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HIGHLIGHTS

approach.

explored.

properties.

• The chemical persistence in real lakes

can be quantified by benchmarking

• The ranges of transformation halflives that can be measured were

 Dominant physical loss processes of chemicals rely on chemical and lake

 Benchmarking will open new chance for slowly degraded chemicals.

G R A P H I C A L A B S T R A C T



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ABSTRACT

The persistence of chemicals in the environment is rarely measured in the field due to a paucity of suitable methods. Here we explore the potential of chemical benchmarking to facilitate the measurement of persistence in lake systems using a multimedia chemical fate model. The model results show that persistence in a lake can be assessed by quantifying the ratio of test chemical and benchmark chemical at as few as two locations: the point of emission and the outlet of the lake. Appropriate selection of benchmark chemicals also allows pseudo-first-order rate constants for physical removal processes such as volatilization and sediment burial to be quantified. We use the model to explore how the maximum persistence that can be measured in a particular lake depends on the partitioning properties of the test chemical of interest and the characteristics of the lake. Our model experiments demonstrate that combining benchmarking techniques with good experimental design and sensitive environmental analytical chemistry may open new opportunities for quantifying chemical persistence, particularly for relatively slowly degradable chemicals for which current methods do not perform well.

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1. Introduction

Persistence in the environment is an undesirable property for synthetic chemicals that escape from the technosphere, and chemical persistence is enshrined as a hazard criterion in many regulatory frameworks for chemical management. The unacceptable thresholds of persistence in water and sediment, expressed as degradation half-lives, generally lie in the range of 1–6 months (Boethling et al., 2009; van Wijk et al., 2009; Hope et al., 2010; Moermond et al., 2012; UNEP, 2008). Therefore, the ability to measure chemical persistence on the time scale of months is a cornerstone of chemical management. However, there are currently very few studies that have directly measured the persistence of organic chemicals in the real environment.



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One approach that has been used to measure persistence in the environment is to compile a complete contaminant mass balance for a well-defined system. Some excellent studies using this approach have been conducted in Swiss lakes to study the persistence of chemicals used in consumer products (Buser et al., 1998; Stoll and Giger, 1998; Poiger et al., 2004). While such studies are valuable, the information obtained about chemical persistence is uncertain because it is directly linked to the accuracy of measurements of chemical fluxes in natural systems that are characterized by large spatial and temporal variability. Studies of chemical persistence in mesocosms, which are reconstructions of a small portion of the natural environment under controlled conditions, avoid most of the problems of temporal and spatial variability. There are guidelines for using mesocosms for higher-tier risk assessment of plant protection products, and they have been used to measure the persistence of some pesticides and other organic pollutants (Knuth and Heinis, 1995; Lahti et al., 1997; Knuth et al., 2000; EEC, 2002; Weaver et al., 2005). However, the ability of mesocosms to reproduce the full complexity of the environment is limited.

Chemical benchmarking is a technique that can be used to overcome the problems of spatial and temporal variability that are encountered with measuring persistence directly in real environmental systems. As typically applied, a benchmark chemical is a substance that behaves in a similar manner to the test chemical of interest, with the exception of the unknown property of interest. By comparing the behavior of the test chemical and the benchmark chemical, one obtains information about the relative magnitude of the unknown property between the two substances. If this property is known for the benchmark chemical, then it can be calculated for the test chemical. The benchmarking principle has been used to obtain information about chemical removal in estuaries by comparing the spatial gradient in the concentration of chemicals originating from the river with the gradient in salinity, a conservative tracer of dilution of the river water (Bester et al., 1998). Benchmarking is the basic principle behind other types of tracer experiments, for instance when a persistent water-soluble dve tracer is added with a water-soluble test chemical to a river to assess chemical removal (Sabaliunas et al., 2003; Whelan et al., 2007). This methodology has been extended to use a persistent chemical already present in a river as a tracer to assess the removal of other chemicals already present in the river (Radke et al., 2010; Kunkel and Radke, 2012).

