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Assessment of contaminant fate in catchments using a novel integrated hydrobiogeochemical-multimedia fate model

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HIGHLIGHTS

- INCA-Contaminants: a novel hydro-biogeochemical-contaminant fate model is presented.
- Climate and geochemical controls on contaminant fate in catchments were described.
- Soil organic matter mineralization was the most influential parameter.
- Diffuse run-off from soil is a main path of PCB mobilization at catchment scale.

GRAPHICAL ABSTRACT

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ABSTRACT

Models for pollution exposure assessment typically adopt an overly simplistic representation of geography, climate and biogeochemical processes. This strategy is unsatisfactory when high temporal resolution simulations for sub-regional spatial domains are performed, in which parameters defining scenarios can vary interdependently in space and time. This is, for example, the case when assessing the influence of biogeochemical processing on contaminant fate. Here we present INCA-Contaminants, the Integrated Catchments model for Contaminants; a new model that simultaneously and realistically solves mass balances of water, carbon, sediments and contaminants in the soil-stream-sediment system of catchments and their river networks as a function of climate, land use/management and contaminant properties. When forced with realistic climate and contaminant input data, the model was able to predict polychlorinated biphenyls (PCBs) concentrations in multiple segments of a river network in a complex landscape. We analyzed model output sensitivity to a number of hydro-biogeochemical parameters. The rate of soil organic matter mineralization was the most sensitive parameter controlling PCBs levels in river water, supporting the hypothesis that organic matter turnover rates will influence re-mobilization of previously deposited PCBs which had accumulated in soil organic matrix. The model was also used to project the long term fate of PCB 101 under two climate scenarios. Catchment diffuse run-off and riverine transport were the major pathways of contaminant re-mobilization. Simulations show that during the next decade the investigated boreal catchment will shift from being a net atmospheric PCB sink to a net source for air

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and water, with future climate perturbation having little influence on this trend. Our results highlight the importance of using credible hydro-biogeochemical simulations when modeling the fate of hydrophobic contaminants.

1. Introduction

Chemical pollution has been traditionally regarded as the result of intentional and unintentional point source emissions from industry, agriculture and waste disposal. However, diffuse pollution sources in the landscape as well as contaminant storage by and remobilization from soils and sediments add enormous complexity to the task of assessing exposure and risk from both experimental and theoretical points of view (Daughton, 2004; Nizzetto et al., 2010; Scheringer, 2002). Developing tools for understanding and predicting the behavior of chemical contaminants with multiple emission pathways and complex environmental behavior is an essential step for prioritizing and implementing effective pollution management strategies.

Based on their specific physical–chemical properties and modes of emission, contaminants can distribute across environmental compartments. Accumulation in natural reservoirs including soils, sediments and vegetation is an important process controlling the cycling and budgets of several persistent and hydrophobic compounds of very high environmental and human health concern. Photo-bio-chemical degradation and natural burial processes in soils and sediments perform a vital service in limiting/retarding the exchange of these contaminants across interfaces between the geosphere and the biosphere. While storage in natural reservoirs is subordinated to persistence and mediated by physical–chemical interactions with the substrate, climate and disturbance in general can lead to contaminant remobilization. Mechanistic predictions of the fate and distribution of these contaminants requires a framework capable of simulating both contaminant and environmental related properties and processes with a sufficient level of accuracy.

