Assessment of Atmospheric PM$_{2.5}$ and PCDD/Fs Collected by Different High-volume Ambient Air Sampling Systems

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ABSTRACT

To evaluate the difference in hazardous air pollutants in PM$_{2.5}$ between reference method (National Institute of Environmental Analysis; NIEA A205) and high-volume air sampler (European standard:EN14907 and Japan method), we set up a sampling station on the campus of National Yang-Ming Chiao Tung University, northern Taiwan, during 2014-2015. Both vapor and solid phases of dioxins were collected.
using high-volume samplers, according to EN14907 and Japan method. The flow rate was set at 500 L min$^{-1}$ and 1000 L min$^{-1}$, respectively. To compare the difference with the high-volume air sampler, we simultaneously used the reference air sampler based on Taiwan NIEA A205.11C, at the flow rate of 16.7 L min$^{-1}$ (BGI PQ200-FRM). The mass concentrations of PM$_{2.5}$ measured with NIEA A205, EN14907, and Japan method were 20.2 ± 8.79, 25.4 ± 10.5 and 28.6 ± 13.9 μg m$^{-3}$, respectively. The difference of the mass concentration of PM$_{2.5}$ obtained from two different methods was lower than 3.9%. Moreover, the concentrations of PCDD/F between solid and vapor phases were 56.9 - 1,090 and 38.6 - 67.1 fg m$^{-3}$ via EN14907 and 51.1 - 1,150 and 18.4 - 81.8 fg m$^{-3}$ via Japan method, respectively. Obviously, there is no significant difference between these two samplers. Compared to the method of NIEA, high volume air sampling method not only provided equivalently good quality data but offer a higher sample quantity for analyzing the trace level chemical component of hazardous air pollutants and the toxicity in different areas.

**Keywords:** PM$_{2.5}$, High-volume air sampler, Impactor, Dioxin
1. INTRODUCTION

Atmospheric fine particulate matter (PM$_{2.5}$) are particles with a size of less than or equal to 2.5 μm (DeCarlo et al., 2004), and they can enter to lung from the upper respiratory tract (Dockery et al., 1993). The sources of PM$_{2.5}$ were divided into natural and anthropogenic sources. As we know, natural sources include the airborne particles, ocean droplets, vegetation dust, forest fires, viruses, etc (Li et al., 2021). Anthropogenic sources could be more complicated, including the vehicle's emissions or industrial emissions such as power plants, incinerators, and industrial processes (smelting, heating, welding) (Querol et al., 2001; Amesho et al., 2021; Mutuku et al., 2021; Wu et al., 2021). Moreover, a previous study also mentioned that PM$_{2.5}$ was influenced by synoptic meteorology (Park et al., 2021).

Many studies also mentioned the health effect of environmental pollution from atmospheric PM$_{2.5}$ (Pope III and Dockery, 2006; Corbett et al., 2007; Perez et al., 2009; Samoli et al., 2014). Besides, PM$_{2.5}$ was already listed as the primary carcinogen by WHO and IARC (International Agency for Research on Cancer, 2011). The earliest industrialized countries like the United Kingdom and the United States also issue the clean air regulation in 1960. Taiwan also ruled the law of air pollution prevention in 1975. Furthermore, the Air Quality Index (AQI) was also used, and it included the monitoring 8 hr value of ozone by the United States in 1999.
other hand, China and Hong Kong used the Air Pollutant Index (API). In 2012, the Taiwan government also ruled the standard of air pollution about emission and atmospheric PM$_{2.5}$.

Hazardous air pollutants are the trace level in the atmosphere and secondary particles from the secondary pollutants. It was the primary chemical reaction from the atmospheric pollutants in the vapor phase and attached to the fine particle matters. As mentioned last part, these pollutants would enter into the human body from the respiratory system and impact health (Tian et al., 2017; Yin et al., 2017; Bowe et al., 2019; Chen and Hoek, 2020; Hystad et al., 2020). That was the reason why particle matter had been listed as the control item for air pollution. In the wintertime, the air quality of Taiwan was affected by long-range transport events like the China haze (Gao et al., 2021). The concentrations of PM$_{2.5}$ were over to 57.1 ± 13.6 μg m$^{-3}$ during the haze period in northern Taiwan (Wang et al., 2016). Moreover, the concentration of PM$_{2.5}$ was 28.4 ± 17.5 and 35.0 ± 22.3 μg m$^{-3}$ for Dacheng and Fangyuan in central Taiwan, respectively (Chen et al., 2016). Yuan et al. (2020) mentioned that there was 28–68% of marine PM$_{5}$ transported to the Matsu Islands by long-range transport. Those studies also indicated that there was a significantly higher PM$_{2.5}$ concentration measured during special event days (including long-range transport events and local pollution events)(Lai and Brimblecombe, 2021). Because of the complicated contents
in PM$_{2.5}$ were in the trace amounts and generally toxic to human and environment, hence, the better time-resolution of chemical composition in PM$_{2.5}$ measurement via high-volume should be further investigated.

