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Highlighted article

Examination of the uncertainty in contaminant fate and transport modeling: A case study in the Venice Lagoon

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ABSTRACT

A Monte Carlo analysis is used to quantify environmental parametric uncertainty in a multi-segment, multi-chemical model of the Venice Lagoon. Scientific knowledge, expert judgment and observational data are used to formulate prior probability distributions that characterize the uncertainty pertaining to 43 environmental system parameters. The propagation of this uncertainty through the model is then assessed by a comparative analysis of the moments (central tendency, dispersion) of the model output distributions. We also apply principal component analysis in combination with correlation analysis to identify the most influential parameters, thereby gaining mechanistic insights into the ecosystem functioning. We found that modeled concentrations of Cu, Pb, OCDD/F and PCB-180 varied by up to an order of magnitude, exhibiting both contaminant- and site-specific variability. These distributions generally overlapped with the measured concentration ranges. We also found that the uncertainty of the contaminant concentrations in the Venice Lagoon was characterized by two modes of spatial variability, mainly driven by the local hydrodynamic regime, which separate the northern and central parts of the lagoon and the more isolated southern basin. While spatial contaminant gradients in the lagoon were primarily shaped by hydrology, our analysis also shows that the interplay amongst the in-place historical pollution in the central lagoon, the local suspended sediment concentrations and the sediment burial rates exerts significant control on the variability of the contaminant concentrations. We conclude that the probabilistic analysis presented herein is valuable for quantifying uncertainty and probing its cause in over-parameterized models, while some of our results can be used to dictate where additional data collection efforts should focus on and the directions that future model refinement should follow.

1. Introduction

Contaminant mass balance fate and transport models are useful heuristic tools extensively used to gain insight into system dynamics and to estimate contaminant concentrations and loadings (Diamond et al., 1994, 2005; Wania and Mackay, 1999). These models are simplifications of reality and as such are inherently uncertain. Model uncertainty derives from both structural and parametric uncertainty (Spear, 1997). Structural uncertainty not only reflects the inability to capture all the significant processes underlying system behavior, but also the inability to express them in an unequivocal manner in the model. For example, even if we develop a model of optimal complexity that comprises all the essential components of the real system, we usually realize that most ecological and physical processes can be described mathematically by a variety of relationships that entail different assumptions and levels of complexity (Arhonditsis et al., 2006). On the other hand, parametric uncertainty refers to the inability to accurately specify the values of the parameters of the mathematical structure used to represent the system. Parametric uncertainty stems from the analytical errors, the imprecision of measurements as well as spatio-temporal parameter variability (McKone, 1996).

The latter source of uncertainty is of particular importance in fate and transport modeling in which the relative effects of individual parameters on model predictions are not well understood and can be contaminant-specific (Matthies et al., 2004).

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The parameters represent physico-chemical properties of substances (e.g., partition coefficients), environmental system variables (e.g., sediment deposition rates) and contaminant emission/loading estimates (e.g., tributary loadings). Many of the input parameters are difficult to measure (e.g., chemical degradation rates), others exhibit significant spatiotemporal variability (e.g., concentration of total suspended sediment), and existing system information cannot unambiguously support their characterization (Arhonditsis et al., 2006). For example, fate and transport models of aquatic systems require an estimate of the sediment resuspension flux which is characterized by considerable seasonal and spatial variation (Sfriso et al., 2005a). Thus, a single value assigned to this parameter is a clear over-simplification that essentially represents an "effective" (spatially/temporally averaged) value. This problem of parameter uncertainty can be partly overcome by increasing the spatial and/or temporal model resolution, but the greater data requirements of the refined grids/increased time steps are rarely met while the models become excessively complex, which introduces further error and uncertainty. Therefore, despite its inability to fully accommodate the spatial and temporal variability of model parameters, the adoption of a coarser resolution in space and time is a pragmatic solution that reflects the inevitable tradeoffs between model articulation and knowledge gained (Costanza and Sklar, 1985).

Because contaminant fate and transport models are often used to inform environmental management decisions, it is important to clearly communicate the uncertainty underlying model outcomes. Decision makers need to consider both the mean predictions and the associated confidence intervals (Reckhow, 1994). The problem of uncertainty of mathematical models has received considerable attention in the field of water resources management and several studies have rigorously addressed issues pertaining to structural and parametric errors (Spear, 1997; Omlin and Reichert, 1999; Pappenberger and Beven, 2006). Nonetheless, a recent meta-analysis showed that most of the aquatic mechanistic biogeochemical models published over the last decade did not adequately assess prediction error; modelers are still disinclined to embrace uncertainty analysis techniques and to assess the reliability of the critical planning information generated by the models (Arhonditsis and Brett, 2004). This reluctance is pronounced in contaminant fate and transport modeling, although efforts to systematically assess the underlying uncertainty can been found in the literature (e.g., McKone and Ryan, 1989; McKone, 1994; Kuhne et al., 1997; Liu et al., 1999; Webster et al., 2004). Recognizing the existing methodological gap, Cowan et al. (1994) and then Wania and Mackay (1999) underscored the importance of developing simple tools for evaluating uncertainty in multi-chemical, multi-media models.

