Citizen Participatory Dioxin Monitoring Campaign by Pine Needles As Biomonitor of Ambient Air Dioxin Pollution

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Introduction

The needle-type leaves of Japanese black pine trees (hereafter abbreviated as pine needles) have been used as an effective bio-monitor of ambient air pollution. Miyata Laboratory of Setsunan University has reported that the pine needles accumulate PCDDs and PCDFs (hereafter abbreviated as D/F) through photosynthesis and respiration during their lifetime.[1] On the basis of this study, we have revealed the correlation between ambient air and pine needle concentrations to be estimated at or near 1:10 by analyzing long term continuous ambient dioxin monitoring data and that of pine needles sampled from the same area as ambient air in the Kanagawa Prefecture in 1999.[2] Since then, the citizen groups of each local area all over Japan have started monitoring the ambient air dioxin concentration levels by using pine needles. Samples analyzed during these 5 years totaled more than 650 throughout Japan. The results of these citizen participatory environmental monitoring activities are the tremendous effects achieved in reducing the dioxin levels. This occurs through observation of the dioxin emission sources such as Municipal Solid Waste Incineration Plants as well as the Industrial Waste Incineration plants, which exist in numbers exceeding several thousands in Japan. This short paper will present the results of 56 municipalities of western Japan where ambient air dioxin levels have improved steadily against local averages during these 5 years.

Method

Sampling and Blending: Prior to the initiation of sampling activities, each citizen group of the target areas was educated in pine needle collection techniques appropriate for the study. The sampling period of pine needles was set from August to September of each year from 1999 to 2003 in 56 areas of western Japan

(Chugoku and Kyusyu Area). The target pine needles, primarily 2 years old, were sampled approximately 1.5 m above the ground level. The sampling points of each target area differed depending upon the scope. From 10 to 30 scattered points in each municipality were utilized to determine the average of the target area. Local citizens, including school children and housewives, who also have been checking the condition of the emission sources of their communities conducted the sampling activities. After completion of sampling, each small portion of the pine needles were packed separately in plastic bags and sent to Environmental Research Institute (ERI, Tokyo). Then, an equally weighted portion of each sample was blended into one sample per target area for a total of approximately 100 g. The procedure is shown in Photo 1.

Photo 1: Sampling Activity, Received samples and Weighting and blending procedure

Analytical method: Miyata Laboratory in Setsunan University originally developed the analytical method of the pine needles[1]. Maxxam Analytics Inc. (Waterloo Lab., Ontario, Canada) has been responsible for all analyses of the pine needles for this specific project, applying the same methodology as developed by Miyata Laboratory. A brief outline of the D/F analytical methodology is shown in Figure 1.[1] The target parameters for analysis were D/F only; the PCB WHO List was not analyzed.

1. Sample (50 g)
   Cut into a size of 3 cm
   Dried by lyophilization
2. Lyophilized sample
   Macerated with an Ultra-Turrax using toluene (500ml)
   Extraction with toluene under reflux (4hr)
   Addition of silica gel (50g) and leave it a whole day and night
   Evaporated to dryness and dissolved with n-hexane (10ml)
3. 30% aliquot of extract

Addition of $^{13}C_{12}$-PCDDs and PCDFs internal standards
Multi layer silica gel column chromatography
Eluted with n-hexane (210 ml)
Concentrated to 10 ml in n-hexane
Alumina column chromatography (neutral, activity I)
Separated into 1st and 2nd fractions
Concentrated to 20 µl in n-decane
4. 2nd fraction
HRGC-HRMS-SIM analysis (R=7000 – 10000)

Figure 1. Outline of analytical method for PCDDs and PCDFs in black pine needle[1]

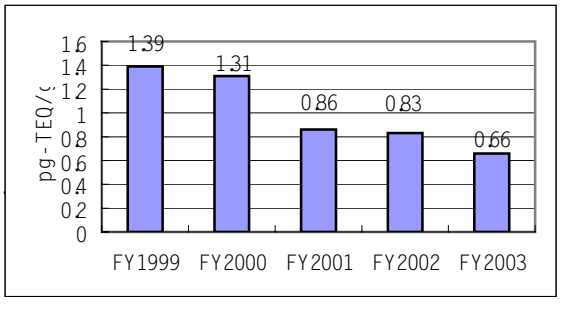
Analysis of the results: After receiving all of the analytical data for the pine needles, ERI illustrated the spatial concentration on a map of Western Japan (Kyusyu and Chugoku Area). Results were based upon the municipal level dioxin concentration data, which were determined by Spline interpolation method.[3,4] The Dioxin concentration map of each year has been printed and distributed to all the participants of the monitoring activities with appropriate comments and implications concerning the dioxin pollution levels and condition of emission sources operated within their neighborhood. This information has been effectively used for citizens' independent activities for reviewing the municipal waste management policies as well as in strengthening the control against the incineration plants and open burning in their communities.

