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Bayesian Uncertainty Analysis of the Global Dynamics of Persistent Organic Pollutants: Towards Quantifying the Planetary Boundaries for Chemical Pollution

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Abstract—We developed a Bayesian emulator of the Finely-Advanced Transboundary Environmental model (FATE) for persistent organic pollutants (POPs), pseudo-FATE, in order to perform uncertainty and sensitivity analysis of the model. FATE predictions for POPs loads and sinks in the global environments are very sensitive to a large number of prescribed input parameters (e.g., atmospheric and soil degradation rates of POPs). The pseudo-FATE was applied to propagate uncertainties in the selected 21 input parameters through to uncertainty on the FATE predictions on polychlorinated biphenyls (PCBs) #28 and #153 for the period of years 1971–80. Significant uncertainties in the PCB#28 loads occurred sporadically in semi-arid terrestrial regions, while those for PCB#153 were organized in equatorial and sub-tropical Africa, sub-tropical South America, and the high-latitudes, especially the Southern Ocean. Our Bayesian sensitivity analysis suggested that PCB#28 loads appear to be very sensitive to the parameters associated with bioconcentration in grasses, whereas uncertainties in the terrestrial and marine PCB#153 loads could be largely modulated by those in tree functional types parameters, and the turnover time of phytoplankton, respectively. The pseudo-FATE predictions point to the uncertain truth behind POPs dynamics, and will potentially be applied to quantify the recently-proposed Planetary Boundaries for chemical pollution.

Keywords: Bayesian emulator, uncertainty analysis, polychlorinated biphenyls, global dynamics, Finely-Advanced Transboundary Environmental model (FATE)

INTRODUCTION

A number of deterministic computer simulators (numerical models) have been developed to diagnose and predict the behavior of complex, global dynamics of

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persistent organic pollutants (POPs; Wania and Mackay, 1995; Scheringer *et al.*, 2000; Suzuki *et al.*, 2004; Macleod *et al.*, 2005; Lohmann *et al.*, 2006). Finely-Advanced Transboundary Environmental model (FATE; Kawai and Handoh, 2009; Kawai *et al.*, 2009) is one of the most recent developments among them. FATE is forced by POPs emission inventory, land-cover, and climate datasets, and is sensitive to a large number of prescribed input parameters (e.g., degradation rate constants of POPs and plant functional types parameters for bioconcentration processes), which is essentially common to all the POPs models. Each of the parameters has aleatory and epistemic uncertainties. Investigations into their impacts on the model outputs (e.g., atmospheric and terrestrial loads and sinks of POPs) are of primary importance to risk assessments of POPs and other toxic chemicals (Ross and Birnbaum, 2003; van Wijk *et al.*, 2009) and to the recently-proposed Planetary Boundaries for chemical pollution (Rockström *et al.*, 2009). However, this demands a series of uncertainty analyses through a conventional Monte-Carlo method, but FATE is too computing-intensive to make the thousands of model runs that are required for such analyses. Therefore, we must look for a computationally-cheap alternative method.

To this end, we developed a Bayesian emulator of FATE (which we will call "pseudo-FATE"), and then applied Bayesian uncertainty analysis (Kennedy *et al.*, 2008; Conti and O'Hagan, 2010) to propagate uncertainty in the input parameters through to uncertainty on the FATE-predicted dynamics of polychlorinated biphenyls (PCBs) #28 and #153.

MATERIALS AND METHODS

Input parameters

Among all the prescribed input parameters, 21 input parameters defining PCBs properties were selected (Table 1), in order to quantify the degree to which their uncertainties are propagated into uncertainty on the FATE-predicted PCBs loads and sinks in the global atmosphere, oceans, soil, and biosphere (terrestrial vegetation and marine phytoplankton). These parameters include degradation rate constants (Wania and Daly, 2002; Malanichev *et al.*, 2004) and plant/plankton functional type constants (Mclachlan and Horstmann, 1998; Dachs *et al.*, 1999, 2002; Seto and Handoh, 2009). Each parameter value was assumed to obey a Gaussian distribution, the means and variances of which are shown in Table 1. For simplicity, we will use "uncertainty" to represent not only the standard deviation of the mean in the input parameters (i.e., input uncertainty), but also the square root of the mean of total variance in the FATE predictions (i.e., output uncertainty).

