On the Long-Range Atmospheric Transport Behavior of Decabromodiphenyl Ether (BDE-209)

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Objective

The objective of our study was to compare and contrast the observed and predicted long-range atmospheric transport behavior of BDE-209 with that of some other polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs).

Background

Decabromodiphenyl ether (BDE-209) is now the most widely used mixture among the polybrominated diphenyl ethers (PBDEs), but our understanding of the long range atmospheric transport (LRT) behavior of this chemical is still limited. We have therefore applied a dynamic multimedia fate and transport model to gain insight in the various processes that control the LRT behavior of this compound. As past model estimates of LRT have been suffering from limited evaluation against monitoring data, we have also compared our model predictions with observational evidence from dated sediment cores.

Results and Conclusions

The model results suggest that chemicals that are both sorbed to particles and potentially persistent in the atmosphere, such as BDE-209, may have a larger potential for LRT than expected, based on results from past model investigations (7). Specifically, the occurrence of dry periods coinciding with strong winds was found to influence the LRT potential of chemicals that combine a significant affinity for atmospheric particles with sufficiently long atmospheric half-life. Furthermore, the forest filter effect (8) may be important for a wider range of chemicals than believed previously, because models assuming constant precipitation fail to account for the impact of differences in dry deposition on days without rain.

BDE-209 was generally detected only in recent sediment horizons from lakes along a latitudinal transect in North America, and sedimentation fluxes were found to decline exponentially with latitude. The empirical half-distance (EHD) for BDE-209 derived from surface flux data was found to be approximately half that of the ΣPCBs. The observational evidence furthermore highlighted that the model seems to underestimate atmospheric loss processes of potential significance to BDE-209, such as dry deposition of particles, precipitation scavenging, and/or photolysis in the sorbed state. A better characterization of these processes is therefore desirable in order to improve current model predictions for BDE-209.

Details of this study can be found in Environ. Sci. Technol. 2006, 40: 4612-4618.

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Methods

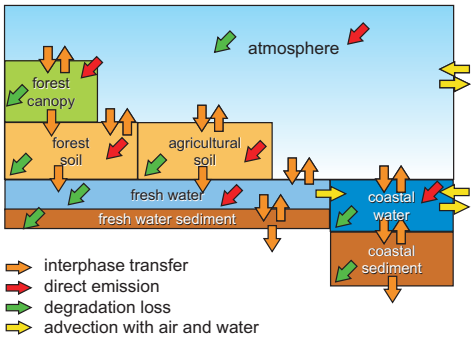

Figure 1: Environmental compartments and bulk processes considered in the original version of CoZMo-POP.

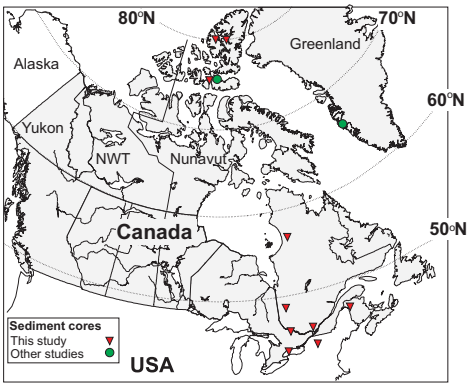

Figure 2: Map of North America showing locations of sediment cores analyzed for PBDEs and PCBs. The locations of cores from two other studies (9,10) are also shown.

Model description

The characteristic travel distance in air (in m) in a model environment, which includes multiple environmental surface compartments in contact with the atmosphere, have been calculated as:

$$L_A = u \bullet M_A / [N_{RA} + \Sigma(N_{ASnet})]$$

where u is the wind speed (m h^{-1}), M_A is the amount of chemical in the atmosphere (moles), N_{RA} is the rate of atmospheric reaction (mol h^{-1}) and $\Sigma(N_{ASnet})$ is the net flux of chemical from the atmosphere to the surface (mol h^{-1}).