To date, a number of mathematical models have been developed to facilitate exposure assessment of chemical contaminants. Important developments occurred primarily within two fields: multimedia fate models (MMFM) and water quality modeling (WQM) (Keller, 2006). MMFMs derive from over 30 years of fugacity-based model development (Mackay, 2001) (although the fugacity formalism is not an essential feature of MMFMs) and focus mainly on describing the distribution of chemicals across multi-media environments in a simplistic, nevertheless holistic way. MMFMs assume a stochastic representation of thermodynamic activities of individual substances within environmental compartments and across interfaces. Based on the level of complexity and deviation from equilibrium, these models can provide predictions of contaminant distribution under thermodynamic equilibrium, steady state, or unsteady state conditions (Mackay et al., 1996, 1992). The representation of biogeochemical processes in MMFMs is typically reductionist. The adoption of a high level of simplification has been instrumental for facilitating MMFM model application and conceptual interpretation of simulation results. MMFMs tend to omit detailed descriptions of the dynamic processes connecting the bio-, hydro- and geosphere with contaminant fate and transport. Hence, MMFMs are useful tools for screening contaminant potential behavior over large spatial scales with a coarse time resolution (Brandes et al., 1996). It is nevertheless important to note that there are multiple examples of successful MMFM applications in highly unsteady conditions and small (e.g. catchment) spatial scales, in which dynamic environmental conditions are drawn from a stochastic representation of monitored data, rather than being mechanistically simulated (Ao et al., 2009; Bogdal et al., 2010; Camenzuli et al., 2012; Hollander et al., 2006; Kern et al., 2011; Mackay et al., 2014; Wang et al., 2012).

On the other hand, WQM-derived contamination models have been traditionally based on hydrological modules (for review see (Chapra, 2008)), and tend to include very detailed dynamic descriptions of the aquatic environment (e.g. (Bowden and Brown, 1984; Feijtel et al., 1997; Futter et al., 2012)). Nevertheless, existing contaminant modules within WQM miss the holistic character of MMFMs for representing environmental distribution of contaminants. WQM typically focus on in-stream processes and typically consider point source emissions along the course of the stream, while generally neglecting contaminant processing in the geosphere.

There are obvious trade-offs between the use of either MMFMs or WQMs and the unbridged gap between these two approaches severely limits the current ability to assess details of contaminant environmental fate. New integrative modeling tools are therefore necessary to predict fate and distribution of multiple contaminants in realistic environments. These tools need to include an adequate level of detail in descriptions of both contaminant-specific and environmental parameters which may vary interdependently over time as a function of climate, land use and land management.

The present paper: (i) introduces INCA-Contaminants (a new integrative hydrological, biogeochemical and multimedia fate model); (ii) describes the results of its evaluation against experimental measurements made in a river network and its catchment; and (iii) analyses the sensitivity of predicted aquatic contaminant concentrations to variations in chemical and biogeophysical parameters. A primary objective of the study was to evaluate the importance of soil organic matter (OM) turnover on contaminant remobilization. The analysis focuses on a set of PCBs in the low range of hydrophobicity (PCB 28, PCB 52 and PCB 101) for which (as a conservative scenario) relatively lower dependence on OM budgets and dynamics can be anticipated (compared to more hydrophobic substances). In addition, an exercise is presented in which the calibrated model was used to simulate long-term future budgets of PCB 101 in a boreal forest catchment under two contrasting climate scenarios.

2. Model description

2.1. INCA-Contaminant structure and INCA models legacy

A new chemical fate model (INCA-Contaminants) was developed by integrating two extensively tested dynamic hydro-biogeochemical models: INCA-C (Futter et al., 2007) (simulating organic matter mass balances in multi-branched, multi-land use catchments) and the sediment transport model INCA-Sed (Lazar et al., 2010) (simulating soil erosion and in-stream transport, deposition and entrainment of sediments). The INCA (Integrated Catchment) models are a family of complex, one-dimensional dynamic flow catchment models designed for water quality assessment (Whitehead et al., 1998a, 1998b). In summary, the discretization of catchment and river structure is provided by determining the order of streams, their length and width between reach points, and by compiling dimensions and characteristics of individual subcatchments (including soil properties and land use information) (Whitehead et al., 1998a). An underlying rainfall-runoff model (PERSiST) (Futter et al., 2014) computes hydrologically effective rainfall and soil moisture deficit time series that are used by INCA models to generate daily run-off and river flow predictions. PERSiST outputs are calibrated using catchment specific observations of river flow. INCA-C computes carbon (C) budgets in the terrestrial compartment, sediment and stream water by linking different pools (solid organic C (SOC), dissolved organic C (DOC) and inorganic C) using calibrated first order transformation kinetics in individual compartments. The same representation of terrestrial soil structure used by INCA-C is employed in INCA-Contaminants. Exchange of C across the different pools, surface