Simulations

We have employed yearly PCBs emission inventory datasets (Breivik *et al.*, 2007), climatological monthly mean primary productivity that are estimated from the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) data (Behrenfeld and

Table 1. Uncertainties in the input parameters prescribed in the Finely-Advanced Transboundary Environmental model (FATE). The mean of each parameter is identical to that of the FATE control run, whereas the variance represents a putative uncertainty of the mean.

Prescribed input parameters		Symbols	Units	PCB#28		PCB#153	
				Mean	Variance	Mean	Variance
Degradation rates	Atmosphere (factor)	dA	cm^3/mole/sec	2.7E-10	1.5E-20	8.1E-11	1.3E-21
	Oceans	dO	1/sec	1.3E-07	3.6E-15	1.6E-09	5.2E-19
	Soil	dS	1/sec	7.4E-09	1.1E-17	1.2E-09	2.8E-19
	Vegetation	dV	1/sec	1.5E-08	2.0E-15	2.3E-09	5.1E-17
	Cryosphere	dC	1/sec	1.3E-07	1.6E-13	1.6E-09	2.4E-17
Empirical constants in a scalar transfer velocity	Oceans	aO	—	2.5E+00	8.6E-02	2.5E+00	8.6E-02
	Soil	aS	—	3.0E-01	8.3E-03	3.0E-01	8.3E-03
	Vegetation	aV	—	3.0E-01	8.3E-03	3.0E-01	8.3E-03
Washout rate for wet deposition		W	—	2.0E+05	5.6E+12	2.0E+05	5.6E+12
Leaf turnover time	Grass	TG	year	1.0E+00	2.0E-01	1.0E+00	2.0E-01
	Evergreen broadleaf	TB	year	5.0E+00	7.8E+01	5.0E+00	7.8E+01
	Evergreen needleleaf	TN	year	5.0E+00	8.2E+01	5.0E+00	8.2E+01
	Phytoplankton	TP	day	7.0E+00	6.8E+01	7.0E+00	6.8E+01
Factor (m) and exponent (n) in K_{VA}-K_{OA} conversion	m: Grass	mG	—	2.3E+01	1.1E+02	2.3E+01	1.1E+02
	m: Deciduous forest	mD	—	1.4E+01	6.5E+01	1.4E+01	6.5E+01
	m: Coniferous forest	mC	—	3.8E+01	4.4E+02	3.8E+01	4.4E+02
	n: Grass	nG	—	4.5E-01	3.3E-01	4.5E-01	3.3E-01
	n: Deciduous forest	nD	—	7.6E-01	3.2E-01	7.6E-01	3.2E-01
	n: Coniferous forest	nC	—	6.9E-01	3.3E-01	6.9E-01	3.3E-01
POPs-phytoplankton interface	Uptake rate	Pu	1/day	8.5E-14	6.4E-28	2.4E-12	5.1E-25
	Depuration rate	Pd	1/day	6.5E-01	3.8E-02	1.1E+00	1.0E-01

Falkowski, 1997) and optimally-interpolated sea-surface temperature data (Reynolds *et al.*, 2002), 6-hourly US National Centers for Environmental Prediction (NCEP)—National Center for Atmospheric Research (NCAR) Reanalysis datasets (Kalnay *et al.*, 1996), and simulation outputs of the Ocean General Circulation Model for the Earth Simulator (OFES; Masumoto *et al.*, 2004) in order to generate the FATE control run, for which each of the 21 parameter was set to its mean. The model was spun-up with the climatological daily mean NCEP-NCAR datasets for 17 years, and was then forced by the individual days of the period of years 1948–80; the rest of the forcing datasets were identical to those in the standard FATE setting (Kawai and Handoh, 2009).

