ATMOSPHERIC CONCENTRATION AND DEPOSITION OF DIOXINS IN THE KANTO REGION OF JAPAN

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Introduction

The emission sources of some species of hazardous chemical pollutants, including dioxins, are often located in densely populated, urban environments. In Japan, waste incinerators are the main source of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). Many small-scale waste incinerators exist in densely populated regions, since much of the waste in such areas is incinerated. Therefore, to evaluate exposure from two or more sources, a grid-type model is necessary.

The distribution of atmospheric concentrations of dioxins, as well as their depositions, were estimated by using the fate model. The model was applied to the entire Kanto region (including the Tokyo metropolitan area) of Japan. The area has more than 30 million inhabitants and more than 200 incineration plants. An analytical domain with a scale of 276 x 224 km was discretized into 40 x 60 grids with about 5-km increments. Figure 1 shows the location and analytical domain used in the simulation.

Materials and Methods

Model overview

The model was designed to estimate a concentration distribution with 5 × 5 km of spatial resolution and a time resolution of monthly averages. Therefore, it was not intended to estimate changes in concentration within a short time or narrow space (e.g., from an accidental discharge). Chronic exposure to chemical substances has adverse effects on the human body and the environment. Persistent pollutants could be transported over wide areas. Therefore, it is necessary to estimate the long-term average concentration of substances that have been continuously discharged over a wide region.

The two-dimensional Gaussian plume and puff models, which assume a uniform concentration below a mixing layer, were adopted as the dispersion process in the simulation model. The distribution of concentration was estimated by overlapping the contributions from all the source meshes onto all the meshes in the analytical domain. The model also included the process of dry and wet depositions to other media (soil and water), dissipation into the upper atmosphere, and degradation. The diffusion calculation was conducted in each classified meteorological condition in order to evaluate long-term averages. The classified meteorological conditions used to estimate concentrations and depositions were set as follows.

1. *Wind direction (16 directions)*
2. Wind velocity (5 categories: 0 -<1, 1-<3, 3-<5, 5-<8, > = 8 m/sec)
3. Atmospheric stability (3 categories: unstable (A,B,C), neutral (D), stable (E,F,G))
4. Time zone (6 categories: 4-hour intervals per day)

The model was validated by comparing calculated and observed trichloroethylene and tetrachloroethylene concentrations in the atmosphere. The observed values were obtained from a monitoring survey conducted over several weeks in summer and winter in the Kanto region. The model is capable of estimating the long-term (e.g., monthly) average concentration distribution of chemicals over a wide, flat area such as the Kanto plain[1].

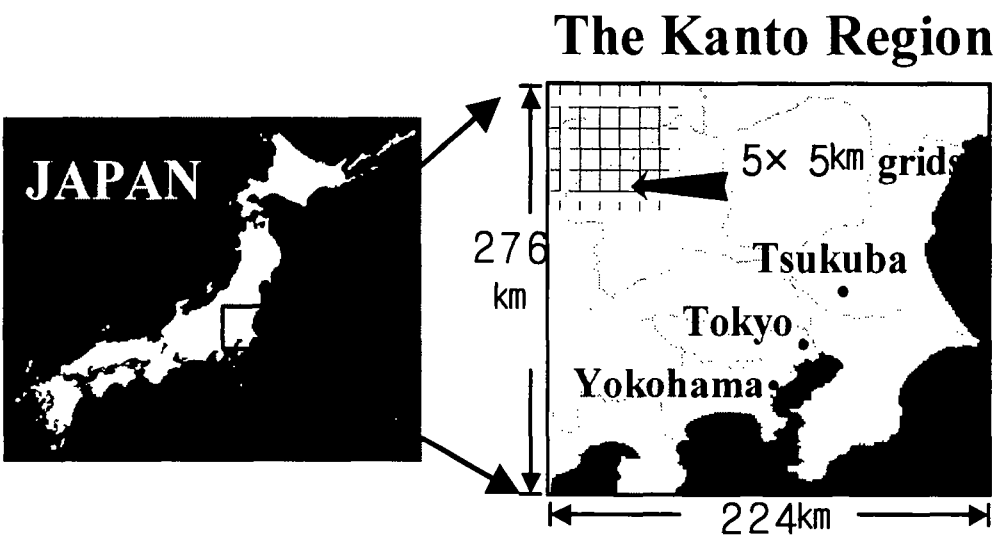

Fig. 1: Location and analytical domain used in the simulation.