Previous studies collected the PM samples at the flow rate of 16.7 - 32 L min$^{-1}$. Lee et al. (2011) collected the PM samples for 24 hr and set the flow rate to 16.7 L min$^{-1}$ in Mt. Lulin, Taiwan’s background station. The air sampling volume was over 23 m$^3$. Moreover, the concentration of PM$_{2.5}$ ranged from 10 - 25 μg m$^{-3}$ during 2003 - 2009. Tsai et al. (2015) also collected the PM$_{2.5}$ samples and used 3 filter package at 32 L min$^{-1}$ in southern Taiwan. In their research, the VAPS (Versatile Air Pollutant Sampler; URG-3000K, URG, USA) was used to collect the fine and coarse particles. Hence, the concentration of PM$_{2.5}$ at the pre, during, and post-Mid-Autumn Festival was 25.4 ± 5.8 μg m$^{-3}$, 25.6 ± 5.2 μg m$^{-3}$, and 15.9 ± 2.9 μg m$^{-3}$, respectively. Furthermore, Hsu et al. (2016) used the BGI PQ200 ambient air particulate sampler to collect PM$_{2.5}$ and PM$_{10}$ in central Taiwan. Each sample was equipped with PTFE filters and set the flow rate to 16.7 L min$^{-1}$ for 24 hr. The annual mean concentration of PM$_{10}$ and PM$_{2.5}$ were 45.0 ± 32.6 μg m$^{-3}$ and 26.6 ± 20.6 μg m$^{-3}$, respectively. In 2012-2013, a previous study (Sugita et al., 2019) also collected the PM$_{2.5}$ samples between the low and high-volume air sampler to compare the R$^2$, and it was also higher than 0.67 in the water-soluble ions. Patel et al. (2021) was also used PM$_{2.5}$
WINS impactor and Tisch PM$_{2.5}$ high-volume air sampler to evaluate the difference between the two methods. The $R^2$ result of PM$_{2.5}$ between different particle sampling methods was higher than 0.95. But this research was not focused on the cyclone separator with PM$_{2.5}$. Even though over the past ten years, many research collected the PM sample by the reference air sampler and analyzed the chemical composition in the atmosphere. We still need to improve the air sampling method with high-volume for the hazardous air pollutants with very low concentration in the ambient environment (e.g. Dioxin, PAHs). Summary for the previous researches that using a low flow rate of the air sampling trains, it was easy to collect the PM$_{2.5}$ sample on filters from the atmosphere. That we can know the basic data from the filters. But there was difficult to analyze the trace level chemical compounds from the filters with low-volume air sampler.

As far, the sampling method of atmospheric fine particle matters by the Environmental Protection Administration of Executive Yuan was constant flow rate (16.67 L min$^{-1}$) to pump the air with specific shape in the inlet of the sampler and used the Very Sharp Cut Cyclone (VSCCTM) separator to collect particle matters to filters. This study chose the PQ200 FRM Ambient Air Particulate Sampler (BGI) from NIEA A205.11C (Taiwan EPA) as the reference method sampler and used the high-volume sampling system for comparison. The first high-volume sampling system
was from Europe, including DIGITEL Aerosol Sampler DHA-80, flow rate: 500 L min\(^{-1}\) and Analitica Air Flow PM2.5-HVS, flow rate: 500 L min\(^{-1}\). The second system came from Japan (Sugita et al., 2019), namely Sibata High-volume Air Sampler HV-1000R, flow rate: 1,000 L min\(^{-1}\). All three sampling systems operated simultaneously to verify the difference in sampling method between NIEA (Taiwan EPA) and high-volume sampler.