Bayesian emulations and analysis

Our Bayesian uncertainty analysis of the PCBs dynamics follows Kennedy *et al.*'s (2008) application to terrestrial carbon fluxes, and is summarized in the following steps: 1) 30 FATE runs, each of which has a specific setting of the 21 input parameters values, were designed by a Maxi–min hypercube method. All the runs were simulated for the period of years 1971–1980, for which the year 1970 of the FATE control run was used as the initial conditions. 2) A Bayesian emulator of the FATE, pseudo-FATE, was constructed for all the FATE grids over the globe, using the results of the aforementioned 30 FATE runs. 3) A Bayesian uncertainty analysis was then applied to account for and to quantify uncertainties in the PCBs loads and sinks that originated from uncertainties in the 21 input parameters.

Given the uncertainties of all the input parameters, a Bayesian sensitivity analysis (Oakley and O'Hagan, 2004), was also performed to quantify the sensitivities of the PCBs loads and sinks to each parameter. This sensitivity analysis will provide new insights into the degree to which parameterizations used in the FATE would influence the results of Bayesian uncertainty analysis. Note, however, that pseudo-FATE does not explicitly reproduce physical parameterizations used in the FATE.

RESULTS AND DISCUSSION

Uncertainties in the POPs loads

The means of surface PCBs#28 and #153 loads predicted by the pseudo-FATE were found to be in good agreement with those of the FATE (not shown). In fact, the FATE predictions over much of the globe fall in the 95% confidence intervals of the pseudo-FATE predictions. This tendency was also confirmed for the rest of PCBs variables (not shown), and thus pseudo-FATE was proven to be an excellent emulator of FATE. The magnitude of quantitative inconsistency between FATE-predicted and observational PCBs concentrations in the atmosphere (Jaward *et al.*, 2004, 2005) and soil (Meijer *et al.*, 2003) are smaller than uncertainties in the pseudo-FATE predictions, which could lend some support to the performance of FATE, provided that pseudo-FATE is a genuinely excellent

