

## CONCENTRATIONS OF ATMOSPHERIC PCDDs/PCDFs AND PCBs IN GYEONGGI AND NEARBY AREA

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### Introduction:

It is important to characterize atmospheric behavior because most dioxins are released into the atmosphere and travel to other environmental media and biosphere through the atmosphere. The dioxins released to the atmosphere are present in the form of gas phase and particulate, and the gas phase moves in the form of dispersion equilibrium/ wet deposition into soil or water. The role of atmospheric monitoring is crucial in identifying local sources and movements. Since the Passive air sampler(PAS) is influenced by weather conditions (wind direction, wind speed, temperature) during the sampling period, the sampling rate may be different depending on the sampling point. In the case of PAS monitoring, it is aimed to identify the long-term average pollution situation in various regions rather than accurate concentration calculation. It aims to provide basic data for environmental management such as local emission source management by accumulating and sharing data continuously.

The objective of this study is to evaluate the effects of POPs in Gyeonggi province and nearby area and to evaluate the pollutant transport route of the coastal industrial complex by the westerlies blowing from the west sea between Ganghwa Island and Yeongjong Island. In order to examine the effects of persistent organic pollutants in the area adjacent to Gyeonggi province, it is firstly evaluated as the pollutant movement route of the coastal industrial complex by the westerlies blowing from the west sea between Ganghwa Island and Yeongjong Island in Bucheon and Incheon area (A) (A2), which is adjacent to Bucheon Metrology Center (A1) and whose straight line distance is within 4.8km, was selected. In Chungnam area (B), four (B2 ~ B5) sites were selected, where coal-fired power plant and steelworks are located. The distance from the coal-fired power plant located in Dangjin to B1 (Pyeongtaek port measuring station) is about 31 km away from the straight line. Fifteen samples of atmospheric sampling PCDD/PCDFs and 12 species of dl-PCBs (dioxin-like PCBs) were investigated using a high volume air sampler (Active Air Sampler: AAS) and a passive air sampler (PAS) from February 2017 to November 2017

### Materials and Methods:

#### *Sampling collection*

Each sample was sampled continuously for 48 hours at a flow rate of 400 ~ 500 L / min by Active air sampler. The total collected flow rate was 1,440 m<sup>3</sup>. Atmospheric samples were taken at different seasons of the year from spring to autumn 2017. PUF disk based air samplers were deployed at six sites. Details for sampling sites were given in Figure 1. The sampling sites were divided into two groups: (a) Bucheon and Incheon, (b) Pyeongtak and CheungNam . All PUF disks (TE-200 PAS, Tisch Environment) were housed in stainless steel chambers to protect the disks from sunlight and precipitation. The samples were collected over 90 days per each season from March 2017 to September 2017. When the sampling was finished, the PUF disks were transferred to laboratory and stored in refrigerator before extraction.

#### *Sample extraction and analysis*

All samples were extracted in large volume Soxhlet extracted with 800mL toluene over 20 hours. These extracts were concentrated to 1mL by a rotary evaporator. The analytical procedure was followed by EPA Method 1613. Briefly, all samples were analyzed on a HRGC/HRMS (DFS, Thermofisher scientific) using isotope dilution method. For quantitation,  $^{13}\text{C}$ -labelled surrogate and internal standards (EPA-1613LCS, EPA-1613ISS, Wellington for PCDDs/PCDFs, 68B-LCS, 68B-CSS, Wellington for dl-PCBs) were used. The recoveries for the  $^{13}\text{C}_{12}$  labeled compound standards were within 50 ~ 120 %.