Input data and calculation conditions

Data from Japan's Automated Meteorological Data Acquisition System (AMeDAS) were used in the simulation as meteorological input data (i.e., wind velocity and direction, precipitation). Calculation was conducted through a year using meteorological data collected in 1997, and the result was used to evaluate the annual average concentration.

The parameter values and conditions used by the simulation are shown in Table 1.

Emission inventory

The gridded emission inventory of PCDDs/PCDFs in the analytical domain was compiled based on published monitoring data on exhaust gas[6], the amount of municipal solid waste (MSW)[7], and the location of each municipal incineration plant[8]. The monitoring data for the simulation were collected in 1997. The data would change in a later survey. Sources other than MSW incineration (emission from industrial waste and medical waste incineration, industrial processes, exhaust gas of automobiles, impurities in PCP, etc.) were not estimated because they had many uncertainty factors. PCDDs/PCDFs emissions were converted to the emission of 2,3,7,8-TCDD equivalent (TEQ) using a toxicity equivalent factor (TEF), and environmental transport calculations were conducted for 2,3,7,8-TCDD. It was assumed that all dioxins/furans behave in the environment in the same manner as 2,3,7,8-TCDD.

Table 1: Parameters used for the simulation.

Parameters	Values	Reference
Evaluation Period	1 year	
Grid Size	5 x 5 km	
Time Resolution	Monthly average	
Dry deposition rate	0.19 (cm/sec)	Carolyn J.K et al.[2]
Wet deposition rate	50000 (-)	Carolyn J.K et al.[2] Bralan D.E. et al.[3]
Half lives in air	0.34	Howard, P.H. et al[4]
Dissipation rate to upper mixing K_t	5×10^{-5}	Markey, D. et al[5]

Results and Discussion

Atmospheric concentration

The spatial distribution of the concentration of PCDDs/PCDFs in the atmosphere is shown in Figure 2. Highly polluted areas include the middle of the Kanto plain (around the border between Tokyo and Saitama Prefecture), the grid including the city of Choshi, and the grid including the city of Kimitsu. The pollution in the middle of the Kanto plain was due to emission and meteorological factors. Many incineration plants exist in this area. Although the concentration of dioxins in the stack flow was low in this area, the large amount of waste incinerated and the large number of incinerators made the concentration of dioxins high. As a meteorological factor, stagnation often occurs around this region, particularly during winter, because of the influences of land and sea breezes. Both Choshi and Kimitsu have incineration plants that emit high concentrations of dioxins. However, because the plants in these two cities were greatly improved in the next year's survey, the concentrations today seem low.

The simulated maximum concentration in the atmosphere is 0.4 pg-TEQ/m^3, which is lower than the measured environmental levels in Japan[9] and is also lower than 0.6 pg-TEQ/m^3, the environmental quality standard in Japan. This result is acceptable since the simulation did not take sources other than municipal waste incineration into account.

Deposition amount

Figure 3 shows the distribution of dry and wet depositions in the Kanto region. This distribution generally reflected the distribution in the atmosphere described previously. The total deposition amount was estimated at 93 gTEQ/y throughout the region; 88 gTEQ/y deposited on the land (including rivers, lakes, and marshes, and 5 gTEQ/y on Tokyo Bay. Total wet and dry deposition amounts in the region were estimated at 47 and 46 gTEQ/y, respectively.

Since the total amount of emission was estimated at 710 gTEQ/y in the region, 13% of discharged substances were deposited within the domain including Tokyo Bay. The other 87% remained in the atmosphere of the domain or disappeared by decomposition or outflow outside the domain.

