CORRELATION OF DIOXIN ANALOGUES CONCENTRATIONS BETWEEN AMBIENT AIR AND PINE NEEDLE IN JAPAN 3

- TREND AND ITS ESTIMATED SOURCE -

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Introduction

The purpose of this research is to identify and to clarify the relation between the dioxin (D/F) concentration in the ambient air and the needle of Japanese black pine in the West Japan area (Kyushu and Chugoku area) focusing on the trend over a period of 2 consecutive years. It is also to find out the relation between the source of D/F emission and its concentration in pine needle.

Method

The sampling period of the pine needle was October to November of 1999 and August to September of 2000. The sampling was carried out with participation and cooperation of the local citizen. The sampling areas were almost same in these 2 consecutive years. The target pine needle of about 2 years age was obtained at the height of about 1.5m from the ground. The pine needle samples from the several scattered points were blended to prepare a representative sample for the target area. The applied analytical methodology was the same to a method developed by Miyata Laboratory of Setsunan University.[2]

Results and Discussion

Figure 1 and Table 1 show the D/F TEQ concentrations of pine needle specimens in 1999 and 2000 in nine prefectures and two ordinance-designated cities with a large population over 1 million levels. Figure 2 shows a fluctuation ratio of the average D/F in pine needles in 42 municipals areas in 2000 versus 1999. Among the 42 areas, the pollution level of 10 areas in 2000 exceeded more than 25% compared to that in 1999. Especially, the number of area with more than 50% was only 5.

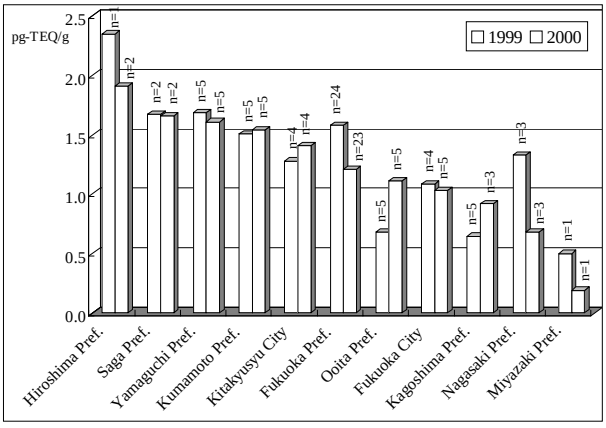

Figure 1 D/F concentration of prefectural avarage in 1999 and 2000 Unit: pg-TEQ/g

On the contrary, 13 municipals decreased to a level of less than 25%. Eighteen of the total 42 municipals were within a small fluctuation ratio of ±25%. It is possible to evaluate that these small changes with an analytical equation.

Table 1 Results of D/F TEQ conc. in pine needles in 1999 and 2000 Conc. Unit: pg-TEQ/g

Prefecture or city	Min.conc.		Max.conc.		Ave.conc.		Mean conc.		N=		Ave.
	1999	2000	1999	2000	1999	2000	1999	2000	1999	2000	+/-
Fukuoka	0.70	0.51	5.43	2.38	1.58	1.21	1.22	0.95	24	23	-23%
Kitakyusyu city	0.70	0.81	2.23	2.32	1.28	1.41	1.10	1.25	4	4	+10
Fukuoka city	0.79	0.51	1.41	2.38	1.09	1.03	1.07	0.81	4	5	-5.5
Saga	0.46	0.97	2.89	2.34	1.68	1.66	1.68	1.66	2	2	-1.2
Nagasaki	0.56	0.54	2.45	0.92	1.33	0.68	0.98	0.59	3	3	-49
Miyazaki	0.50	0.19	0.50	0.19	0.50	0.19	0.50	0.19	1	1	-62
Kumamoto	0.89	0.93	2.28	1.99	1.51	1.54	1.56	1.47	5	5	+2.0
Oita	0.45	0.28	1.14	1.57	0.68	1.11	0.63	1.36	5	5	+63
Kagoshima	0.39	0.33	1.07	1.82	0.65	0.92	0.54	0.61	5	3	+43
Yamaguchi	0.71	0.57	3.84	5.59	1.68	1.61	1.56	0.59	5	5	-4.2
Hiroshima	2.35	0.55	2.35	3.27	2.35	1.91	2.35	1.91	1	2	-19

Note:N= indicates the number of analyzed municipal samples
Kitaikyusyu City and Fukuoka City are two ordinace-designated cities in Fukuoka Pref.

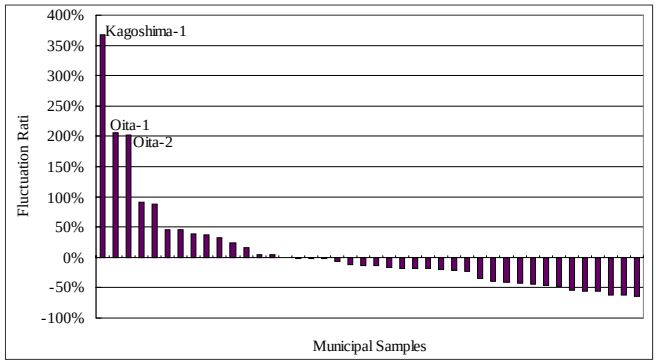

Figure.2 Fructuation ratio of the D/F Concentration in Pines in 1999 and 2000 in West Japan Area (42 municipal or area wide avearge samples)

