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Concentration of Dioxins in Ambient Air Over the Past 20 Years (II)

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Introduction

In order to understand chemical exposure trends to biological organisms and effectively regulate their effects, it is important to look at the annual trends regarding the concentration of these compounds in the environment. Methods to understand the past include using objects that contain natural records regarding historical trends (e.g. sediment core, ice core, amber) and using samples actually collected and stored over time (specimen banking). While the previous method allows trend comparison over a longer period of time, the later method is superior in terms of accuracy and resolution. On the National Institute of Public Health's building in Tokyo Japan, a High Volume Air Sampler has collected dust samples from ambient air in a quartz paper filter over the past 20 years. Theses samples were then stored in a deep freezer. In the present study, portions of these samples were analyzed in order to investigate the trends regarding the concentration of dioxins (PCDDs/PCDFs and coplanar PCBs) in ambient air over the sampling period. The high volume air sampler was periodically calibrated to assure accurate and reliable sampling. Authors have reported dioxins concentrations in ambient air from 1980 - 1999. This time, analyses of the year samples from 2000 to 2001 have been completed.

Method

The filter samples stored in a deep-freezer (-80 degree C) were taken, and divided according to the season, Spring (March - May), Summer (June - August), Autumn (September - November) and Winter (December - February). Samples were collected and stored every six days so each group included approximately 15 samples. In order to accurately quantitate the concentration of dioxins, after combining quartz filter papers that had absorbed a sample size of approximately 1200 m^3 of air, a portion (2 pieces of filter paper x 47mm diameter) of filters were punched and analyzed. After punching to appropriate size, filters were dried, internal standards of all dioxins with an assigned WHO-1998 TEF were added and followed by soxhlet extraction with toluene. After solvent exchange to hexane, multi-layer silica gel and activated carbon column chromatography was employed for sample cleanup. Samples were then assayed by HRGC/HRMS. Polychlorinated-*p*-dixions, polychlorinated furans, and the 12 coplanar PCB's with assigned TEF's were assayed. All samples preparation, extraction and cleanup were performed in a clean room, and all solvents used were distilled by sub-boiling method in a clean room prior to use. It should be noted that no PUFPs were sampled in this study. Analyses were performed in accordance with ISO/IEC 17025(JCLA4).

Results and Discussion

The assayed concentration and TEQ of PCDDs/PCDFs and co-PCBs (#77, #81, #126, #169, #105, #114, #118, #123, #156, #157, #167, #189) from 1980 to 2001 for each season is shown in *Figure-1* and *2* respectively. The average TEQ (PCDDs/PCDFs) of all years was 0.68 pg-TEQ/m^3, with observed concentrations varying between 0.13 pg-TEQ/m^3 (2001) and 1.1 pg-TEQ/m^3 (1994). During recent years (1996 - 2001), a decreasing trend in concentration is seen. In Japan, regulation has started from 1997 regarding stack gas of incinerator. Obtained results show effect of regulations. Co-PCB's concentrations have clearly been decreasing every year from 0.063 pg-TEQ/m^3 in 1980 to 0.029pg-TEQ/m^3 in 2001. Difference of secular changes for PCDDs/PCDFs and co-PCBs between measured concentration and TEQ descriptions shows there were differences of isomer/homologue composition regarding each year.

Also, the average concentrations of PCDDs/DFs were high in the winter and low in the summer (*Figure-3*). It is thought that this could be a reflection of the difference in the quantity of airborne dust. There was no significant difference between the seasonal concentrations of co-PCB's. It is thought that this could be due to the fact that a great deal of PCB's exist in the gas fraction.

Currently, samples after 2001 are being analyzed, so we are waiting for these results.