2. MATERIALS AND METHODS

2.1 Sampling sites and samples collection

Taiwan is located at the receptor region from the long-range transport event, like the Northeast monsoon and Southeast Asia biomass burning. This study used the high-volume air sampler from Taiwan EPA, Europe, and Japan to collect the air samples for PM\(_{2.5}\) on a school located in Taipei city during the winter of 2014 and summer of 2015. The sampling site was a hillside terrain, and Taipei Veterans General Hospital was 1 km away, the MRT station was 1.5 km away. Also, there were Anbu Weather Station (Yang-Ming Mountain) and Beitou Thermal Valley, which were 3 km away. Moreover, we also chose eight sampling sites far away from the residential area and close to the riverside, two sampling sites close to the roadside, and one sampling site at the expressway to collect the PM\(_{2.5}\) in central Taiwan. Every sample was
collected for 24 hr. The sampling method of Taiwan EPA collected a sampling volume of 23 m³. On the other hand, the total sampling volume of Europe and Japan was more than 900 m³, even 1400 m³.

2.2 Difference in Three Sampling methods

2.2.1 PM$_{2.5}$ sampling method in Taiwan (NIEA A205)

The sampling method of the Environmental Protection Administration, Executive Yuan, is pumping air through Very Sharp Cut Cyclone (VSCC$^{TM}$) separator to the filter with a constant flow rate of 16.67 L min$^{-1}$. By the inertial motion and cyclone separator, aerosol was collected of which aerodynamic diameter less than or equal to 2.5 on the sampling media.

2.2.2 PM$_{2.5}$ sampling method in Europe (EN14907)

The sampling method of European standards, such as EN12341 and EN14907, collected the particle matters by the flow rate of 500 L min$^{-1}$. As mentioned, the air sampler was set at a high-volume, and we also used the electronic control microprocessor to stabilize the flow rate during sampling. At the same time, ambient temperature and atmospheric pressure were recorded. In the mechanism of the EN14907 method, the built-in impactor was employed. While aerosol past the sampling inlet and along the specific track, particle matters with aerodynamic diameters more than 2.5 μm would be blocked on the impactor, and those less than 2.5
μm would be collected on the filter.

2.2.3 PM$_{2.5}$ sampling method in Japan

Another high-volume air sampler was made in Japan (Sibata High-volume Air Sampler HV-1000R) (Sugita et al., 2019). In this high-volume air sampler, we set the flow rate up to 1000 L min$^{-1}$. The impactor (based on an HV-1000R 2.5 impactor, SIBATA Scientific Technology Ltd., Saitama, Japan) of this method employed an extra impactor filter. During sampling, PM < 2.5 μm was be collected on the bottom filter, and PM > 2.5 μm was trapped on the top filter.

2.3 Sample preparation

We used the quartz fiber filters and polyurethane foam (PUF) plugs for ambient air sampling to collect the solid and vapor phase compounds in PM$_{2.5}$. Before sampling, the Whatman quartz fiber filters (Grade QM-H, 150 mm diameter; 8 × 10 inch; Merck KGaA, Darmstadt, Germany) were baked for 400°C (5 hours) to remove the impurity on the filters and matrix interference. Gravimetric analysis was done after stabilizing the filter at constant humidity (45% ± 5%) and temperature (18°C) for at least 24 hr and stored with a glasses petri dish. Additionally, as with the preparation of filters, polyurethane foam (PUF) was pre-wash using Toluene by Soxhlet purification for 4 hr and reflux more than 4 cycles per hour. Before sampling, the
polyurethane foam (PUF) was added to the internal standard (Method 1613) 20 µL.

