HISTORICAL EMISSION INVENTORY AND DYNAMIC FATE MODELLING OF PCBS IN JAPAN

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Introduction

Current environmental levels of PCB are caused by (1) primary anthropogenic emissions from production, use, leak and disposal of technical PCB, (2) environmental recirculation of PCBs emitted in the past and (3) unintentional formation and release of PCBs from thermal processes such as municipal solid waste incineration (MSWI). Determination of the relative importance of the above three sources is a key factor in assessing the effectiveness of risk reduction measures, such as destruction of PCB waste, remediation of contaminated sediments, and improvement of waste incineration. Previous studies successfully estimated the relative contributions of {(1) + (2)} and (3) by conducting a principal component analysis of PCB congener profiles[1] or by conducting a mass balance calculation between the atmospheric emission from MSWI and the deposition flux to the surrounding area[2]. However, the separation of the past emission from the current emission is more challenging[3] because the congener profiles of the two sources are similar. An effective approach to answer this question is a combination of historical emission inventory[4] and dynamic fate modeling[5].

The aim of this paper is to apportion the sources of PCB into the above three source categories. To achieve this, historical emission inventory of 22 PCB congeners released to air and soil in Japan was estimated by a substance flow analysis of PCB. This emission estimate was coupled with dynamic fate modeling of PCB to predict the PCB concentrations in air, water and soil. The predicted concentrations were compared with the observed concentrations to validate the emission estimate.

Methods

In order to estimate the emission of PCBs from production, use, storage and disposal/destruction processes of technical PCB, we used a substance flow model shown in Figure 1. Based on the substance flow model by Breivik[4], we added some modifications to the model: (1) ‘storage of PCB waste’ is introduced as a product life cycle; (2) ‘emission to soil’ is included in addition to emission to air; (3) emission from MSW incineration (PCB as unintentional by-product) is included; (4) congener profile of Kanechlor[6] (KC-300, KC-400, KC-500, KC-600) was used. To evaluate the uncertainty of the model parameters, we used three emission scenarios: Low, Mid, and High.

We used the Mackay Level 4 (dynamic multimedia fate) model to estimate the fate of PCBs emitted into the environment. The model parameters for the fate model are taken from our previous study[7].

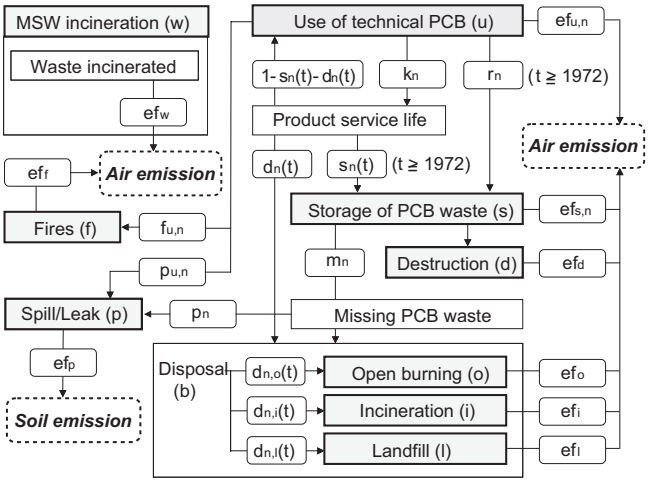

Figure 1: Overview of a substance flow model for estimating PCB emissions.

Results and Discussion

Figure 2 shows the cumulative emission of each PCB congener in terms of (a) emission media and (b) emission sources. The emission source profiles of PCB 126, 169 and 189 were different from the other PCB congeners. This result agrees well with the previous studies[2] which suggested that incineration is a major source of PCB 126, 169 and 189. Figure 2(b) shows that contribution of 'PCB use' is relatively higher for lower chlorinated congeners, reflecting the higher emission factors from this process for PCB congeners with relatively high vapor pressures. For all PCB congeners, emissions from 'appropriate storage of PCB waste' and 'destruction of PCB waste' were negligible.

