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Invited paper

Remoteness from sources of persistent organic pollutants in the multi-media global environment☆

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ABSTRACT

Quantifying the remoteness from sources of persistent organic pollutants (POPs) can inform the design of monitoring studies and the interpretation of measurement data. Previous work on quantifying remoteness has not explicitly considered partitioning between the gas phase and aerosols, and between the atmosphere and the Earth's surface. The objective of this study is to present a metric of remoteness for POPs transported through the atmosphere calculated with a global multimedia fate model, BETR-Research. We calculated the remoteness of regions covering the entire globe from emission sources distributed according to light emissions, and taking into account the multimedia partitioning properties of chemicals and using averaged global climate data. Remoteness for hypothetical chemicals with distinct partitioning properties (volatile, semi-volatile, hydrophilic, low-volatility) and having two different half-lives in air (60-day and 2-day) are presented. Differences in remoteness distribution among the hypothetical chemicals are most pronounced in scenarios assuming 60-day half-life in air. In scenarios with a 2-day half-life in air, degradation dominates over wet and dry deposition processes as a pathway for atmospheric removal of all chemicals except the low-volatility chemical. The remoteness distribution of the low-volatility chemical is strongly dependent on assumptions about degradability on atmospheric aerosols. Calculations that considered seasonal variability in temperature, hydroxyl radical concentrations in the atmosphere and global atmospheric and oceanic circulation patterns indicate that variability in hydroxyl radical concentrations largely determines the seasonal variability of remoteness. Concentrations of polybrominated diphenyl ethers (PBDEs) measured in tree bark from around the world are more highly correlated with remoteness calculated using our methods than with proximity to human population, and we see considerable potential to apply remoteness calculations for interpretation of monitoring data collected under programs such as the Stockholm Convention Global Monitoring Plan.

1. Introduction

The potential of a chemical to pose an environmental and/or human health risk as a result of long-range transport is a defining property of a persistent organic pollutant (POP). As such, long-range transport potential is identified in the Stockholm Convention as one of the screening criteria for determining whether a substance should be classified as a POP, and subject to global regulation. According to Annex D of the convention text (S.C, 2009), measured levels of a chemical "in locations distant from its release" should be supplied as evidence of a chemical's long-range transport potential during the screening process. Therefore, in the planning and design of measurement campaigns, and also, when analyzing measurement data from monitoring networks, it is critical to determine how distant or remote sampling locations are from pollution sources.

In a pioneering study, von Waldow et al. (2010) introduced a quantitative metric of the remoteness of regions at a global or regional scale from emissions of pollutants to the atmosphere, called remoteness index. Von Waldow et al.'s (2010) remoteness index is calculated from atmospheric transport modeling of a suite of volatile tracers with different atmospheric half-lives emitted at a constant rate on a specified geographical distribution. The results of the tracer modeling are fit to a non-linear function that provides a

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location-specific parameter that indicates the remoteness of locations in the model domain from the emissions. Westgate et al. (2010) subsequently proposed three methods to quantify the remoteness of air sampling sites. They define pertingency index values calculated for sampling sites as a measure of the proximity (1/remoteness) of the sites to spatially distributed emissions. Two of the methods proposed by Westgate et al. (2010) are based on the geographical distance of sources from sampling sites, and one method uses Lagrangian trajectory modeling. They applied these methods to determine the proximity of the sampling sites in the Global Atmospheric Passive Sampling (GAPS) study to estimated global emissions of polycyclic aromatic hydrocarbons (PAHs). Similar methods using Lagrangian trajectory modeling to quantify the remoteness of sampling sites in studies were later used by Westgate and Wania (2011) and Westgate et al. (2013).

In the approach proposed by Westgate et al. (2010), the location of a sampling site and also the specific time period for sampling are required to obtain the air-mass trajectories that end at the site. While, in principle, the whole globe can be divided as a collection of cells and the air-mass back trajectories for all cells can be calculated, it is more practical to use this method to quantify remoteness of a limited number of predetermined sampling sites to a given emissions distribution to avoid extensive computations. Von Waldow et al.'s (2010) method, on the other hand, used a simplified global atmospheric circulation model to calculate long-term average circulation patterns at steady-state as the basis of the remoteness index. Von Waldow et al.'s (2010) method, therefore, produces time-independent remoteness values unlike the methods based on air-mass back-trajectory calculations.

