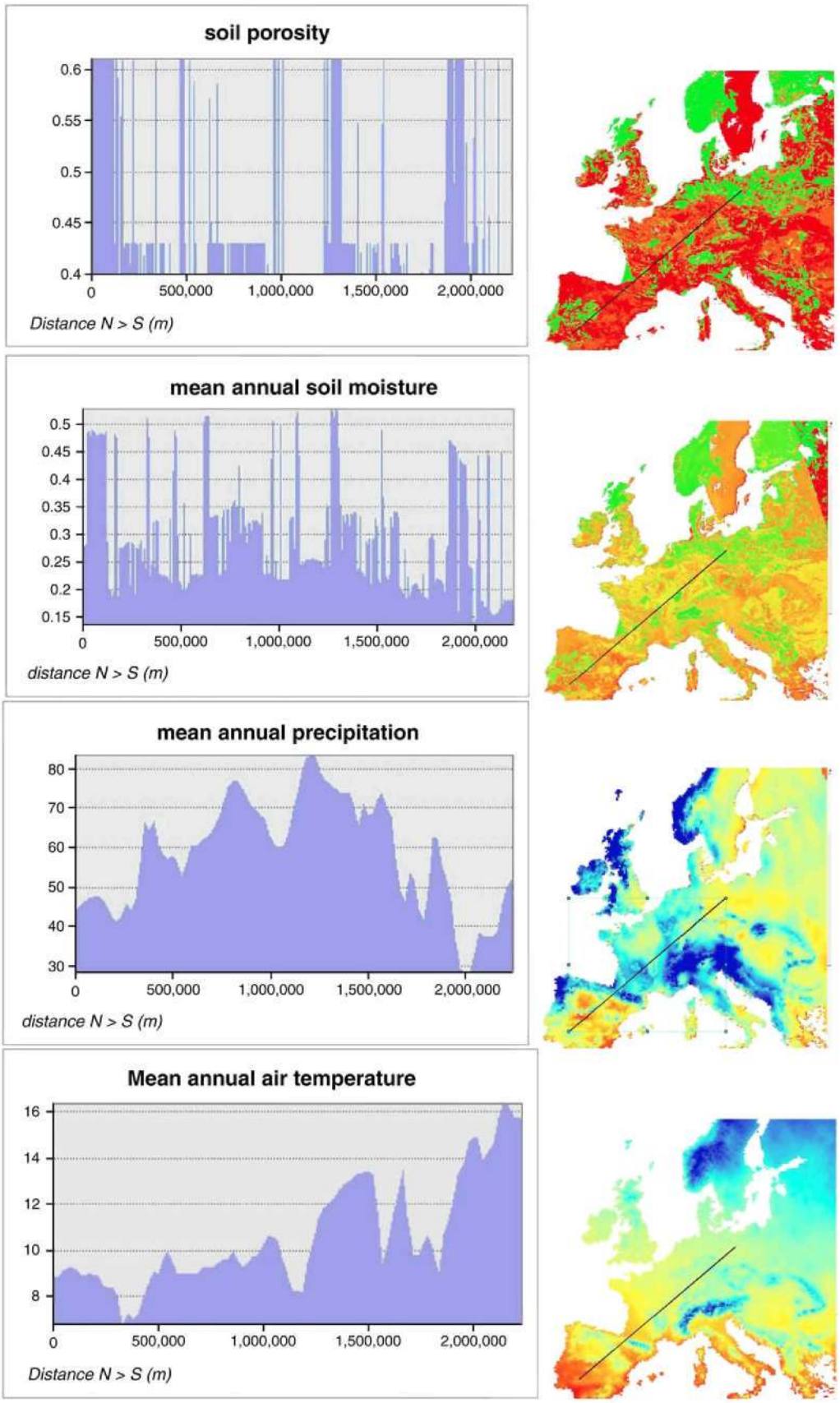

Fig. 5. Examples of landscape/climate parameters used in multimedia fate and transport models, showing spatial trends (precipitation, air temperature), no trend (soil porosity) or a weak trend (soil moisture, roughly corresponding to a combination of precipitation and soil porosity). The graphs display the variables measured along the cross section line indicated in the corresponding maps; distance along the line (*x*-axis) is measured from North (N) to South (S). The graphs are representative of a broad area as indicated on the map of mean annual precipitation.