## Results and discussion

### *PCDD/PCDFs and dl-PCBs concentrations using Active Air Sampler*

The regional characteristics of Bucheon and Incheon area indicate that the A1 area of Bucheon has a lot of small scale factories surrounding it, and the A2 point of Incheon is a residential area with almost no fixed pollution source near the city atmosphere measuring station and it is close to Geyang Mountain. PCDD/PCDFs and dl-PCBs in the Bucheon area were higher than those in Bucheon and Incheon in summer except rainfall at sampling time. The mean concentrations of the three measurements in spring, summer and autumn were 0.042 pg-TEQ/S  $\text{m}^3$  in Bucheon area (A1) and 0.022 pg-TEQ/S  $\text{m}^3$  in Incheon area (A2). The mean concentrations of dl-PCBs were 0.0004 pg-TEQ /S  $\text{m}^3$  for A1 and A2. The concentration of particulate and gaseous PCDD/PCDFs is higher than that of A2 at the A1 point, which is considered to be due to the particulate matter in the exhaust gas emitted from factories in the industrial area. The ratio of particulate dioxin in the case of actual concentration was about 56.7 ~ 93.5%, the ratio of particulate dioxin was about 50.0 ~ 94.7% in case of toxic equivalent conversion concentration (except during summer) %, which is generally higher than that of gaseous dioxins. In the case of toxic equivalents, the proportion of particulate dioxins is reduced compared to the actual concentration, which is believed to be due to the presence of gaseous dioxins in the gas phase and high levels of dioxins in the particulate form. In the case of B region, the proportion of particulate dioxin was higher than that of A region. This, as in A1 and A2, is believed to be due to particulate matter in the effluent from the plant in the industrial area. Concentrations of PCDD/PCDFs and dl-PCBs at B1 site (PCDD/PCDFs averaged 0.038 pg-TEQ/S  $\text{m}^3$  and dl-PCBs concentration 0.0005pg-TEQ/S  $\text{m}^3$ ). The results were higher than B2 ~ B5 region. PCDD/PCDFs concentration ranges from B2 to B5 were ranged from 0.007 to 0.035 pg-TEQ/S  $\text{m}^3$  and dl-PCBs concentrations ranged from 0.0001 to 0.0004pg-TEQ/S  $\text{m}^3$ . The PCDD/PCDFs distribution ratio in B region is about 76.6 ~ 99.1% and the ratio of particulate dioxin is about 95.3 ~ 100 %, the proportion of particulate dioxins was higher than that of gaseous dioxins. In the case of toxic equivalent conversion concentration, the proportion of particulate dioxin slightly increased compared with the measured concentration. Seasonal particulate matter and gaseous dioxin distribution showed that the ratio of particulate dioxin was the lowest in summer and the highest in autumn. According to the results of previous research in Gyeonggi Province, the majority of dioxin concentrations in the particle-gas distribution characteristics are distributed in the form of particles, and the particle-gas distribution varies according to the seasonal temperature. Seasonally, the winter period is the highest, but the winter period is not included in this survey.

### *PCDD/PCDFs and dl-PCBs concentrations using Passive Air sampler*

Mean Concentrations of PCDD/DFs, A1 was 0.072 pg-TEQ/day and A2 was 0.063 pg-TEQ/day. And B1 was 0.339 pg-TEQ/day, 0.164 for B2, 0.137 for B3 and 0.195 pg-TEQ/day for B5 respectively. dl-PCBs were measured as 0.0023, 0.0024, 0.0037, 0.0019, 0.0032, and 0.0021 pg-TEQ/day, respectively. According to the survey results of PCDD/PCDFs and dl-PCBs, the concentrations of B areas were higher than those of A areas.

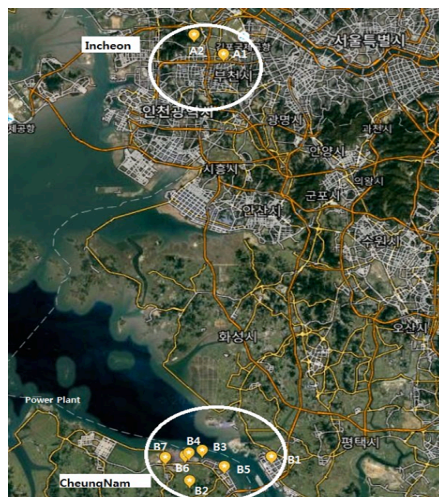
In the case of AAS, PCDD/PCDFs and dl-PCBs concentrations in A areas were higher than those in B, where coal-fired power plants and steel mills and factories are adjacent to each other. Comparing these results of HAV and PAS, the concentration of PCDD/PCDFs and dl-PCBs may be different according to the weather conditions

at the time of sampling and sampling method. It is also believed that it is necessary to accumulate data continuously rather than single-shot PCDD/PCDFs and dl-PCBs measurements.

#### *Dioxin homolog and isomer distribution*

The ratio of the isotopic enantiomeric concentration and the measured concentration of toxic equivalents of A1 site and A2 site are similar to each other in Spring, but in summer and autumn, the homologues and isomers distribution tendencies of A1 and A2 points are different. Based on the isotopic distributions of measured concentrations and toxic equivalents, A1 and A2 areas seem to be affected by the same pollutants in the spring due to wind. In summer and autumn, there are totally different homologs and isomers, which are considered to be influenced by other pollutants. In the B areas, the measurement points B2 to B4 in Chungcheong province have similar isomer distribution characteristics in spring and are affected by the same pollutants. However, there is a slight difference in the distribution of isomers between B2 and B4 in Chungnam and Pyeongtaek (B1), and it is considered to be affected by other pollutants besides the same pollutants. In the case of summer and autumn, the isotopic distribution is also observed at the measurement points in Chungnam area. The wind direction of Chungnam during the sampling time of spring was northwest and north, and the wind direction of Pyeongtaek was west and southwest. In the case of Pyeongtaek(B1), it was southeast, south-south, and southwest. Pyeongtaek in the autumn (B1) was a westerly wind in the northwest and northwest directions.