In this paper we explore the potential of the benchmarking technique to quantify the persistence of chemicals in the real environment. In doing so, we use benchmarking to quantify several unknown characteristics of the environmental system, thereby broadening the definition of a benchmarking chemical to be a substance which is used as a reference point for the behavior of another substance. We focus on lakes, as they offer the possibility of studying persistence in water and sediment on time scales that correspond to the regulatory thresholds for persistence. We postulate that for some chemicals persistence can be quantified based on the change in concentration ratio of a test chemical and a benchmark chemical in (i) the medium that is the major vector of chemical input to the lake (e.g., inflowing water or an emission source) and (ii) the water flowing out of the lake. Our goal is to delineate the limitations of this methodology, and specifically to define the range of transformation half-lives that could be quantified with such a study and how this range depends on the partitioning properties of the test chemical, the properties of the lake system, and the uncertainty in our determination of the concentration ratios. We envisage that our assessment will provide a basis for designing field studies to quantify the persistence of chemicals in the aquatic environment under real conditions.

2. Theory

2.1. Model

We use a one-box model that assumes steady state and a chemical partitioning equilibrium between water and sediment to assess the potential to measure the persistence of contaminants in lake systems. The model includes water, suspended sediments and surface sediment in the lake, where the surface sediment consists of that volume of sediment that readily exchanges chemicals with the water column (i.e., non-buried sediment). It is assumed that the system is well-mixed. Chemical input to the lake, which could be via inflowing surface water, inflowing groundwater, atmospheric deposition, or direct emissions, is treated as a single term. Four processes for chemical loss are considered: advection, volatilization, sediment burial, and transformation (see Fig. 1).

The chemical mass balance is described by the following equation:

$$I = (G_{\mathsf{W}} + k_{\mathsf{W}}Af_{\mathsf{D}} + k_{\mathsf{B}}AK_{\mathsf{SW}}f_{\mathsf{D}} + k'_{\mathsf{R}}(V_{\mathsf{W}} + K_{\mathsf{SW}}f_{\mathsf{D}}V_{\mathsf{S}}))C_{\mathsf{W}}$$
(1)

where *I* is the rate of chemical input into the lake from all sources, mol h⁻¹; *G*_W is the flow rate of water out of the system, m³ h⁻¹; *k*_W is the overall air–water mass transfer coefficient for the chemicals referenced to the water phase, m h⁻¹; *A* is the surface area of the water body, m²; *f*_D is the fraction of the chemical in water that is freely dissolved; *k*_B is the burial rate of bulk sediment, m h⁻¹; *K*_{SW} is the sediment/water equilibrium partition coefficient, m³ water m⁻³ bulk sediment (i.e., the concentration of chemical in the bulk sediment divided by the concentration of freely dissolved chemical in the water); *k*'_R is the first order rate constant for transformation of the chemical in the system, h⁻¹; *V*_W is the volume of water, m³; *V*_S is the volume of surface sediment, m³; *C*_W is the total (freely dissolved plus sorbed) concentration of chemical in the water, mol m⁻³.

The half-life of the chemical in the lake due to transformation processes, $t_{0.5R}$, is defined by $t_{0.5R} = \ln 2 / k'_R$. Note that this definition includes transformation in both the water and the sediment compartments. It therefore gives a measure of the persistence in the lake as a whole, not media-specific persistence. The model does not specify where in the lake the transformation is occurring; it could be in the water column, in the surface sediment, or in both.

A version of the mass balance model coded in Excel was used to explore the potential of the theoretical framework for determining chemical persistence in lakes and to assess the sensitivity of the determined transformation rate constants to certain sources of uncertainty. In the model, k_W , K_{SW} and f_D were defined according



Fig. 1. Schematic illustration of the lake model showing chemical inputs (mol h^{-1}) in green and four removal fluxes in purple (mol h^{-1}), i.e., volatilization, advection, burial and transformation (the terms shown in the equations are defined in the text); red and black dots represent suspended solids and bottom sediments, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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