continental distribution of PCBs and dioxins in Europe (Pistocchi, 2008b); to the fate of pyrethroid insecticides in Europe (Pistocchi et al., 2009); to the distribution of lindane in Europe (Vizcaino et al., in preparation); and industrial emissions from the European Pollution Emission Register (EPER) (Pistocchi and Galmarini, in press).

5. Relevant landscape and climate parameters for spatially explicit chemical fate models

5.1. Data availability on the Digital Earth

An aspect of utmost importance in the use of spatially explicit chemical fate models is the choice of landscape and climate parameters. There is now an unprecedented availability of spatial information covering the whole globe, and more in detail specific regions such as the US or, more recently, Europe. In the years from the first theoretical discussions on the "Digital Earth" concept (Wikipedia, not dated) in the early 1990s, to the implementation of Google Earth and similar tools in mid 2000s, all human activities, and particularly scientific investigation, have seen their geographic dimension enormously empowered and made explicit. In Europe, the adoption of a directive on an infrastructure for spatial information (EC of the Directive, 2007) promises to consolidate a scenario where landscape and climate parameters no longer represent a problem for fate and transport modeling; similar conditions occur in other parts of the world. Fate and transport models require a potentially endless list of landscape and climate variables, of which the most common are summarized in Table 2. It is not uncommon that new mechanisms, and consequently new environmental parameters required for their representation, come to the attention of researchers. For instance:

- In recent times, studies on the effects of intermittent rainfall have suggested the utility of the number of dry days per month as a proxy in fate and transport modeling (Jolliet and Hauschild, 2005);
- The importance of sinking fluxes associated with organic matter has suggested the use of chlorophyll in seawater (Dachs et al., 2002); and
- Studies on the sorption of chemicals to suspended matter in air or water support the adoption of more complex parameterization of these variables (Götz et al., 2007).

A relevant impulse to the development of spatial data sets has been given by increasingly available remote sensing products, more and more used also in the field of chemical fate modeling (e.g. Vijayaghavan et al., 2008; Jurado et al., 2004; Montzka et al., 2008; Boschetti et al., 2006).

5.2. Data assimilation

In particular, data assimilation techniques bringing together remote sensing datasets with deterministic modeling results bear great potential for improving further the realism of environmental characterization provided by the respective models. This can be of particular importance when assessing the link between environmental pressure and human health risk (Sarigiannis et al., 2002, 2004). A good example of the usefulness of data assimilation in this regard is given by the study of particulate matter pollution in the ambient air at the regional scale. At this scale several problems with the spatial and temporal coverage of the environmental monitoring networks hamper the efficacy of pollution pattern determination. Lack of accuracy in key input parameters, including emission inventories, constrain the applicability of complex atmospheric models. Assimilation of particulate matter data from ground-based measurements with optical indicators of pollution such as turbidity as measured by the optical depth of atmospheric aerosol can enhance both the spatial extent and validity of the air pollution data in a spatially resolved manner. This allows the generation of a model for translating optical indices into pollutant concentrations with the aid of ancillary models and data sets such as meteorological and landscape information (Fig. 3(A)). Further fusion of this model with results from deterministic atmospheric fate modeling using decision-theory based algorithms to optimize the assimilation of the underlying data on the basis of self-estimates of uncertainty (e.g. using the well known Kalman filter) results in a meta-modeling system, which optimizes the uncertainty profiles across the computational domain (Fig. 3(B)). The model fusion algorithm assigns automatically variable weights to the different data sources (e.g. the atmospheric fate models, see Fig. 3(C)) depending on the determination of the minimal residual error of the method across the computational domain. The final result is an improved map of the horizontal profile of PM10 concentrations, characterized by optimal use of all relevant datasets and models (Fig. 3(D)).