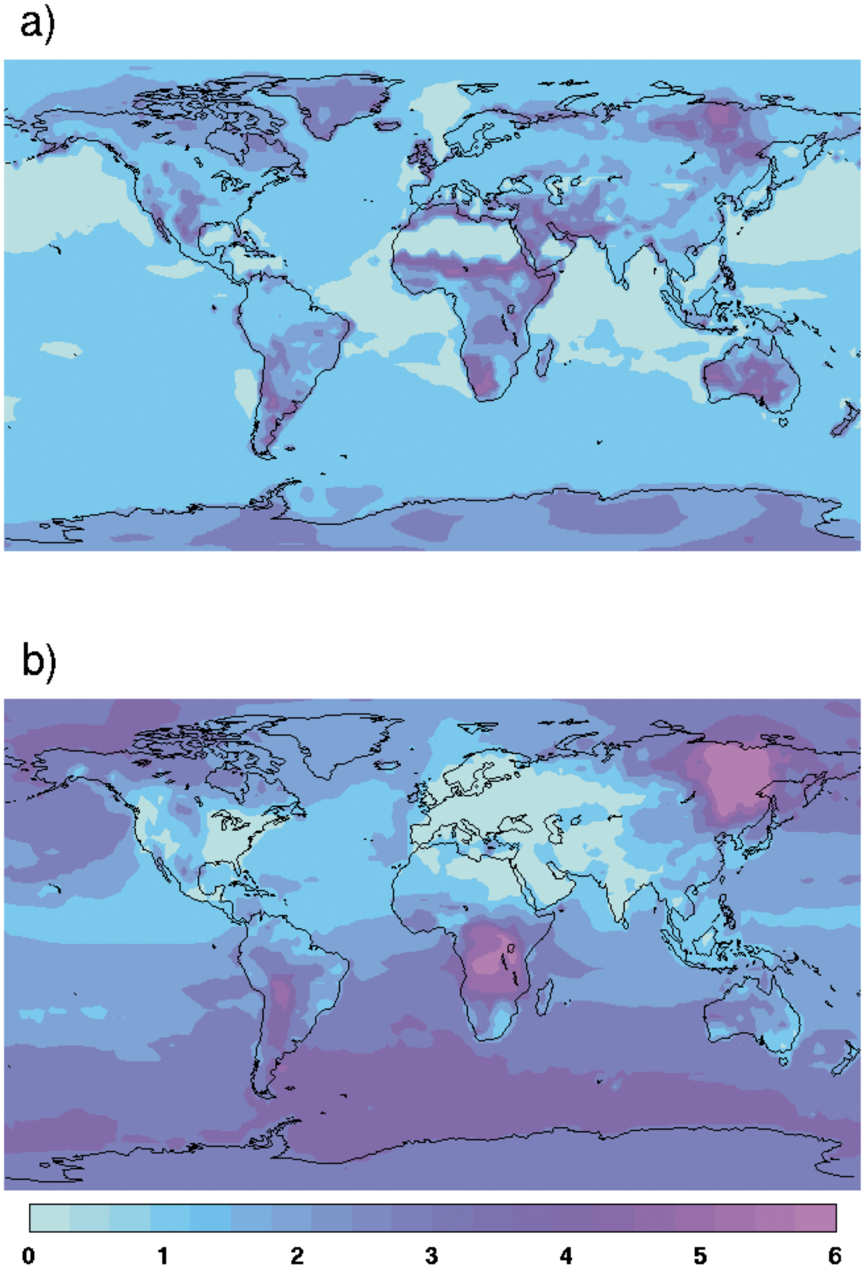

Fig. 1. Predicted uncertainties in the log-transformed annual mean surface loads (log [ng/m^2]) of a) PCB#28 and b) PCB#153 for the year 1980.

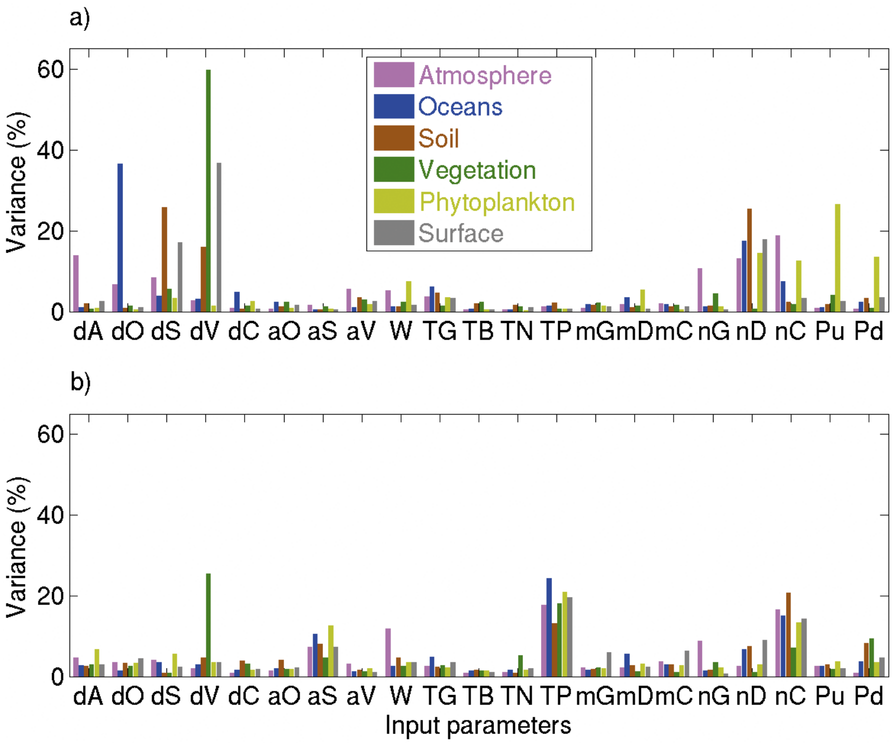

Fig. 2. Relative importance of the selected input parameters. Variances (%) of pseudo-FATE predictions for PCBs loads (atmosphere, oceans, soil, terrestrial vegetation, and surface total) and sink (marine phytoplankton) explained by each input parameter are shown. a) PCB#28 and b) PCB#153 for the year 1980. See Table 1 for the symbols of the input parameters.

emulator of FATE. Hereafter, we discuss the results of our Bayesian uncertainty analysis for annual mean PCBs loads of the year 1980 (Fig. 1).