2.4 Chemical and Statistics Analysis

This study collected the PM$_{2.5}$ on the quartz fiber filters and polyurethane foam (PUF) plugs. Then, we used the Soxhlet extraction with Toluene for 16 hr. After the extraction, every sample was cleaned with an activated carbon impregnated silica gel column. Washing the filler and reagent of the silica gel column with 50 mL Hexane. Then transfer the concentrated extract sample to the column and rinse the tube with 5 mL hexane. Eluted with 40 mL toluene to completely wash the silica gel column. Last, concentrated the fraction till its volume to 50 µL under a stream of nitrogen, waiting for analysis (Bi et al., 2020). The injection volume was 1.5 µL, and the sample volume was 0.5 mL. A laboratory blank and filed blank were analyzed for quality control. Furthermore, a matrix spike sample (2.0–20 pg µL$^{-1}$ PCDD/Fs) also were analyzed after every eight samples. Atmospheric seventeen 2,3,7,8-substituted PCDD/F congeners were analyzed by high-resolution gas chromatography (TRACE GC, Thermo Scientific, USA)/high-resolution mass spectrometry (HRMS) (DFS, Thermo Scientific, USA) equipped with a fused silica capillary column DB-5 MS (60 m x 0.25 mm x 0.25 µm, J&W). The mean recovery efficiency of standard for $^{13}$C$_{12}$-2,3,7,8-substituted PCDD/Fs ranged 49 - 113%. The method of detection limits (MDL)
ranged from 0.056 to 0.362 pg μL⁻¹. The QA/QC of the PCDD/Fs analysis was followed the regulations by the Taiwan Environmental Analysis Laboratory, Taiwan EPA (NIEA A810.13B) (EPA, 2010).

The IBM SPSS Statistics 24 software calculated the Pearson coefficient. On the other hand, we also calculate the difference (%) between three sampling systems (NIEA, Europe and Japan) by the Eq.(1):

\[
\text{Difference (\%)} = \frac{HV - \text{reference}}{\text{reference}} \times 100\% \quad (\text{Eq.1})
\]

HV: Concentration of PM₂.₅ in high-volume (μg m⁻³).

reference: Concentration of PM₂.₅ in NIEA (μg m⁻³).

3. RESULTS AND DISCUSSION

3.1 Atmospheric PM₂.₅ mass concentration between different sampling methods

The metrological data with the ambient air sample in 2014-2015 was obtained during the sampling period. In this study, the wind direction is major for the northeast and southwest during the winter and summer in Taiwan. However, the wind speed was not more than 4.00 m s⁻¹. The wind direction and the wind speed would influence particle deposition. The wind speed was ranged from 0.10 - 4.00 m s⁻¹ during the sampling period. Moreover, there was construction during the sampling period in the wintertime, and it was the reason that would influence particle deposition.
Furthermore, rainy might also influence particle deposition in the atmosphere.

The geographic location of Taiwan in the subtropical leads to high temperatures in summer and northeast monsoon in winter, respectively. In our research, we had two sampling periods for the sampling method of Taiwan (NIEA) vs. Japan method and EN14907, respectively. To verify the sampling method of a different standard, we combined the data for the sampling period with NIEA vs. Japan and NIEA vs. EN14907. As a result showed the PM$_{2.5}$ mass concentration of NIEA and Japan was $26.5 \pm 10.7 \mu g m^{-3}$ and $28.6 \pm 13.9 \mu g m^{-3}$ in the winter, 2014, respectively. The difference between NIEA and Japan was around 7.92%.

On the other hand, the PM$_{2.5}$ mass concentration of NIEA and EN14907 was $20.2 \pm 8.79 \mu g m^{-3}$ and $25.4 \pm 10.5 \mu g m^{-3}$ in 2015, respectively. The same difference with the NIEA and EN14907 was calculated around 25.7%. Comparing the difference of NIEA with Japan method and EN14907, the reason for the difference was the sample size and the theory of sampling. It means the cyclone and impactor sampling method was different and probably caused particle bounce. Another reason was the sample size of NIEA with Japan method ($n = 8$) and EN14907 ($n = 34$).

Moreover, the atmospheric temperature, pressure, and humidity were also the causes, and we will discuss them in section 3.3. In the NIEA and Japan method sampling period, no rainy and high temperature caused the difference. On the other
hand, for the sampling period of NIEA and EN14907, there were spring and summer
in 2015. It means there was much uncertainty in these seasons, like the afternoon
thundershower in summer, monsoon events in spring. So that comparing the two
differences of sampling method of NIEA with Japan method and EN14907, Fig. 1(a)
showed the coefficient of determination of the sampling method. For the PM$_{2.5}$ mass
concentration of the sampling method, the coefficient of determination in NIEA and
Japan method and EN14907 were 0.825 and 0.608, respectively. The high correlation
showed no difference between these two sampling methods. For the result of the
high-volume impactor with WINS PM$_{2.5}$, the R$^2$ was higher than 0.95 (Patel et al.,
2021). Our research also obtained the high agreement in the high-volume impactor
with cyclone separator.

