微量化学物質の環境循環モデルを用いたダイオキシンの挙動解析

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1. Introduction

The international environment protection policy is to identify and control the sources of some chlorinated organic compounds including polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), often known collectively as dioxins. A major issue associated with emissions of PCDD/F is the zone of influence of major emission sources, in other words, whether the effects of the dioxin source are contained to a local area with potentially high PCDD/F concentrations, or whether they are diffused over a large region with overall dilute PCDD/F concentrations. However, because there have been few studies on the atmospheric fate and long-range transport of dioxins, there is not sufficient information to resolve this matter. In order to understand the nature of dioxin transport, it is important to know whether PCDD/F deposition at a given location is the result of local sources or distant regional/global sources. It is also important to characterize the movement of dioxins in the environment and the locations of potential reservoir sources.

We present here analysis and comparison of the mixture PCDD/F concentrations in order to understand the phenomenon of long-range transport. In the literature of environmental science, data on the particular subject of dioxin transport are lacking, and there exist significant limitations in the analytical methodologies to assess the relative fractions of emitted PCDD/F that are deposited locally versus transported at regional or global scales. Although we studied the same class of chemicals in each location, individual environmental circumstances must be taken into consideration in order to identify the major factors describing the general case of long-range dioxin transport. Considering the numerical models that predict distribution for each medium and attenuation of dioxin transport in each medium of dioxins are required for risk assessment, for the purposes of this study, the actual medium level of distributions for each medium was investigated as the first step. The novel element in our initial investigation in this paper is the new parameter called the compartment distribution coefficient (D_C). It is believed that D_C has significant potential to improve the predictive ability of resulting models for dioxin transport. An accurate model might identify districts at high risk for dioxin contamination caused by long-range transport. Such foreknowledge might allow for the possibility of informed intervention to stem global diffusion of dioxins.

2. Data and Methods

2.1 Data sources

Data on the levels of PCDD/Fs in air, water, and land, were gathered from different cities in Japan, the United States, the United Kingdom, and Australia. The conditions in these locations differ in terms of temperature, precipitation, wind, and complexity of the local terrain. Databases, which are available for public use from national authorities, Environmental Protection Agency (EPA), the European Commission, and international organizations, were exploited to the fullest extent necessary for the purposes of the study. Furthermore, the analysis was also applied to the statistical data summarized by Rainer Lohmann and Kevin C. Jones.

2.2 Theory and Methodology

Only official statistical information and data from experimental investigations on the levels of PCDD/Fs in air, water, and land, were used for calculating the estimates of PCDD/F potential tendency. Various sources and methods were used for the estimation of emission factors. We have proposed a new parameter to describe the multimedia distributing situations of PCDD/Fs. The new parameter, entitled the compartment distribution coefficient (D_C), can be described quantitatively with the following equation:

$$D_C = TEQ_C / TEQ_T$$

Where TEQ_C is the WHO-TEQ levels of PCDD/F concentration in different compartments including air, water, and soil. TEQ_T is the total WHO-TEQ levels of PCDD/Fs obtained from the basis on the generation source discharge data of dioxin in each country. The subscript of D_C signifies the general compartment. Therefore, D_A, D_W, and D_S represent the compartment distributing coefficient of PCDD/Fs in air, water, and soil, separately. For each location, estimates of D_C were made for the maximum possible to identify the transport tendency of PCDD/Fs.

2.3 Procedure

1. We collected the statistical data of the levels of PCDD/Fs in air, water, and land, for the selected locations.
2. We calculated the values of D_C for each compartment.
3. We identified estimates of the potential reservoir sources of dioxins.
4. We will establish the relationship models between the D_C values and Physico-chemical properties of PCDD/Fs.
5. We will perform more accurate quantitative estimates of the levels of pollution emitted by the potential reservoir sources of dioxins.

3. Results and Discussion

3.1 The levels of PCDD/F mixtures in the environment

There is interest in the mixture of PCDD/Fs present in air, water, and soil, as these levels will be related to intensity of source inputs and weathering processes. It has often been reported that there is a quite consistent homologue pattern of PCDD/Fs in air, except close to important local sources (Hippelein M et al., 1996; Jonws KC and Duarte-Davidson, 1997). Overall summaries of mixture PCDD/Fs for Japan, the U. S., the U. K., and Australia are shown in Table 1.

3.2 Comments on D_C

Atmospheric transport is the primary distribution pathway moving PCDD/Fs from atmospheric emission sources toward deposition to terrestrial and aquatic ecosystems. There is evidence that atmospheric source reductions have been occurring over the past couple of decades. However, there are still considerable uncertainties about the relative importance of atmospheric sources of PCDD/Fs and their fluxes into the environment in terms of their contribution to overall environmental pollution. According to literature, the level of soil repository of dioxin had built up to a point such that it could provide a secondary source of PCDD/Fs back to the atmosphere as initial atmospheric levels declined. As a result, there is much interest in the long-range fate of PCDD/Fs in the environment. Indeed, the long-range dynamics are governed by different physical, chemical, and biologically mediated loss processes.

In order to understand the long-range transportation mode of PCDD/Fs in the different environmental compartments, the concentrations measured at each location were converted to those values of D_C shown in Table 1. Due to much information included in it, D_C can be regarded as the result of a combination of geologic, topographic, hydrologic, and climatic factors, as well as historical modifications and changes, both natural and manmade. Because the accurate of future predictions of the long-range transport of contaminants is dependent by the uncertainties of the variables in any current model, it is crucial that careful and thorough site characterization is conducted now.