Results and Discussion

Table 1 and Figure 2 show the trend of average annual dioxin concentration levels of the target area based on the results of continual investigation for the past five years. The dioxin concentration levels have gradually improved during 5 years. Japanese citizens' concern about dioxins had reached its peak in 1999, the same year, relatively high dioxins were found in the vegetables (0.64-0.75pg-TEQ/g.wet in spinach) near waste incineration plants. This was also the case for dried tea leaves grown in the areas where incineration plants were densely populated in the suburbs of Metropolitan Tokyo.[5]

Table 1 Average Dioxin Level And Target Areas

	Average Conc.	Target Areas



FY 1999	1.39	51
FY 2000	1.31	56
FY 2001	0.86	55
FY 2002	0.83	55
FY 2003	0.66	56

Unit of Conc.: pg-TEQ/g (dwb)

Figure 2 Trend of Dioxin concentrations in Pine needles

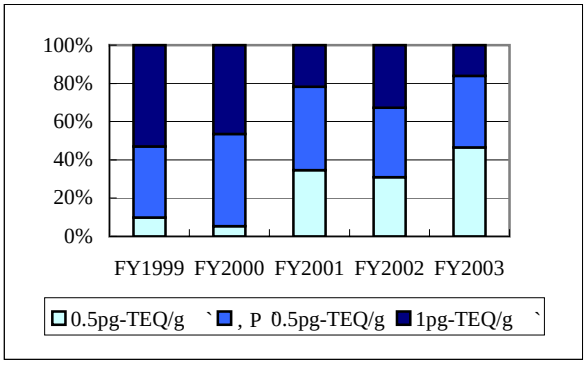
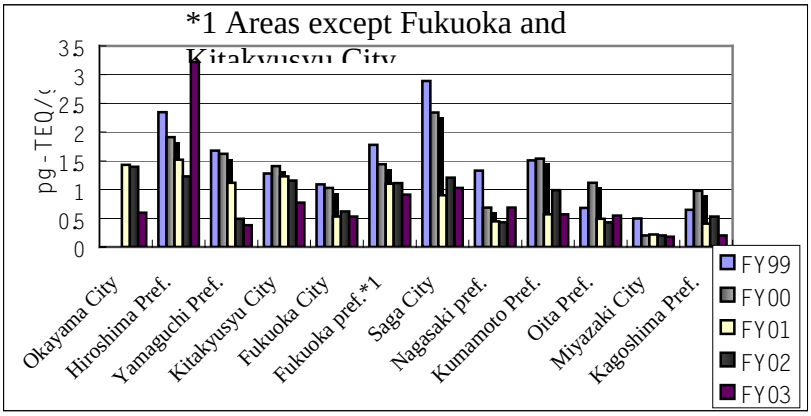


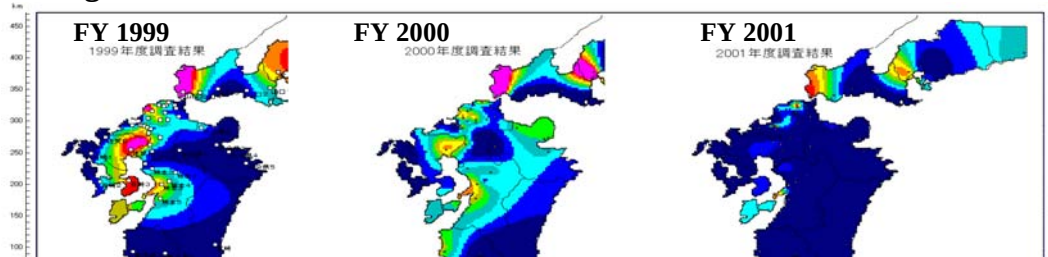
Figure 3 shows the clear shift of concentration ranges during the 5 year monitoring campaign. In 1999 and 2000, the concentration levels over 1pg-TEQ/g were about 50%, but reduced to 16% by 2003. This improvement had been achieved partly because the enforcement of the new emission standards for the facilities that came into effect as of 1 December 2002.

