FATE OF THE POLYCHLORINATED BIPHENYLS IN THE GLOBAL OCEANS PREDICTED BY THE FINELY-ADVANCED TRANSBOUNDARY ENVIRONMENTAL MODEL (FATE)

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Introduction

Over the last few decades, a number of multi-compartment models have been developed to predict the dynamics of Persistent Organic Pollutants (POPs) (see intercomparison study by Shatalov *et al.*, 2004 [1]). Some of these models were capable of quantifying biogeochemical cycles of POPs in the global environment (*e.g.*, Wania and Mackay, 1995 [2]). While it has been pointed out that the POPs exports to the deep sea by both of the biological (*i.e.*, organic carbon exports) and physical (*i.e.*, overturning circulation) processes play important roles in the sink of POPs from the environment (Daches *et al.*, 2002 [3]; Wania and Daly, 2002 [4]; Lohmann *et al.*, 2006 [5]), these processes have been poorly understood.

The authors have developed a global multi-compartment model for POPs, named FATE (Finely Advanced Transboundary Environmental model; Kawai *et al.*, 2009 [6]; Kawai and Handoh, 2010 [7]; http://www.gfate.iobb.net). The previous version of the model did not incorporate the 3D oceanic transports. In this study, we developed a high resolution ocean compartment of the FATE, and investigated the distributions of PCBs in the global oceans.

Materials and methods

FATE is a high resolution global model, which solves non-steady POPs biogeochemical cycles in/between five environmental compartments (atmosphere, ocean, vegetation, soil, and cryosphere). Except for the oceans, the horizontal resolution of the model is 2.5°×2.5°, and the atmosphere is vertically divided into 10 ů-layers. A newly developed high resolution (0.5°×0.5°×54 layers) ocean compartment which hosts 3D advection processes is replaced to the previous simple box scheme. The processes solved in FATE is the same as previous version (see in detail in Kawai *et al.*, 2009 [6], and Kawai and Handoh 2010 [7]) except for the internal physical processes in the ocean. FATE solves 3D atmospheric advection and diffusion, intra- and inter-compartment abiotic POPs dynamics (*i.e.*, degradations, phase partitions, dry and wet depositions, and interfacial POPs exchange processes), and the bioconcentration in vegetation and marine phytoplankton. The modified Crowley advection scheme (Bott, 1989 [8]; Li and Chang, 1996 [9]) with a 4th-oder polynomial

expansion in the concentration fields was applied commonly to the atmosphere and oceans. This scheme simultaneously solves conservation equations for a scalar and the fluid mass, and therefore, conserves the mass of transported scalar through advection calculations.

FATE is forced by climate data, and the other global inputs (see in detail in Kawai *et al.*, 2009 [6]). NCEP/NCAR reanalysis I 6-hourly data (1948-2007; Kalnay *et al.*, 1996 [10]) and OFES (Ocean general circulation model For the Earth Simulator) outputs (Masumoto *et al.*, 2004 [11]) were, respectively, used for meteorology (velocity, temperature, and precipitation) and for oceanography (velocity and temperature). For the emission, "high scenario" of Breivik's annual PCBs emission inventory (1930-2007; Breivik *et al.*, 2007 [12]) was used.

In this study, we run the FATE for PCB153 for the period of years 1930-2007, and the annual mean outputs for the year 1970 were discussed.

Results and discussion

Figure 1 shows the FATE-predicted annual mean concentrations of PCB153 in the atmosphere and the ocean for the year 1970. Four panels shown in Figure 1 are (a) concentrations at lower part (σ = 0.975) of the atmosphere, and (b, c, and d) concentrations in the ocean. We analyzed concentrations at three depth in the oceans; the surface (8 m) which interacts directly with the atmosphere (Figure 1 (b)), 96 m depth; the depth is characterized by less interaction with the atmosphere, but is biologically productive (Figure 1 (c)), and 1045 m depth, which is the topmost layer of the bathypelagic realm (Figure 1 (d)).

Not surprisingly, the spatial distributions of PCB153 in the surface oceans (8 m) are largely correlated with these of the lower part of the atmosphere. Around the countries with high historical emissions (*i.e.*, USA, Europe, and Japan), areas where PCB153 emitted from such countries, are swept away downwind (*i.e.*, along temperate westerlies). The Arctic showed high concentrations.