5.3. Types of variability in landscape and climate data, and their relevance in spatial predictions

It should be stressed that the required detail in the description of the environment depends on the purpose of modeling. Some parameters are intrinsically so highly variable, that an explicit account of their spatial distribution increases the variability of predicted chemical concentrations, but does not provide any better insight on the spatial trends, hot spots etc., which are dominated by the interplay between emissions and environmental parameters. As an example, let's consider the case of a random emission to soil at a number of locations (Fig. 4(A)), for a chemical subject to degradation only (i.e., all other removal mechanisms are negligible). Let's also assume that degradation follows a strong exponential trend with temperature, increasing with location number. The distribution of predicted mass, i.e. ratio of emission to degradation rate, will follow an exponential decrease consistent with the trend in degradation (Fig. 4(B)). The oscillations of mass across the trend depend on the variability of emissions. Let's now assume that degradation rates are distributed randomly, following a Gaussian distribution with the same mean and standard deviation as the temperature-dependent rates. We have then a distribution of predicted masses as in Fig. 4(C). If we eventually replace the random-varying rate of Fig. 4(C) with the average of removal rates, we obtain the distribution of predicted masses of Fig. 4(D). In Fig. 4(C), inclusion of the spatial variability of the degradation rate increases significantly the variability of mass, but does not allow detecting trends. Therefore, it is appropriate to include the variability of the degradation rate if one is interested in extremes, but not if one wants to highlight hotspots or geographical trends.

In reality, the different parameters used in models may or may not show a spatial trend. Fig. 5 shows examples of such parameters, which appear to have different evidence of spatial trends. An objective way to detect spatial trend, as is well known, is to draw the variogram of a spatially distributed variable, i.e. the plot of the variance of data points at a given spatial distance, as a function of the distance itself (see e.g. Clark, 1979), as is common practice in the geostatistical analysis of data.

6. The evaluation of spatially explicit models

An issue of key importance in any modeling exercise is the evaluation of models with observations. In the specific case of spatially explicit fate models of chemicals, evaluation poses a number of issues. First of all, chemicals are typically measured in the environment at trace levels, the collection of sufficiently representative samples may be extremely difficult and, sometimes, analytical procedures are not sufficiently consolidated, and do not provide sufficient quality assurance (e.g. Lepom et al., 2009; Quevauviller et al., 2007).

Even when good sampling and analytical procedures exist, the costs of sample collection and analysis often make data generally rather rare and sparse; especially for substances of high variability in time and space, it is extremely difficult to build a data set of samples

well representing the "true" spatial distribution. More often, samples only reflect the statistics of environmental concentrations, and not values that can be considered stable point-wise, hence used for the point-wise assessment of models. For this purpose, only observational data should be used that were originally conceived and developed as "spatial"; these data are increasingly available for large regions or the globe, e.g. from monitoring of lipids from commercial milk products (Kalantzi et al., 2001; Weiss et al., 2005; Malisch and Dilara, 2007), air passive samplers (Gioia et al., 2007; Jaward et al., 2004), and systematic spatial sampling of a given environmental medium (Mejier et al., 2003; Loos et al., 2009).

For these reasons, with spatial predictions we do not necessarily seek to match monitoring data to an extreme accuracy: modeling exercises have been judged successful or useful even when highlighting orders-of-magnitude discrepancies with observations; models may happen to be considered acceptable when matching orders of magnitude with no or weak correlation with observations in space, and such correlation has been demonstrated to be sufficiently high only in a few cases. When selecting the level of computational complexity of fate models, the fact that model evaluation is at least troublesome and sometimes impossible with the given data available cannot be ignored. Therefore, on the one hand using comprehensive models describing detailed mechanisms of transport in the environment may be useful in order to provide a benchmark; on the other hand, considering that no model represents the truth and even highly physics-based models introduce at some point semi-empirical parameterizations and arbitrary assumptions, complex models may tend to lack transparency and hide some of their assumptions behind their own complexity, thus making the tracking of model errors a difficult task; moreover, their realistic-looking output based on great computational capabilities tends to define a virtual reality obeying the rules included in the model, and thus pushing simpler models to mimic their output rather than observations, in turn scarce and difficult to retrieve. This basically obliterates the lines of research not included in the development of the complex models used as benchmark.