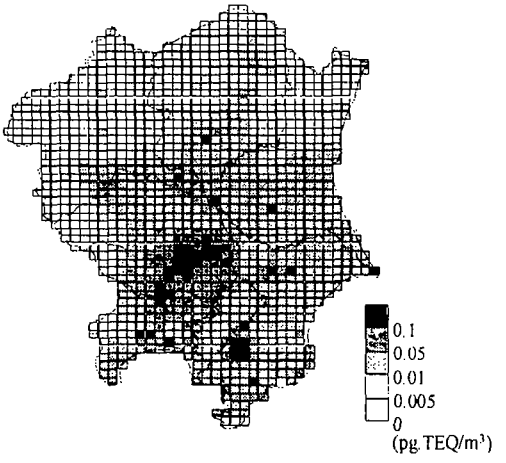

Fig. 2: Estimated concentration of PCDDs/PCDFs in the atmosphere over the Kanto area.

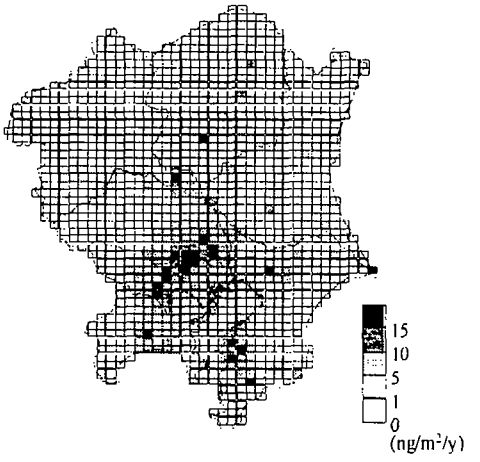

Fig. 3: Estimated deposition of PCDDs/PCDFs over the Kanto area.

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Table. Isomer composition of sludge samples from sludge pits of State Unitary Enterprise "Ufachimprom", ng/g.

Isomers	6	5			4		
PCDD/PCDF	0-30 cm	0-5 cm	50 cm	150cm	0-5 cm	50 cm	150 cm
2,3,7,8-TCDD	3.56	21.9	8.15	17.76	1.01	6.56	56.39
1,2,3,7,8-PeCDD	16.64	7.28	25.04	20.97	< d.l.*	< d.l.	< d.l.
1,2,3,4,7,8-HxCDD	1.46	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.
1,2,3,6,7,8-HxCDD	9.02	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.
1,2,3,7,8,9-HxCDD	2.58	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.
1,2,3,4,6,7,8-HpCDD	28.6	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.
OCDD	58.6	294.8	664.7	567.0	220.5	2234.3	2258.3
2,3,7,8-TCDF	1.08	175.46	96.01	181.46	24.14	68.58	100.46
1,2,3,7,8-PeCDF	1.12	3.59	6.94	11.33	< d.l.	< d.l.	< d.l.
2,3,4,7,8- PeCDF	1.37	4.62	5.13	9.09	< d.l.	< d.l.	< d.l.
1,2,3,4,7,8- HxCDF	1.62	4.94	1.68	6.57	< d.l.	< d.l.	< d.l.
1,2,3,6,7,8- HxCDF	2.39	3.85	2.46	5.96	< d.l.	< d.l.	< d.l.
1,2,3,7,8,9-HxCDF	1.04	4.78	1.94	6.89	< d.l.	< d.l.	< d.l.
2,3,4,6,7,8- HxCDF	2.28	4.37	1.76	4.43	< d.l.	< d.l.	< d.l.
1,2,3,4,6,7,8- HpCDF	11.9	18.4	39.2	48.7	11.7	16.8	23.9
1,2,3,4,7,8,9- HpCDF	2.4	23.7	34.9	47.4	10.3	26.6	32.6
OCDF	29.2	536.1	756.8	637.8	125.6	1247.5	1645.8
TEQ, ng/g.	15.3	45.3	33.8	51.9	3.9	17.3	70.9

Note: *d.l. – detection limit of 0.1 ng for TCDD/TCDF; 10 ng for hepta- and octa- isomers.