Neither the von Waldow et al. (2010) nor the Westgate et al. (2010) approaches to quantify remoteness take into account the properties of specific pollutants. In this study, we propose a method to quantify remoteness from global sources of POPs that considers the multimedia fate and transport properties of chemical pollutants. We use a global multimedia fate model to derive remoteness values of regions covering the entire globe for a set of hypothetical pollutants assuming a representative generic global emissions distribution. Running the fate model in steady-state mode yields long-term average remoteness values similar to the von Waldow et al. (2010) approach. However the global multimedia fate model is capable of dynamic simulations, and therefore, seasonal variations in the remoteness of locations can be analyzed with our method when time-dependent remoteness values are of interest.

Our method of quantifying remoteness is based on the transfer efficiency concept introduced by MacLeod and Mackay (2004). Transfer efficiency is a model-based metric of the contribution of emissions distributed in a defined geographical pattern to the flux of chemical that reaches a specific receptor area. In MacLeod and Mackay's (2004) analysis, the source area was the continent of North America and the receptor area was the Great Lakes. In this study, the whole globe is considered as both the source and the receptor.

This study presents an investigation of the relationship between the multimedia partitioning properties of pollutants and the remoteness of regions from pollution sources. Hypothetical POPs that represent different categories of partitioning properties are used in the analysis. A case study with polybrominated diphenyl ethers (PBDEs) is included as an evaluation of the methodology. The proposed approach for quantifying remoteness allowed us to conduct a critical analysis of the 2-day-half-life-in-air criteria that is used to screen for long-range transport potential in the Stockholm Convention and under various national regulations as it is applied to pollutants with different multimedia partitioning properties.

2. Methods

2.1. Global multimedia fate model – BETR-Global

We used the global multimedia fate model BETR-Global (MacLeod et al., 2011) to calculate the global transfer efficiency values for the hypothetical and real organic chemicals analyzed in this study. BETR-Global is a member of the Berkeley-Trent (BETR) family of spatially resolved multimedia fate models. In BETR models, the environment is conceptualized as a collection of interconnected regional mass-balance models. Each region has seven compartments: upper atmosphere, lower atmosphere, vegetation, freshwater, ocean, soil, and freshwater sediments. The regions are connected by bi-directional flows of air and water between the atmosphere, ocean and fresh water compartments. Chemical mass balance equations for each of the compartments in each of the model regions are defined, and numerical solution of the resulting set of equations provides the spatial and temporal distribution of the simulated chemical.

Since its first introduction (MacLeod et al., 2005), BETR-Global has been applied to analyze many different global-scale chemical pollution problems (e.g. Armitage et al., 2009a; Armitage et al. 2009b; Gusev et al., 2010; Lamon et al., 2009; Li et al., 2015; Wöhrnschimmel et al., 2012). These applications have built confidence in the model structure and assumptions by conducting model evaluations where modeled and measured concentrations of chemicals are compared. Lamon et al.'s (2009) study showed satisfactory agreement between the dynamic model results and long-term monitoring data for globally distributed concentrations of PCB 28 and PCB 153 in air. Wöhrnschimmel et al. (2012) modeled the global fate of α- and β-HCH and evaluated the model performance by comparing the model results with measured concentrations in air and ocean water. Recently, two new software implementations of BETR-Global has been published (MacLeod et al., 2011). BETR-Global 2.0 is coded in Visual Basic for Applications and runs as a Microsoft Excel macro (https://sites.google.com/site/betrglobal). BETR-Research is a re-implementation of BETR-Global in the Python programming language (http://betrs.sourceforge.net). BETR-Research is an open-source project aimed at researchers who would like to modify the model code and add new capabilities to the model according to their specific research interests. Some of the modifications to BETR-Research since its first introduction include increasing the resolution of the global data sets to model the global environment on $3.75^\circ \times 3.75^\circ$ spatial resolution (in addition to the base resolution of $15^\circ \times 15^\circ$), implementing a fast numerical solver, and adding new code to track mass fluxes between compartments. In this study, the newest version of BETR-Research that allows fast simulations with high spatial resolution was used. Code introduced to the model to track mass fluxes between compartments (Wöhrnschimmel et al., 2013) was critical in calculating the transfer efficiency values.

2.2. Quantifying remoteness

Our proposed multimedia remoteness measure is based on the global distribution of transfer efficiency values calculated by the BETR-Research model. The definition of transfer efficiency used in this study is a slight modification of the definition given by MacLeod and Mackay (2004). In this study, transfer efficiency is calculated by dividing the rate of contaminant flux to a selected target environmental compartment (including emissions) by the rate of global emissions:

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