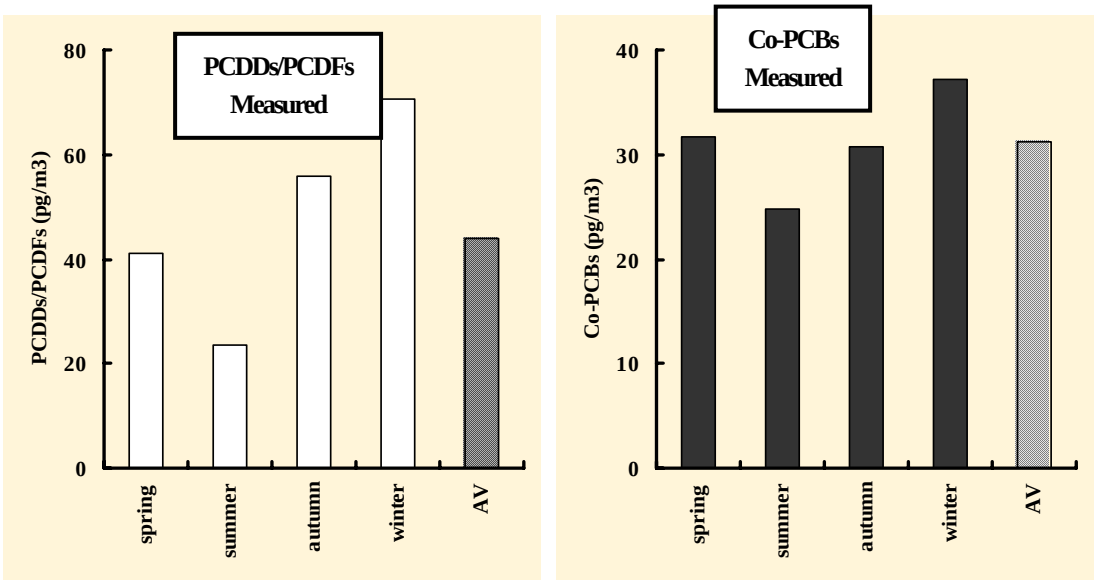

Figure-3. Seasonal Variations of PCDDs/PCDFs and Co-PCBs in Ambient Air for Past 20 Years.

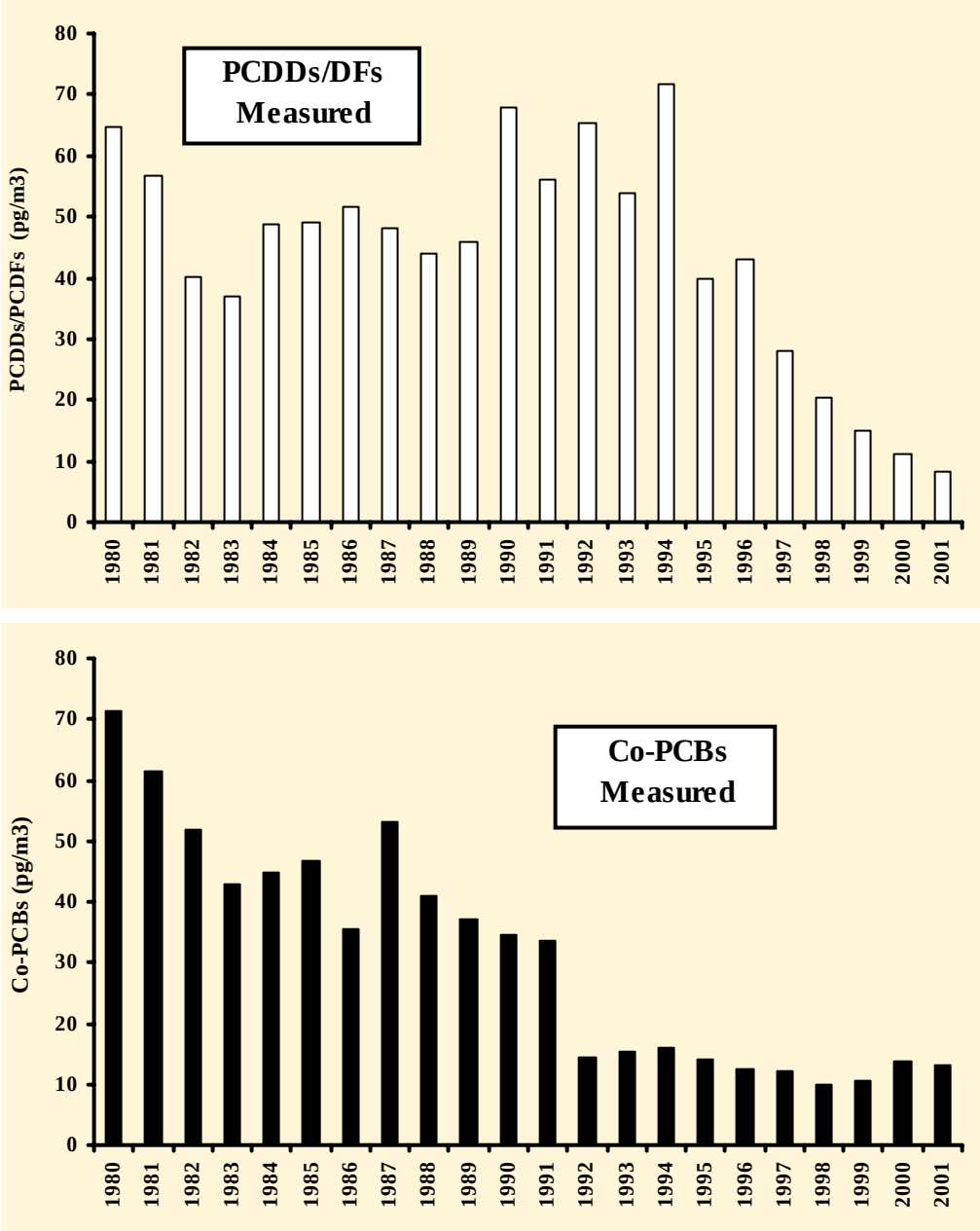

Figure-1. Secular Changes of Measured Concentrations for PCDDs/PCDFs and Co-PCBs.

