Transport and fate characteristics of persistent organic chemicals around geo-referenced Japanese environment by spatially-resolved/geo-referenced model (G-CIEMS) methodology

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Introduction

Transport characteristics of the persistent organic pollutants (POPs)[1] have been among the major concerns for the purpose of evaluating their regional and global behaviour. National monitoring program on POPs is now underway in Japan[2-4], however, it is often difficult to interpret the monitoring output in terms of environmental transport of chemicals. Assessment of long-range transport potential (LRTP) by multimedia modelling technologies has been discussed in international framework, and LRTPs derived from generic multimedia models could be effective measures for the assessment of chemicals in terms of transport characteristics[5,6], however, it is still difficult to link model-derived LRTPs to the real geographical distribution of environmental levels.
The authors recently developed a geo-referenced multimedia environmental fate model (G-CIEMS) on the surrounding area of Japan, which can simulate the multimedia fate and transport processes of the chemicals in the spatially-resolved environment[7,8].
The aim of this study is to evaluate the transport characteristics of the chemicals in the realistic geographical distribution by (a) deposition characteristics in the real land-ocean compartments, and (b) comparison of expected site-specific levels and transport characteristics.

Methods and Materials

G-CIEMS (ver.2) model and modelling domain of this study: The G-CIEMS (ver.2) model is a multimedia fate model with spatial resolution, on the geo-referenced GIS (geographical information system) environment. The model compartment consists of air (grid cell structure with 4 vertical layers), river and lake (geo-referenced river network structure), soil (catchment polygon with 7 land-use categories), sea (polygonal area with vertical 4 layers), and sediments beneath all aquatic media. Figure 1 shows the outline of schematic diagram of the model[7]. Area of this study is shown in Figure 2, and the summary of modelling domain is shown in Table 1. Model parameterization and validation has been performed for dioxin and several chemicals[8]. Steady-state solution was employed in this simulation.

Target compounds, emission estimation and CTD (Characteristic Travel Distance): Target compounds in this study were; hexachlorobenzene (HCB), PCB #28 (PCB-28), tetrabromodiphenylether (TeBDE), and pentachloro-dibenzofuran (PeCDF). Emission estimation for HCB, PCB-28, and TeBDE was assumed to be a hypothetical unit emission only from Japan. The total unit emission was allocated into 5x5 km grid resolution by using the population as allocating factor. This means that emission of those 4 compounds was same for both total amount and geographical distribution, but only physico-chemical properties were different.
Emission estimation for PeCDF was based on the real dioxin emission inventory[9] in Japan, but total amount of emission was normalized to be a unit amount as the other compounds. Geographical pattern of the emission in the existing emission inventory was used as the allocating factor of the unit emission. Current emission from air was used for all compounds in this study, as the major objective of this study is to show the geographical distribution of air levels at the steady-state condition. CTDs from generic ELPOS model[10] were calculated by using identical chemical properties but with default environmental setup in the model.

Methods and measures of the simulation outputs: Vapour pressure values used in the simulation were 0.24, 0.09, 0.00013, and 0.000017 Pa for HCB, PCB-28, TeBDE and PeCDF, respectively. Multimedia environmental concentration was calculated in this study, however, only the air concentration was discussed in later section. Air levels of simulation output were all expressed as the normalized levels, by putting the maximum air levels within simulation area set to 1, for the purpose of inter-chemical comparison.
Mass-balance calculation at the two boundary lines A and B in the modelling domain (shown in Figure 2) was made using the model output. Line A in Figure 2 is the boundary of the "region A" area covered by the 5x5 km grids of air, which covers the entire terrestrial region of Japan. Model output in this region can be considered as the geo-referenced, realistic information. Line B is the boundary of the "region B" area covered by the 80x100 km grids of air, which covers the mainly oceanic, and land area other than Japan's islands. The net mass fluxes across the lines A and B, which are designated as the outward fluxes from regions A and B, respectively, were calculated from the model output. The outward fluxes from region B can be essentially comparable to those obtained from the generic models, because of the lack of information of land area other than Japan's islands and detailed ocean information for region B at this moment.
For the purpose to estimate the impact of rain on the transport, cases by arbitrary setting rainfall of all area to zero were also calculated.

Results and Discussion

Geographical distribution of air levels, deposition characteristics, and outward fluxes from the region: Geographical distribution of normalized air levels from model simulation are shown in Figure 3 for HCB. Figure 4 shows the simulated relative proportion of the mass fluxes removed from air based on the constant-rain assumption. Figure 4 also shows the relative proportion of area in the modelling domain. Figure 4 shows that roughly 60% of TeBDE and PeCDF are removed from air by the deposition onto soil compartment, although soil area occupies only 7% in the region. Direct deposition of the two compounds onto oceanic area is nearly 40%; however, the proportion is significantly less than expected from the geographical proportion of the oceanic area (93%), because of the significant differences of air concentration among locations. Ratios between

removal fluxes onto soil and ocean for PCB-28 and HCB are both comparable to the ratios between geographic areas of land and ocean, although the contribution from reaction loss is different between two compounds. This discussion suggests that transport characteristics of less-volatile compounds like TeBDE and PeCDF in this region is still significantly controlled by the runoff from terrestrial area after deposition onto soil compartment, although the land area of Japan's island is relatively small than the oceanic area.