The results of AAS showed that the concentrations of PCDD/PCDFs and dl-PCBs in A area were higher than B area, where thermal power plants and steel mills were adjacent. However, PAS results were higher in B areas. The dioxin concentration in AAS may vary greatly depending on the weather conditions at the time of sampling and the operation status of the discharge facilities. Therefore, it is necessary to continue to monitor the dioxins using both AAS and PAS.



**Fig. 1. Sampling sites**

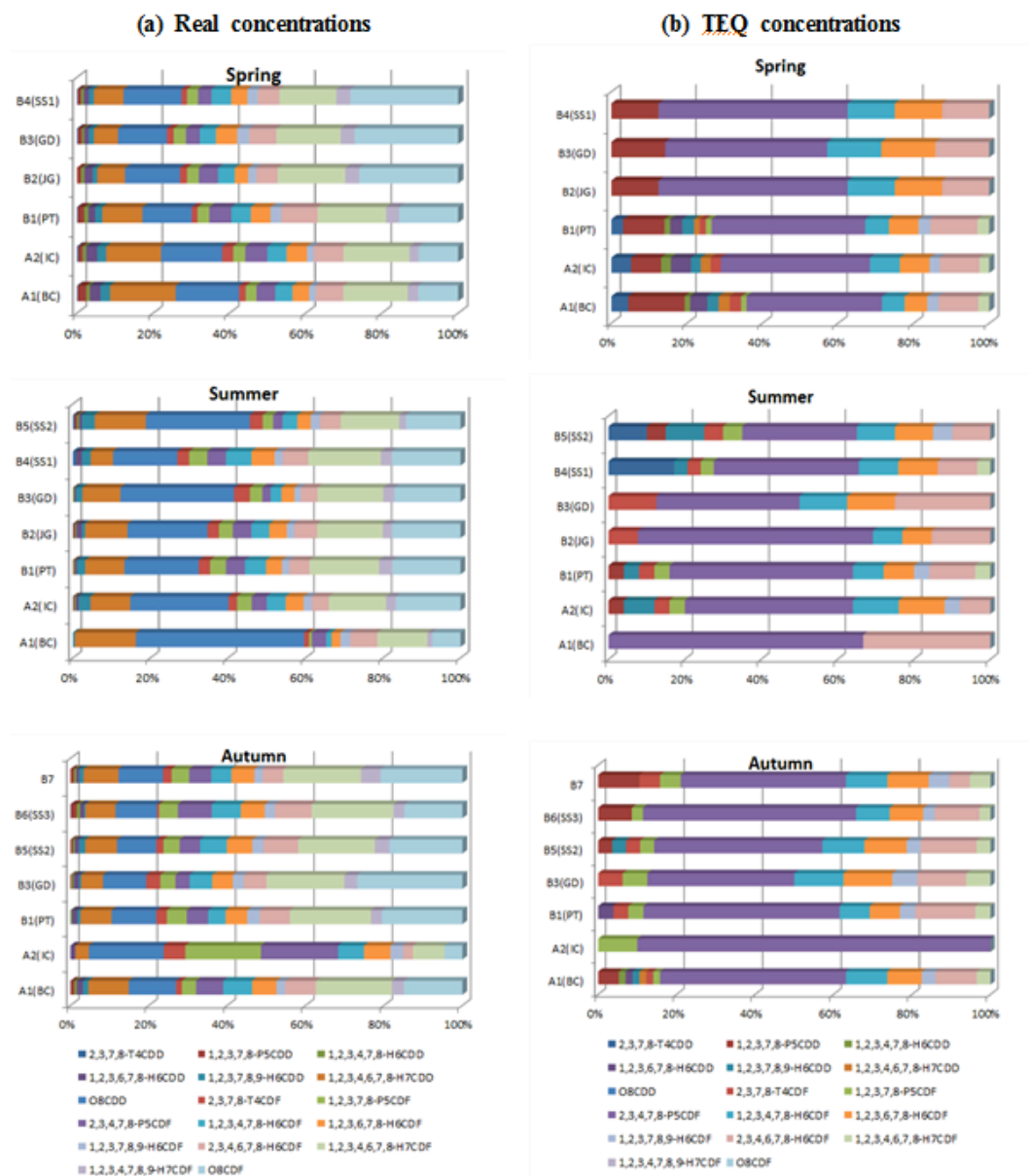


Fig. 2. The distribution of PCDD/PCDFs congener profiles by Active Air Sampler

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