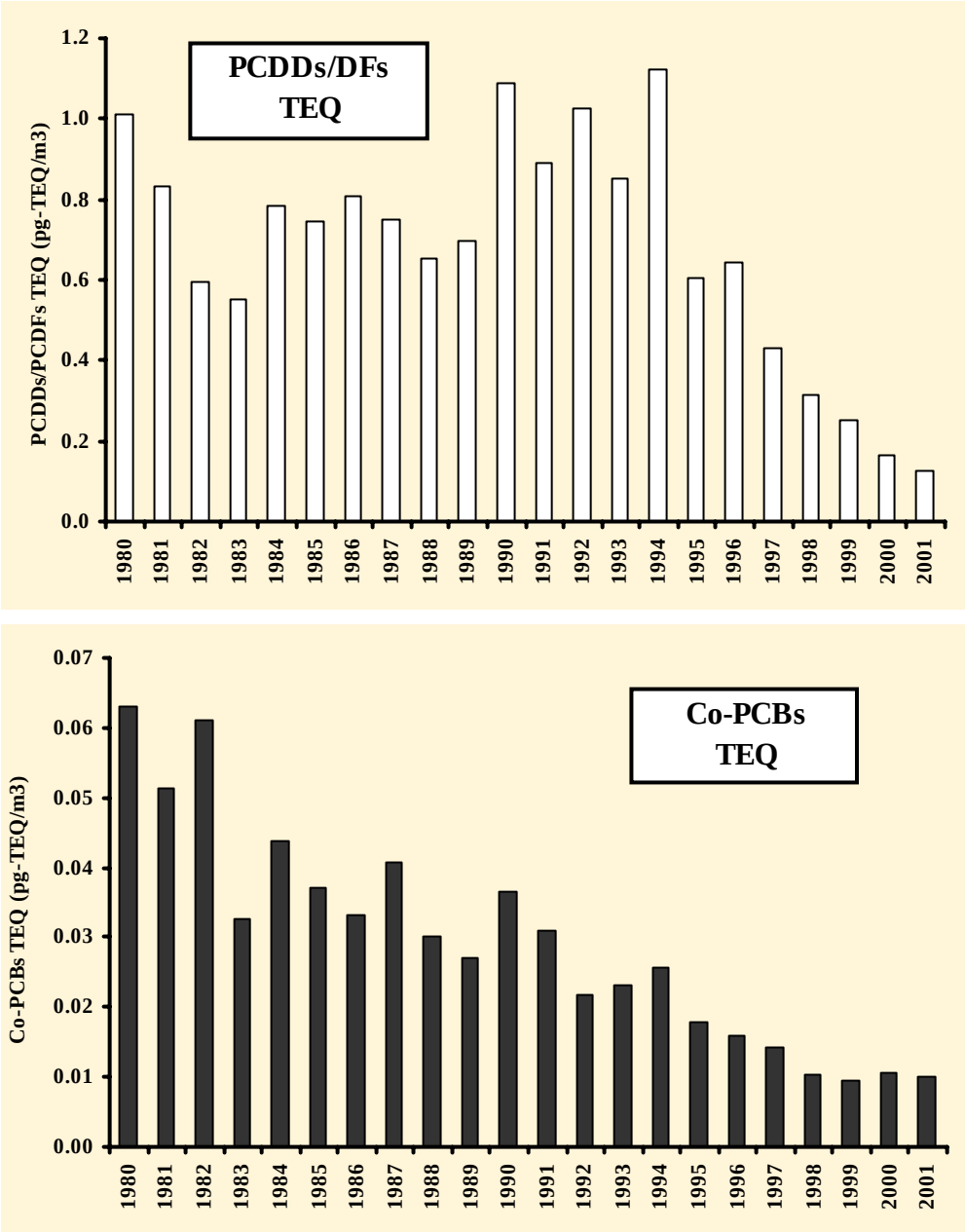

Figure-2. Secular Changes of TEQ for PCDDs/PCDFs and Co-PCBs.