Uncertainties associated with the pseudo-FATE-predicted loads were systematically larger in PCB#153 than in PCB#28. The difference was up to an order of 1–2. In general, geographical distributions of the uncertainties differed significantly between the PCBs congeners. Although both PCBs exhibited the largest uncertainties over the northern China and central-eastern Siberia, uncertainties in the PCB#28 loads occur sporadically in semi-arid terrestrial regions such as sub-tropical Africa and Australia, while those for PCB#153 were organized in equatorial and sub-tropical Africa, sub-tropical South America, and the high-latitudes, especially the Southern Ocean. It is important to note that there are no significant uncertainties in much of Europe for PCB#153.

The uncertainties over such terrestrial domains could be attributed to those in plant functional type constants. In fact, our Bayesian sensitivity analysis for the influential parameters suggested that PCB#28 load is relatively sensitive to the factor and exponent in K_{VA}-K_{OA} conversion for grasses (mG and nG), whereas uncertainties in the terrestrial PCB#153 load is largely modulated by those in parameters for evergreen needle-leaved forests in Siberia (nC), and deciduous and/or evergreen broad-leaved forests in the equatorial-subtropical Africa and subtropical South America (nD and TB). However, in the global mean, this tendency does not seem to be evident (Fig. 2). PCB#28 sink by marine phytoplankton is found to be relatively sensitive to the POPs uptake and depuration rates (Pu and Pd). By contrast, PCB#153 loads on the oceans appear to be very sensitive to the turnover time of marine phytoplankton (TP), which accounts for the above-mentioned uncertainties in the high-latitudes (Fig. 2b).

Degradation rates of the environmental media directly influence POPs loads and sinks in the media. This evidence is pronounced by PCB#28 sensitivities to the degradation rates, but not so by PCB#153, for which the shorter persistency of the former might be responsible. Noting that the oceans, soil, vegetation and cryosphere interact directly with the atmosphere, we stress that uncertainties in the global surface loads are likely to influence those in the atmospheric PCBs loads, and *vice versa*.

Uncertainties in the emission inventories, land-cover, and climate datasets should not be ignored. For example, wrong configurations of land-cover profiles will, by definition, result in wrong predictions in the POPs loads and sinks through plant functional type constants. Climate datasets such as air temperature, precipitation, and mixed layer depth directly and indirectly modulate wet and dry depositions, surface exchanges, advections and diffusions, and bioconcentration processes of POPs, respectively (Kawai and Handoh, 2009; Kawai *et al.*, 2009). Interestingly, our estimates of uncertainties in the POPs loads due to uncertainties in the input parameters could potentially dominate those associated with emission inventories that have been considered to be the most significant source of uncertainties (Breivik *et al.*, 2007). The uncertainties in the predictions might have been overestimated, however, because uncertainty in each input parameter was prescribed as a putative and plausible maximum estimate.

Contributions to Planetary Boundaries debates

A Bayesian emulator of FATE, pseudo-FATE has enabled us to quantify the propagations of uncertainties in the selected 21 input parameters, for which no more than 30 FATE model runs were required. Our pseudo-FATE predictions point to the uncertain truth behind POPs dynamics. A combination of FATE and pseudo-FATE are very likely to be employed to crudely assess fully-aggregated POPs loads and sinks in the global environment, provided that "virtual POPs", whose multi-variate physicochemical properties and emission statistics obey a set of *ad hoc* Gaussian distributions and characterize all the chemical compounds under the Stockholm Convention on POPs (Hagen and Walls, 2005), are defined. Therefore, our study will potentially be applied to quantify Rockström *et al.*'s (2009) Planetary Boundaries for chemical pollution.

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