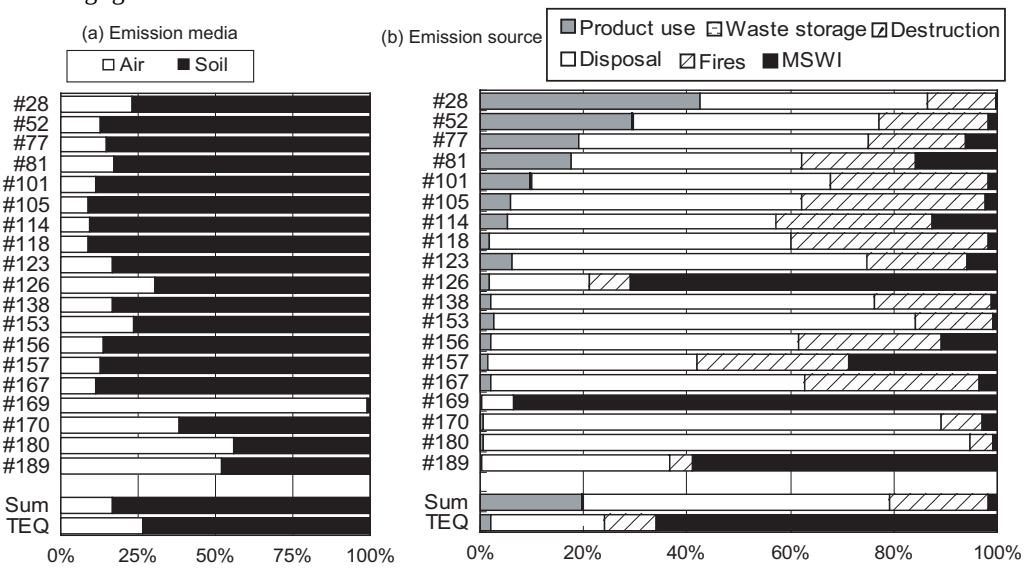

Figure 2: Comparison of air and soil emission of PCB (left) and source composition of the cumulative air emissions of PCB from 1950 to 2002 in Japan (right).

Figure 3 compares the predicted and observed PCB concentrations in air, water and soil. The observed concentrations were taken from national urgent survey on dioxin-like compounds conducted by Environmental Agency of Japan in 1998. The predicted concentrations are those for 'Mid' scenario. For all three media, the predictions were within the range of the observations. The predictions for air and water were close to the median of the observations, while the predictions for soil were about a factor of 10 higher than the median of the observations. One of the reasons for this overestimate is that the applied model dealt soil as one uniform compartment. Separate modeling of landfill soil from the rest of the soil would have resulted in better agreement with the observations. Inclusion of PCB emissions to the water needs to be addressed in our future work. Given the uncertainties in emission estimate and variances in observed concentrations, the simulation of PCB fate in Japan will show satisfactory agreement with monitoring data.

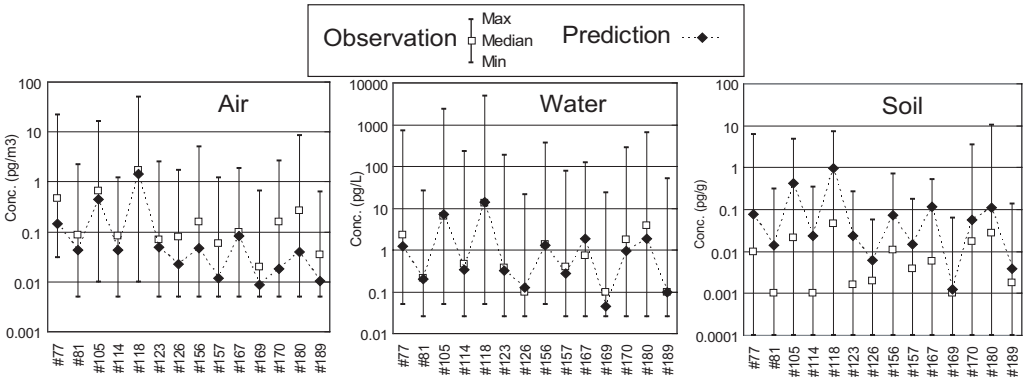

Figure 3: Predicted and observed PCB concentrations in air, water and soil of Japan in 1998.