Figure 3 Shift of Concentration range during 5 consecutive years



Not only the reduction of the limit values by the government, but the increase of the citizen's awareness and consciousness to reducing the incineration of waste played a big role in this steady improvement.

Figure 4 Results of Dioxin Monitoring Campaign; Prefectural and City Average



FY 2002 **FY 2003**

Kyusyu & Chugoku Area

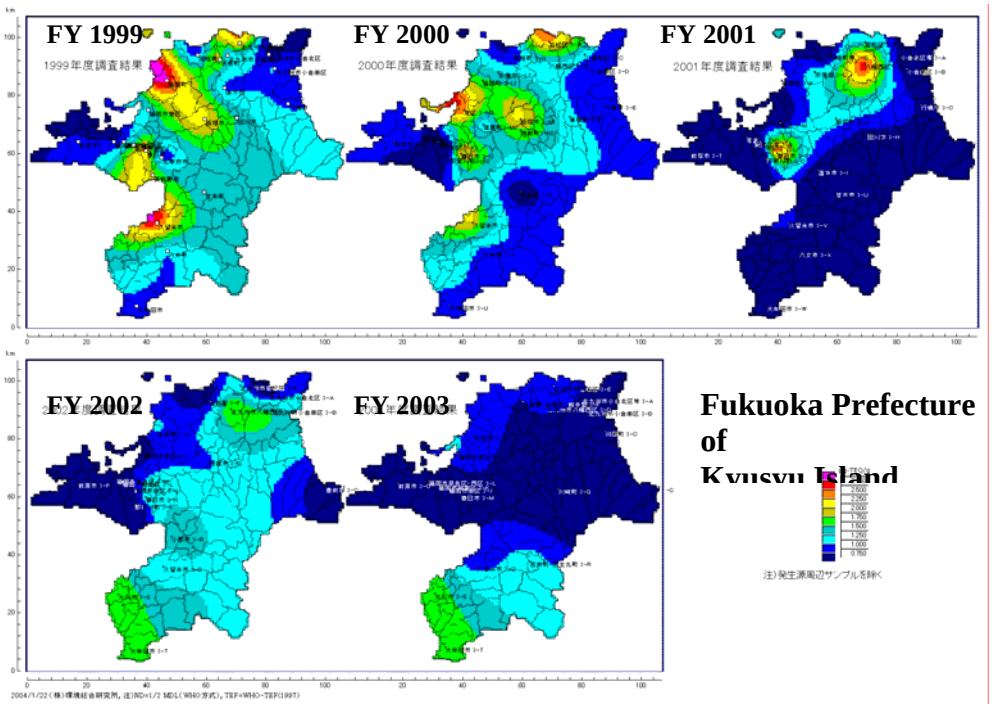


Figure 5 Dioxin Concentration Map of Kyusyu & Chugoku Area, Fukuoka Prefecture

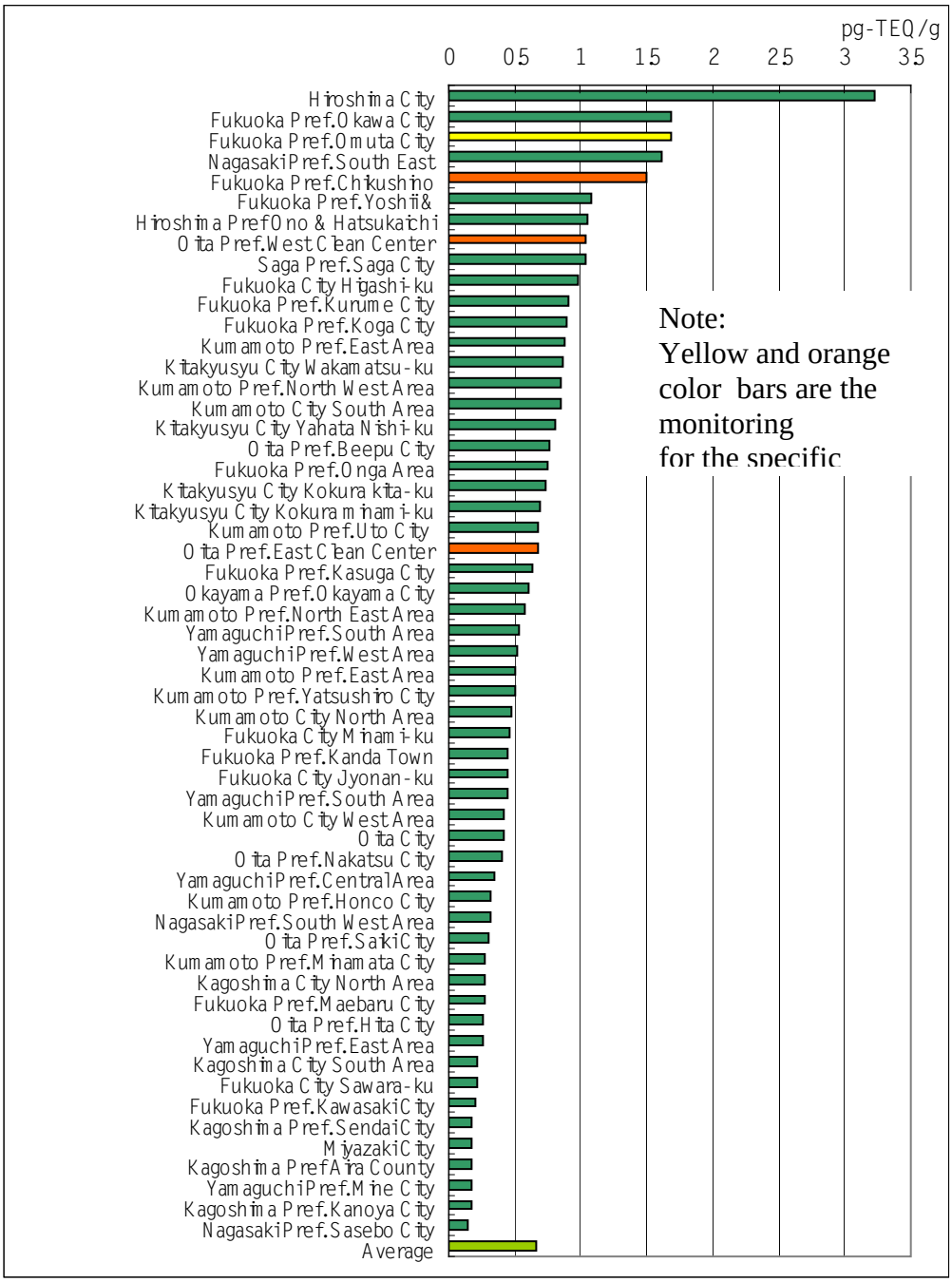


Figure 6 Pine Needle Dioxin Ranking of 56 monitored areas in Campaign 2003

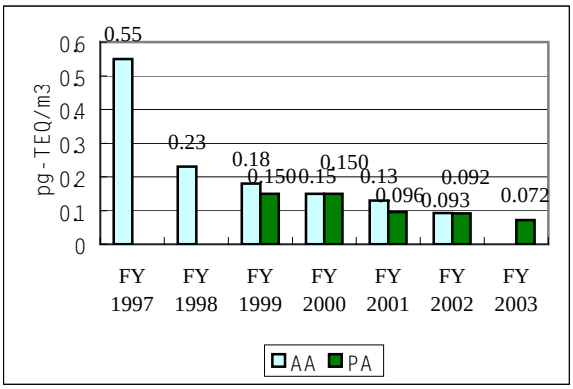


Figure 7 Comparison of Government Measurement of National Average Ambient Air[6] and the Estimated Ambient Air Concentration from Pine Needle. Legend: AA is Ambient Air Measurement Data, PA is Estimated Ambient Air Values from Pine Needles.

Figure 5 details the map of dioxin concentrations in pine needles of Western Japan and Fukuoka Prefecture located in the northern part of Kyusyu Island (most populated Prefecture among the target areas). Figure 6 is the latest results of the 56 areas monitored in 2003. After receipt of the D/F results of pine needles, we estimated the total dioxins (D/F and Co-PCB) of each sample assuming that 10% of whole dioxins could be attributed to Co-PCBs on average. Finally, the total dioxin values were used to estimate the ambient air dioxin concentrations in correlation of 1:10. Figure 7 shows the strong correlation between the measured data of ambient air and estimated values from the pine needles. This indicates that the pine needle monitoring campaigns for ambient air are scientifically meaningful and an effective tool in environmental education and policy making for waste management in local municipalities.

Acknowledgment

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