From the data shown in Table 1, a difference was observed between the values for D_A, D_W, and D_S in the same location. According to the observed relationship of $D_A < D_W < D_S$, we can conclude that it is more easier for PCDD/Fs to have the greatest tendency to remain in the soil. Difference values of D_C were also found in the same compartment. The lower D_A values were found in the U. S., the bigger D_A values were found in Japan and U. K., and D_A value found in Australia was higher than those in America cities were. The usefulness of this analytical method is in its ability to identify locations of potential reservoir sources without having to identify the subtle and non-obvious site characteristics that make them high risk. For example, it might be expected that Australia cities would have lower D_A values than those in America cities

because of less industry. As seen from Table 1, however, this is not the case. Such a finding may indicate that the reported spot in Australia is a potential reservoir source.

3.3 Identification on the generation source of PCDD/Fs

As indicated by the data in related studies, there are several factors that could potentially affect or control atmospheric PCDD/F concentrations. We concluded that current background levels of PCDD/Fs could be classified according to three sources: the natural background, the local emission and regional transportation. It is assumed that the value of D_C is stable at a particular location under normal circumstances. If this assumption is valid, then we can identify the generation source of PCDD/Fs by calculating the on-line ratio (D_{CQ}). D_{CQ} is calculated through the similar method to the calculation of D_C; that is, in order to obtain D_{CQ}, the on-line WHO-TEQ level of PCDD/F concentration in each compartment is divided by the total WHO-TEQ levels. If the ratio (D_{CQ}/D_C) greatly exceed one, it suggests that there is PCDD/F contamination. If the possibilities of contamination by natural causes or from human activities are eliminated, then there is a strong likelihood that long-range transport is occurring at this location. If the ratio of D_{CQ}/D_C were observed to keep on the tendency of exceeding one for a long time, it should be an alarm for us to take measurement to control dioxin contamination.

3.4 Concluding remarks and outlook

With the application of D_C, it was found that PCDD/Fs have the greatest tendency to remain in the soil. D_C values are different from each other because of different transportation modes or site characteristics. Smaller D_A values were found in the U. S., bigger D_A values were found in Japan and the U. K., while a higher than expected D_A value was found in Australia. This result may indicate that this location in Australia is a potential reservoir source of dioxins in the future.

Through analysis and comparison of all of the available data using the parameter D_C, the quantities of dioxins due to natural background, long-range transportation, and current background may be calculated. Through this method using the ratio (D_{CQ}/D_C), it is possible to predict the reservoir sources of dioxins without requiring exhaustive source characterizations of the PCDD/F mixture or detailed knowledge of the environmental fate. Indeed, the new parameter D_C proposed in this paper may offer a shortcut to the identification of reservoir sources and offer clues to the timing and magnitude of dioxin transport. Such information may then expedite subsequent efforts to characterize the key processes underlying dioxin contamination and exposure pathways. Although there remain many uncertainties and data gaps in the research of dioxin assessment, Preliminary results suggest that strategies to manage dioxins should be improved.

In order to test the predictive ability of the new parameter D_C, it is necessary to undertake in-depth studies of the fate and behavior of PCDD/Fs in the environment. Several areas of future research need to be addressed for the purpose. These include the practice of the theory of D_C in the context of the individual classes of PCDD/Fs, the establishment of a quantitative structure activity relationship model to predict D_C, a multimedia simulation to understand the transportation mode, and the identification of districts at high risk for contamination due to long-range transport of dioxins.

References

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Table 1 Levels of PCDD/F mixture in the environment and the Calculation values of compartment distributing coefficient D_C for different cities

Country	Location	Comment	TEQ			ΣTEQ [b] (gI-TEQ/y)	Ratio D_C [c]			Year
			Air (pg/m^3)	Water (pg/L)	Soil (pg/g)		$-\log D_A$	$-\log D_W$	$-\log D_S$	
Japan[a]	Nishiwaki	Urban	0.27		2.1	230	2.930		2.040	2000
	Neyagawa	Urban	0.22	1.0			3.019	2.362		2001-2002
	Kobe	Urban	0.12	0.98	1.2		3.283	2.371	2.283	2000
	Nagoya	Urban	0.21	0.48	1.0		3.040	2.680	2.362	2000-2001
US	Phoenix, AZ	Urban	0.25			2921	4.068			1996
	Ohio	Urban	0.081				4.557			1995
	Bloomington	Rural	0.033				4.947			1986-1989
	Wisconsin	Rural	0.058				4.702			1989
	Connecticut	Urban	0.11				4.424			1987
UK	Manchester	Urban	0.41			480	3.068			1991-1993
	Cardiff	Urban	0.19				3.402			1992-1993
Australia	Brisbane	Rural	0.011			16	3.163			1996

Note: In general, most mean values are shown in order to describe the Levels of PCDD/F mixture in the environment for different cities summarized by Rainer Lohmann and Kevin C. Jones.

[a]Japanese data, cited from a comprehensive report by the Japanese Government on the Japanese environment downloaded from internet.

[b]ΣTEQ, the main amounts of generation source discharge of dioxin in each country downloaded from the Internet or obtained from the related reports (UNILABS, 2002; Her Majesty's, 1997; USEPA, 1995).

[c]Ratio D_C, calculated according to the method described in the former part on Theory and Methodology. D_A, D_W, and D_S represent the multimedia distributing situations of PCDD/Fs in air, water, and soil, respectively.