Figure 4 compares the contribution of MSWI and technical PCB to the amount of PCB in air, water and soil. For PCB 118, technical PCB contributed dominantly to the PCB amount in all three media, while for PCB 126 MSWI source had relatively strong contribution to the amount of PCB especially in air.

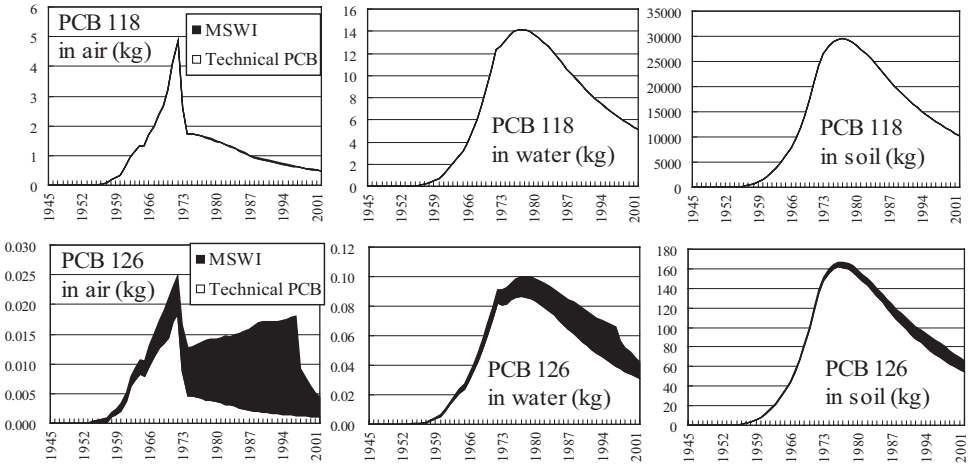

Figure 4: Predicted time-trends of PCB amount in air, water and soil categorized by the sources.

Figure 5 shows the time trend of PCB 118 and PCB 126 amounts in air, water and soil categorized by three emission periods: (a) 1950-1971: before the ban of PCB production in Japan; (b) 1972-1996: before the start of dioxin regulation on MSWI in Japan; and (c) 1997-: after the start of dioxin regulation in Japan. The results show that for both PCB 118 and PCB 126, most of the PCBs present in the current environment were released after the ban of PCB production in 1972. It is also shown that current emission of PCBs (1997-) has strong influence on the PCB amounts in current air, while the past emission of PCBs still has strong influence in current water and soil.

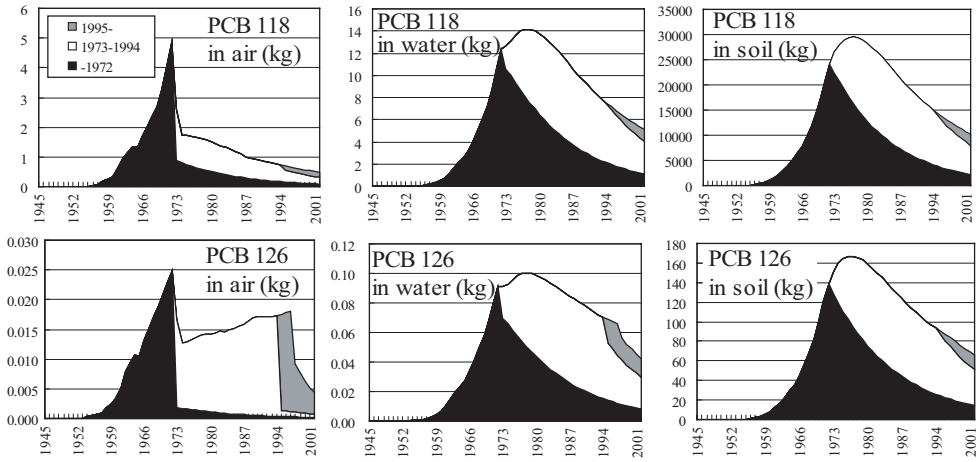

Figure 5: Predicted time-trends of PCB amount in air, water and soil categorized by the periods of PCB emission.

Acknowledgments

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