The reason for the concentration fluctuation was analyzed from various aspects. The first aspect was in differences concerning sampling such as sampling period, sampling points, sampling areas and number of pine needle sample blended. The two months difference of the sampling period of the year 1999 and 2000 would not affect the D/F concentration in pine needle because tested needle samples were mostly 2 years old and the D/F accumulation trend in the pine needle had been confirmed to be stable after 4-6 months from the sprout.[2] Therefore it is reasonable to compare the results of these 2 years analysis for the trend evaluation of D/F concentration in those target areas. Regarding the sampling point, it had to be noted that the sampling point in each municipal and the number of individual blended needle samples were not completely equal in these 2 years surveys. However most municipal or area wide samples were blended by 10-30 samples in both year. Therefore, each analyzed sample was

considered to be as a representative for each municipal area.
The second aspect was environmental factors such as geographical and meteorological conditions of the target areas. In this project, the analyzed pine samples are not the particular single pine. Since all the pines were selected from the view points of long term survey and area wide average analysis, it was possible to say that the pine needles themselves reflected on the local environmental characteristics, especially for the ambient air pollution.

The third aspect was emission sources of dioxins under operation. It was possible to estimate that in these municipals where D/F concentration had been greatly fluctuated for 2 years, there might be their certain conditional and/or operational changes. The great elevated pollution level might be caused by additional emissions from the incineration plants in the target areas. On the contrary, the case of the big decrease might be estimated that required countermeasures had been taken at waste incineration plants after the enforcement of the national law for dioxin control. The citizen's severe and increased interest against the incineration plants in their vicinities might have influenced the operation of incinerators to decrease dioxin emission. However, it is difficult to corroborate the yearly pollution trend from the aspect of emission sources of the areas, because the flue gas dioxin concentration data are not fully publicized.

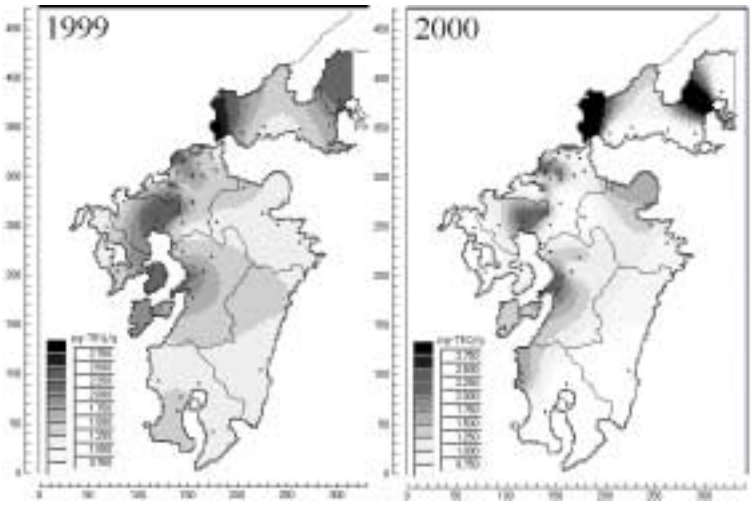

Fig.3 Pine needle D/F concentration in west Japan at municipal levels in 1999 and 2000 unit: pg-TEQ/g

On the other hand, Fig.3 shows the spatial concentration map analyzed by spline interpolation method[3,4] based on municipal level pine D/F concentration data. The dots in the map indicate a center of gravity for multiple scattered sampling points in each municipal or sampling area.

Thus, the yearly trends of the concentration in pine needle would be caused by various factors such as emission gas temperature, gas D/F concentration, gas volume, stack height of the existing incinerator in the target areas as well as regional geographic and meteorological conditions. Although it is rather difficult to find out the specific reason of the yearly fluctuation of D/F concentration in each local area, the pine needle is quite useful and effective as a bio-monitor of local ambient air pollution of dioxin.

Table 2 shows the results of the D/F concentration focused on the adjacent areas to waste incinerators in Fukuoka in west Japan and Kanagawa prefectures in central Japan. The target pine needles were sampled from more than 10 scattered points around the incinerator within radius of 500m from the stack and blended into one. These results were comparatively analyzed to reveal the area wide avarage D/F concentration of pines sampled from more than 10 scattered points within the municipal.

Table 2 Pine needle D/F concentration in adjacent area to Incinerator Conc: pg-TEQ/g

Sampling	Incinerator			Municipal			Inc./Muni.	Type of
Municipal	Ave.Conc.	N=	n=	Ave.Conc.	N=	n=	Ratio	Incinerator
Chikuho	5.4	1	4	1.7	1	20	3.2	Industrial
Tagawa	4.2	1	12	1.3	2	26	3.3	House waste
Chikushino	3.5	1	3	1.8	2	16	2.0	Industrial
Sagamihara	10.5	1	14	5.4	1	285	2.1	Industrial
Sakae	3.4	1	1	1.5	1	89	2.3	House waste
Ayase/Yamato	21.3	5	5	3.1	3	146	6.9	Industrial

Note: N: Number of analyzed samples, n: Number of sampling point
Industrial: Incinerator for industrial waste, House waste: Municipal incinerator for house waste

It became obvious that the D/F concentration in the pine needle adjacent to the incinerator was 2.1-6.9 times higher than that of area wide municipal average concentration level. Since, the dioxin level in pine needle reflects on the long term average of ambient air dioxin pollution level[2], it is reasonable that the pine needle sample from adjacent area to the incinerator will reflect 2-7 times higher D/F concentration levels than that of the areawide municipal average.

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