Table 2 shows the simulated outward net fluxes from regions A and B, calculated from the G-CIEMS simulation. Net outward fluxes from regions A and B were significantly different among compounds. Majority of HCB and PCB-28 is shown to be transported across both the A and B lines, whereas the minor part of TeBDE and PeCDF is shown to be transported across the same lines. However, when rainfall was set to zero, the transport characteristics of the latter two compounds were significantly enhanced, although those of HCB and PCB-28 were nearly unchanged. The significant impact of rain assumption on the transport nature of less-volatile compounds has apparent effect on the discussions above, especially on the relative proportion of removal mass fluxes. This point should be studied further in future study.

Relation among simulated levels at sites 1 to 5, outward fluxes and CTDs: Sites 1 to 5, which are shown in Figure 3, are arbitrarily selected to represent the northern (1), eastern (2), western (3), southern (4), and relatively remote (5) edge of simulated geographical distribution of air concentration shown in Figure 3. As the all sites locate on the ocean without any apparent emission sources, contaminants obtained at these sites should be attributed to the transport from sources on the Japan's islands in this simulation. This means that the relative level of normalized concentration at each site should reflect the relative transport characteristics of the compound among the all target compounds.

Figure 5 shows the relation of relative air levels for 4 compounds at sites 1 to 5 and the CTDs to the outward fluxes from region A. All values were normalized to 1 at the maximum, which always appears on HCB cases. As shown in Figure 5, relative air levels at site 5 show almost linear relationship to relative proportion of the outward fluxes from region A. On the other hand, relative air levels for sites 1 to 3 are roughly constant for all compounds. Relative air levels at site 4 show intermediate level of relationship to the outward fluxes from region A. The CTDs show consistent tendency to the outward fluxes from region A, however the direct comparison to the site observations should be difficult, because of the poor quantitative relation to the outward fluxes calculated by the geo-referenced model output. Figures 3 and 4 suggest that the main plume of contaminants is simulated to go south-eastern direction from the sources on Japan's islands. The good correlation of the outward fluxes from the area and the expected relative levels at site 5 can be attributed to the characteristic location and distance from the sources on this site, which may give implication to the selection of monitoring site locations. Another implication from this discussion is that the possible observation at the sites other than site 5 is not very sensitive to the emission from Japan's island, and possible observation at those sites may be attributed to other sources and/or mechanisms that are not considered in this study.

Expansion of the model to the east-Asian region is now planned. By combining the information from LRTPs, monitoring-based estimation, and the geo-referenced modelling approach, we believe that transport and distribution of the chemicals will be assessed in a more realistic way.

References

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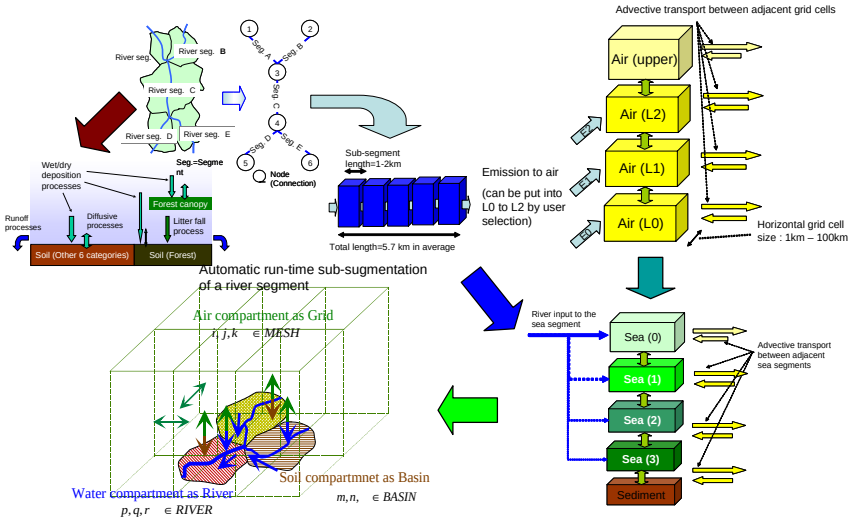