7. Conclusions and perspectives: GIS-supported revival of simple models?

7.1. Models and observations in hypothesis testing

In the last decades, voices have been raised against the development of overly complex models, especially in such fields as hydrology, where terrific computational developments have eventually shown that little further progress can come from modeling, and relevant steps forward are necessarily related to developments in measurement techniques (e.g. Beven, 2001). On the contrary, a very strong movement has grown in parallel, aimed at fostering the use of models as tools for hypothesis testing rather than accurate simulators of reality (e.g. Beven, 2006). This has brought researchers and practitioners to reconsider simple models not just as remainders from the times when computation was expensive, but rather as parsimonious ways to interpret the complex reality in the presence of limited experimental information, hence preferable according to the Ockham's razor principle. The same considerations apply to chemical fate assessment, an area where hardly any further development is expected on the computational side, if not paralleled by substantial advances on observations.

7.2. The crucial role of chemical emission maps

Current limitations in the development of chemical fate models are identified mainly in the lack of information on chemical emissions to the environment; therefore, developing complex and computation-intensive models, which involve a high effort in development, is likely to bring no better results than much simpler calculations that can be crudely summarized as estimates of chemical masses by algebraic combination of maps of emissions and dilution or removal factors (Eq. (4)). This is true in general, but particularly so for spatially explicit models, where emissions explain usually a very large amount of the variance in environmental concentrations of a given chemical. In this case, the most critical piece of information, and the most relevant area of research, seems to be about emission inventorying (e.g. Pistocchi and Bidoglio, in preparation; Hollander et al., in preparation; Pistocchi and Loos, 2009).

7.3. Way forward: maps and simple calculations for hypothesis testing

In this paper, we have discussed a few aspects related to the development and use of spatially explicit models for the prediction of chemical fate at the scale of the European continent and similarly large regions. A shift in model use from process simulation to hypothesis testing is under progress, in which explaining the discrepancies between observed and computed chemical concentrations in the environment takes importance over prediction *per se*.

This shift may take advantage of using simple models with residual uses of complex models for detailed studies, under conditions of high uncertainty concerning emissions and environmental processes such as phase partitioning and degradation. In addition, GIS technology and methods invite to developing models by reasoning in terms of maps, keeping the model structure extremely simple (as to be implemented with map algebra) and capitalize on spatial data to refine the spatial representation of environmental drivers such as landscape and climate variables, and better emission estimates.

The problem, in this perspective, becomes one of producing appropriate maps of emissions, on the one hand, and removal rates, on the other hand; the latter capitalize on available spatial data sets of landscape and climate parameters such as air temperature, vegetation, precipitation, soil organic carbon content, runoff, etc., which are increasingly available for large regions also as a consequence of developments in remote sensing of the earth system. Such simple calculations based on a wealth of spatial information on environmental variables maintain an ease of tracking errors, visualizing intermediate results and discussing in a quick and productive way the orders of magnitude of phenomena of which the interplay results in the environmental concentration of chemicals. At the same time, this approach lends itself to reducing the error of the overall environmental fate assessment across the spatial domain of study. This can be achieved by the application of model fusion techniques employing decision-theory approaches and information filters (such as the Kalman filter) to optimize the relative importance of different superimposed modeling results on the basis of a weight-of-evidence approach. This approach, which so far has been successfully implemented in atmospheric pollution problems for integrating satellite-based remote sensing with atmospheric models and ground-based air quality monitoring data, could be a viable advancement towards the enhanced utilization of GIS-based environmental fate models at the continental and regional scale.

It can be stated, in summary, that the problem is not "how to compute" (given the wealth of available numerical methods, spatial/temporal model ressolution, quantitative uncertainty and sensitivity analysis techniques, etc.) but "what to compute", as often our knowledge of emissions and environmental processes, and our chemical monitoring systems limit the application of even very simple models.

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