3.2 The variation of PCDD/Fs concentration between EN14907 and Japan
method

To compare the hazardous air pollutants in the atmosphere, we analyzed the trace
level toxicity compound, namely PCDD/Fs, for comparison. Fig. 1(b) showed the
coefficient of determination between EN14907 and Japan method (R$^2 = 0.9007$) in the
solid and vapor phase concentration with PCDD/Fs. Accroding to the Eq.(1), we also
calculate the difference between EN14907 and Japan method. Fig. 2 and 3 showed the
difference in the solid and vapor concentration of PCDD/Fs between the EN14907 and Japan methods in these sampling period, respectively. There was a stable condition in solid-phase concentration of PCDD/Fs. Especially for the high chlorine compound of PCDD/Fs (2,3,4,6,7,8-H6CDF; 1,2,3,7,8,9-H6CDF; 1,2,3,4,6,7,8-HCDF; 1,2,3,4,7,8,9-H7CDF; OCDF; range from -7 - 31%). On the opposites, it was not stable in the vapor phase concentration of PCDD/Fs.

The fluctuation in vapor phase dioxin was limited in sample numbers and the capture efficiency could also be caused by the sampling flow rate, the size of sampling media (PUF) of each sampling system, even the vapor pressure of PCDFs (Fig. 2 and 3). Compared with the mass concentration of EN14907 and the Japan method, it was no difference between these two theories of sampling method (mass concentration of EN14907 was 32.3 μg m⁻³; mass concentration of Japan method was 32.2 μg m⁻³). Moreover, the result of chemical composition showed the concentration of PCDD/Fs in ambient air sample were also no different. The average PCDD/Fs concentration in solid and vapor phase at EN14907 method was 560 fg m⁻³ and 50.4 fg m⁻³, respectively. Furthermore, the concentration of PCDD/Fs in solid and vapor phase at Japan method was also 598 fg m⁻³ and 47.3 fg m⁻³, respectively. It was a similar PCDD/Fs concentration trend between EN14907 and the Japan method. 

Besides, Sugita et al. (2019) evaluated the chemical compositions with high-volume
air sampler in PM$_{2.5}$. It was showed higher R$^2$ in the water-soluble ions and PAHs. In the methodology of sampling with environmental media that was a lack of puf in the pollutants of vapor phase. There was also set the lower flow rate (700 L min$^{-1}$) in the high-volume air sampler. For the time resolution and analysis limited of trace level pollutants, we set the flow rate up to 1000 L min$^{-1}$. Moreover, we also used the PUF to collect the vapor phase of PCDD/Fs.

Summary for the results of mass and concentration of PCDD/Fs, there was no difference in these two sampling methods. But there was still some limitation in our research. Because of the seasonal difference in metrological, the species of PCDD/Fs distribution was different. Especially the convection of atmospheric was influence the metrological. Hence, there was the influence of typhoon Soudelor. That was also the reason why the concentration of PCDD/Fs was different between summer and winter.

### 3.3 Confounders in different methods

Expect the correlation between these three air sampling methods. We also used the statistical software (SPSS 24) to analyze the correlation and Pearson coefficient with the temperature, wind speed, PM$_{2.5}$ concentration of three sampling methods and humidity (Table 1). The statistical analysis result showed a significant association between NIEA and EN14907 and Japan method (Pearson coefficient of PM$_{2.5}$
concentration (NIEA) and EN14907 and Japan method = 0.777**; 0.908**).

Simultaneously, there was also a significant difference between EN14907 and Japan method (Pearson coefficient = 0.913**). It meant that the high-volume air sampling method of Europe and Japan could replace NIEA. On the other hand, there was a negative significantly related PM$_{2.5}$ concentration (NIEA) and temperature (pearson coefficient = -0.358*). However, there was still differentiated result at the extreme value because of the different situations in the atmospheric metrological. Fig. 4 showed the confounder's indifference of these sampling methods. In Fig. 4 (a), when the temperature range was 15-20°C, there was a stable difference. When the range of temperature was 20-35°C there had a wild range difference in the temperature. Fig. 4 (b) and Fig. 4 (c) showed there was no certain range of humidity and wind speed that would influence the difference between the sampling methods. Additionally, there was a significant difference of lower wind speed. Finally, when the concentration of PM$_{2.5}$ measured by NIEA method was higher than 30 µg m$^{-3}$ or lower than 20 µg m$^{-3}$ that showed a stable difference. However, there still had the outliner point of difference that exits in our research because of the uncertainty in the atmospheric metrological (Fig. 4 (d)) condition. Additionally, the metrological confounders were defined as the major reason for PM$_{2.5}$ concentration. The sampling volume was also one of them.