In this paper, we present a Monte Carlo analysis for quantifying environmental parameter uncertainty in a multi-segment, contaminant fate and transport model based on the fugacity/aquivalence approach of Diamond et al. (1992) and Mackay (2001). We analyze the fate of two metals (Cu and Pb) and three POPs (octachloro dibenzo-*p*-dioxin (OCDD), octachloro dibenzofuran (OCDF) and PCB-180) in the Venice Lagoon. Sommerfreund et al. (2009) provide full details of the model, its parameterization and results from a deterministic analysis. In this study, scientific knowledge, expert judgment and observational data are used to formulate prior probability distributions and to characterize the uncertainty pertaining to 43 environmental system parameters. Uncertainty propagation through the model is then assessed by a comparative analysis of the moments (central tendency, dispersion) of the model output distributions. Finally, we apply principal component analysis (PCA) in combination with correlation analysis to identify the most influential parameters and to gain insight into the system dynamics, at least as depicted in our model.

2. Material and methods

2.1. Study system

The Venice Lagoon (Fig. 1) is composed of three basins (north, central and southern) defined by three inlets to the Adriatic Sea (Lido, Malamocco and Chioggia). It is a network of channels and mud flats with few principal deep channels (>15 m), covering a 550 km^2 area with a mean depth of 1.1 m (Solidoro et al., 2004a). The amplitude of the prevailing diurnal tidal flow can be greater than 1 m (Umgiesser et al., 2004). The Venice Lagoon receives inflows from 12 major tributaries resulting in water quality (dissolved oxygen, organic matter, suspended sediment) and physical parameter (e.g., salinity) gradients from the main land to the Adriatic Sea (Solidoro et al., 2004a). The lagoon is turbid, with net sediment loss from the central basin as a result of resuspension promoted by anthropogenic activities, e.g., the introduction of manila clams, clam fishing (Sfriso et al., 2005b), historic river diversion (Degetto and Cantaluppi, 2004) and a high frequency of boating and mechanical dredging (Pranovi et al., 2004).

Sediment profiles suggest that peak contaminant loadings to the Venice Lagoon from the industrial area of Porto Marghera occurred from 1950s to 1980s (e.g., Frignani et al., 2001; Frignani et al., 2005). Present loadings mainly originate from the industrial area of Porto Marghera (Bellucci et al., 2002; Carrer and Leardi, 2006), export from the watershed via tributaries (Collavini et al., 2005), remobilization of contaminated sediments (Sommerfreund et al., 2009), direct atmospheric deposition to the lagoon (Gambaro et al., 2004; Guerzoni et al., 2004) and discharges from municipalities. Our study focuses on the contaminants with the most complete data sets; namely, OCDD, OCDF, PCB-180, Cu and Pb.

2.2. The Venice Lagoon model

Full details of the Venice Lagoon model are presented by Sommerfreund et al. (2009). The model is based on the fugacity/aquivalence approach of Mackay (2001), Diamond et al. (1992) and Bhavsar et al. (2004), to estimate the concentrations of single species POPs and multi-species metals. To accommodate the spatial variability of the system, a ten-box model was constructed in which each segment consists of three compartments, i.e., water, upper and lower sediment (Fig. 1). Model segmentation follows that of Solidoro et al.'s (2004b) two-dimensional hydrodynamic model used to reproduce the spatial variability of the lagoon water quality. We have adopted their reference scenario which reflects steady-state hydrologic circulation patterns in the system, assuming no wind, mean annual tributary flows and an idealized sinusoidal M2 tide level at the inlets (Solidoro et al., 2004b).

The model considers both depositional (segments 1–5, 9, 10) and erosive (segments 6–8) environments. In depositional environments, net sediment accumulation occurs and water exchanges mass with the upper sediment compartments via particle-bound deposition/resuspension and bidirectional diffusion. The exchanges of mass between upper and lower sediment layers occur by biological/physical mixing and bidirectional diffusion. Contaminants are

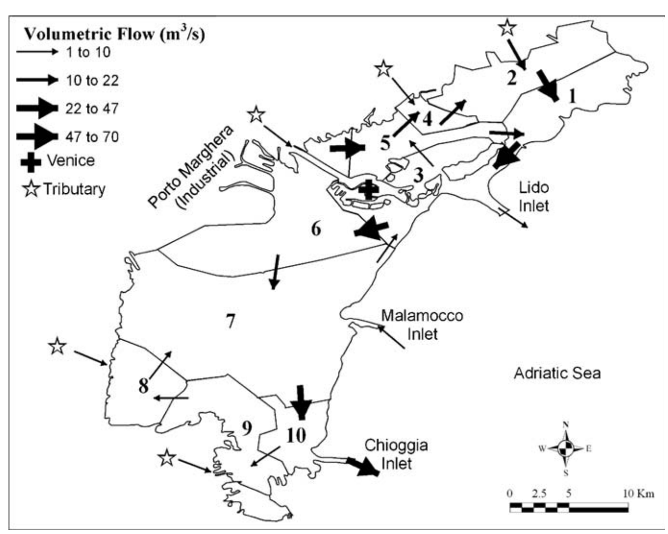

Fig. 1. The ten segments and the hydrologic circulation patterns considered by the Venice Lagoon model (Solidoro et al., 2004a). The industrial area of Porto Marghera neighbors segment 6. Segments 6–8 were modeled as erosive areas, while erosion of in-place pollution occurs in segment 6. The northern, central and southern basins are represented by segments 1–5, segments 6–8 and segments 9 and 10, respectively.