The horizontal distributions at 96 m depth are distinctly different from those at the surface. Interestingly, at this depth, PCB153 tended to be accumulated into lower latitudes (subtropics). Although the physical mechanism causing this tendency may differ between the hydrographical sections, the elevated concentrations found in the North Atlantic could be explained by the clockwise subtropical gyre with weak subsidence flow of high-salinity water body originating from the Mediterranean.

At more deep inside the ocean (1045 m), we could see two sections with relatively high concentrations; high latitudes in the North Atlantic and the Weddell Sea, in which deep-water formations such as NADW and AABW occur. The levels of PCB153 concentrations are much lower than those in the upper part of the oceans at this point of the simulation. Note, however, that the amount of deep sea exports by overturning circulation is sensitive to the PCBs degradation rate in the oceans which was prescribed to the FATE; the rate of PCB153 in the deep sea could be much smaller than that in the surface ocean, which may result in the

underestimation of bathypelagic PCBs concentrations.

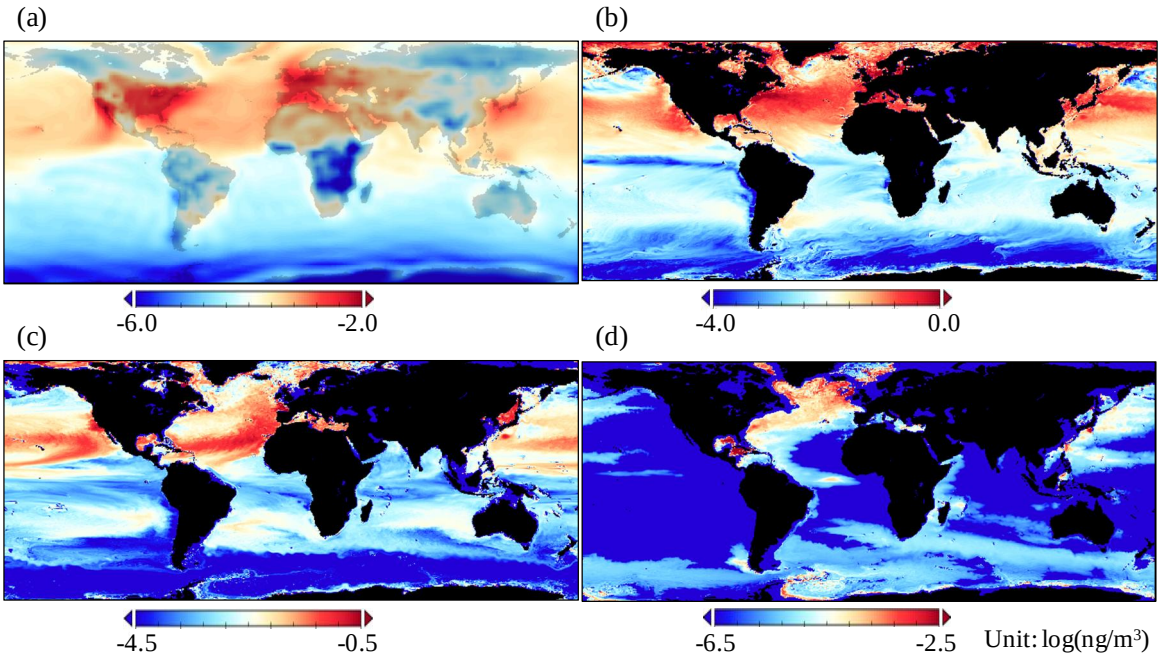

Figure 1: FATE-predicted spatial distributions of the annual mean PCB153 concentrations for the year 1970. (a) the lower part of the atmosphere (σ = 0.975), and for (b), (c), and (d) the selected three depths in the ocean (*ca*. 8m, 96m and 1045m, respectively).

The improved FATE has enabled us to address the distributions of PCB153 in the global oceans. Noting that the availability of validation data is largely limited, the uncertainties in FATE predictions need to be examined. To this end, our group has also applied Bayesian uncertainties and sensitivity analyses to FATE (Handoh and Kawai, 2010 [13]). We believe that combination of more comprehensive modelling of POPs, such as coupling of the marine/terrestrial food web models with FATE, and extensive uncertainty analysis will lead to our better understanding of the full POPs dynamics.

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