Traditionally, the sampling volume was calculated by average flow rate before and
after sampling multiplied by total sampling times. There was a lack of precision of volume calculation. In our study, we found the concentration of PM$_{2.5}$ would be higher in the Japan method mostly. There was a similar trend of PM$_{2.5}$ mass concentration between the method of NIEA and EN14907. Because of the theory of sampling and air sampler that difference would be obtained on these three air sampling methods. For the trending of ultra pollutants in the atmosphere, it was necessary to develop the method of high-volume air sampling.

4. CONCLUSION

In this study, we evaluated the difference in the method of high-volume air sampling to analyze the chemical compounds. Especially focus on the 17 PCDD/Fs. The ambient air samples were compared to the concentration of PM$_{2.5}$ and PCDD/Fs. The correlation and pearson coefficient of PM$_{2.5}$ in the sampling method of Taiwan (NIEA) vs. EN14907 and Japan method were similar. It means that there was no wild rang of difference between the three sampling methods. The coefficient of NIEA, Japan method, and EN14907 were 0.825 and 0.608, respectively. Additionally, the correlation of PCDD/Fs between EN14907 and Japan high-volume sampling methods was higher than 0.90. Moreover, compared to those high-volume air sampling methods in PCDD/F measurements, there was no significant difference (-7-31%) with
in the solid-phase PCDD/F measurement between EN14907 and Japan methods. Besides, metrological was also one of the confounders leading to the difference between high-volume air sampling methods. Specifically for temperature and wind speed (negative correlation around -0.358 to -0.495 with a significant level < 0.05).

Based on the results obtained from this study, it showed higher interrelationship of PCDD/Fs in PM$_{2.5}$ can be measured via the high-volume air sampling methods. For the trace level pollutants and chemical compounds in the PM$_{2.5}$ in the atmosphere that is necessary to develop the high-volume air sampling.

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**DISCLAIMER**

There are no real or perceived financial conflicts of interests for any author.
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Table 1. Pearson coefficient of confounders in the PM$_{2.5}$ concentration.

<table>
<thead>
<tr>
<th>Pearson Coefficient</th>
<th>PM$_{2.5}$ (NIEA, Taiwan)</th>
<th>PM$_{2.5}$ (EN14907, Europe)</th>
<th>PM$_{2.5}$ (Japan)</th>
<th>Temperature</th>
<th>Wind Speed</th>
<th>Humidity</th>
</tr>
</thead>
<tbody>
<tr>
<td>PM$_{2.5}$ (NIEA, Taiwan)</td>
<td>1</td>
<td>0.777**</td>
<td>0.908**</td>
<td>-0.358*</td>
<td>-0.023</td>
<td>-0.251</td>
</tr>
<tr>
<td>PM$_{2.5}$ (EN14907, Europe)</td>
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<td>0.913**</td>
<td>-0.257</td>
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<tr>
<td>PM$_{2.5}$ (Japan)</td>
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<tr>
<td>Temperature</td>
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<td>-0.495**</td>
<td>-0.009</td>
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<tr>
<td>Wind Speed</td>
<td>1</td>
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<td>Humidity</td>
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** Significant level < 0.01  
* Significant level < 0.05
Figure 1. The coefficient of NIEA, Japan method and EN14907 on PM$_{2.5}$ mass concentration and PCDD/Fs concentration.
Figure 2. The difference in the solid phase concentration of PCDD/Fs between the EN14907 and Japan method (n=7).
Figure 3. The difference in the vapor phase concentration of PCDD/Fs between the EN14907 and Japan method (n=3).
Figure 4. The difference of sampling method between confounders (including (a) temperature, (b) humidity, (c) wind speed, and (d) PM$_{2.5}$ mass concentration) in the atmosphere.