L_A was calculated using CoZMo-POP, a non-steady state, non-equilibrium fugacity-based multimedia fate and transport model that has been developed to describe the long-term fate of POP-like substances in a coastal region (*1,2*: Fig. 1). In this work, we used a modified version of CoZMo-POP that includes a dynamic water balance (3). Wind speed, temperatures, atmospheric OH-radical concentration and precipitation rates (as well as L_A) are assumed to vary on a daily basis in the model, but not from year to year.

Sampling and analysis

Sediment cores were collected from selected lakes along a north-south transect from southern Ontario and upper New York state to Ellesmere Island between 1998 and 2001 (Fig. 2). Except for Lake Ontario, all lakes were remote or isolated. Sediment slices were dated using ^{210}Pb and ^{137}Cs and sedimentation rates and dates were estimated using CRS and CIC models (*4*). Sub-samples from cores with interpretable ^{210}Pb profiles were analyzed for a suite of halogenated organics following methods of Muir et al. (*5,6*) with minor modifications to allow for isolation of PBDEs. All sediment sample extraction and cleanup steps were conducted in a certified clean room at NWRI. Depositional fluxes of PBDEs were determined by multiplying sedimentation rates for each horizon by concentrations and dividing by the sediment focusing factor estimated for each core. Where concentrations were <MDLs, fluxes were expressed as <1/2 MDL·sedimentation rate. Empirical half-distances (EHDs) were calculated using PBDE and PCB results for isolated/remote lakes by simple linear regression of log surface flux vs latitude.

Table 2: Empirical half-distances and estimated annual average and median L_A at quasi steady-state for the default scenario [in km]. The daily maximum and minimum values for L_A and the numbers of years simulated to reach quasi steady-state are also presented for the default scenario. Also given is the estimated annual average L_A at quasi steady-state for (a) a scenario without forest compartment, as well as (b) static environmental conditions (assuming steady-state forest growth and decay, wind speed, precipitation, OH-radical concentrations and temperatures based on annual average values), and (c) steady state environmental conditions and no forest compartment.

Chemicals[1]	Empirical half-distances (EHDs)[2]	TaPL3 (L_A)[4]	ELPOS (L_A)[4]	CoZMo-POP (L_A)							
				Non-Steady State environmental conditions						SS environment	
				Default Model Scenario (with forest)					*no forest*	*forest*	*no forest*
				$L_{A,avg}$	$L_{A,median}$	$L_{A,max}$	$L_{A,min}$	Years	$L_{A,avg}$	$L_{A,avg}$	$L_{A,avg}$
PCB-28/31	754±324 (1,088)	4,881	5,233	6,272	4,417	87,070	316	12	8,381	5,040	8,045
PCB-52	895±602 (1,291)	7,411	9,050	7,013	3,535	162,400	340	18	9,484	3,929	7,164
PCB-101	740±202 (1,068)	6,337	16,314	5,115	2,533	90,230	301	48	8,591	2,621	5,488
PCB-118	736±151 (1,062)	3,463	12,739	1,483	1,526	4,322	188	49	3,228	1,346	2,952
PCB-138/163	878±153 (1,267)	2,628	18,364	1,350	1,366	2,408	163	60	3,224	1,229	2,970
PCB-153	846±117 (1,221)	2,843	26,908	1,438	1,490	3,808	181	82	3,389	1,297	2,990
PCB-180	947±281 (1,366)	912	6,889	1,134	1,029	1,917	128	89	2,379	973	2,202
BDE-47	1,168±942[3] (1,685)	1,113	2,483	952	778	1,827	104	14	1,808	758	1,470
BDE-99	-	610	1,194	1,540	961	3,877	118	14	2,902	860	1,440
BDE-209	566±101 (817)	480	735	9,718	4,471	48,020	161	14	22,172	2,658	3,788

[1] *Model calculations do only include the first out of two PCB congeners listed in the same row.*

[2] *EHD = log(0.5)/slope x 112 km/deg. Numbers in parentheses are the ETDs (=EHD / ln(2)).*

[3] *Calculated with 6 lakes, 4 from this study and Lakes DV09 (9) and G (10)*

[4] *Data from Wania and Dugani (7).*

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