Figure 1. Schematic diagram of the G-CIEMS (ver.2) model

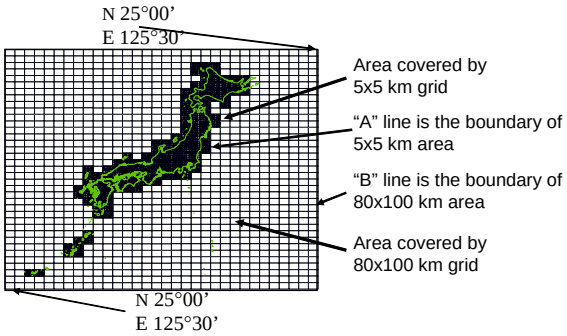

Figure 2. Simulation area and "A" and "B" lines around region A and B, for the flux calculation

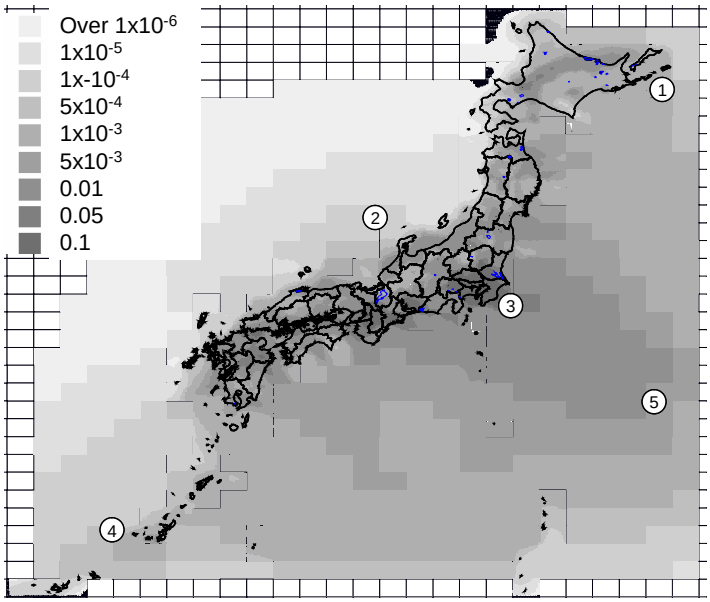

Figure 3. Estimated relative levels of HCB in air from Japanese sources, by the normalized level at the highest level set to 1. Sites 1-5 are also shown in the figure.

Table 1. Summary of model domain in this study

Compartment	Description
General	Rectangular area between 22° 30' E to 149° 30' E and 22° 30' N to 46° 40' N, including all of the Japanese islands, a generic sea area
Air	Grid cell: 5 km by 5 km for Japanese terrestrial area (about 40,000 grid cells) and 100 km by 80 km for oceanic area (about 7000 grid cells) area[a]
River, lakes, and soil	Total of 38,000 catchments, consisting of river path and catchment area.
Coastal sea	About 300 polygons for all coastal area of Japan's islands

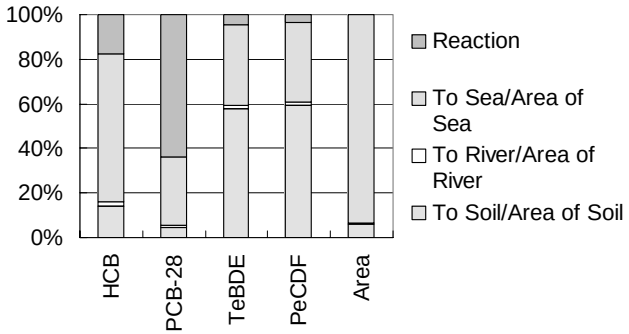

Figure 4. Relative proportion of mass fluxes removed from air and relative proportion of area in the geographical domain.

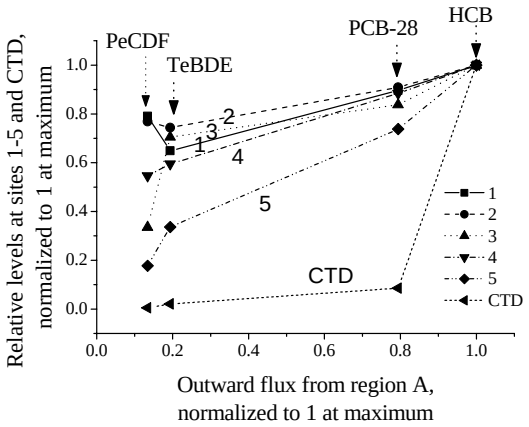

Figure 5. Comparison of relative levels of 4 compounds at sites 1-5 for air concentration, CTD and outward fluxs from region A. All values are normalized to 1 at the maximum.

Table 2. Ratio of net fluxes outward from the region A and B, with and without rain.

	With constant rain				*Without rain*			
	HCB	PCB-28	TeBDE	PeCDF	HCB	PCB-28	TeBDE	PeCDF
Region A	0.97	0.77	0.19	0.13	0.97	0.78	0.51	0.45
Region B	0.97	0.68	0.10	0.040	0.96	0.70	0.41	0.30
Total losses	0.045	0.32	0.90	0.96	0.042	0.30	0.59	0.70

*Values are the ration by total emission normalized to 1.