Composition of $PM_{2.5}$ and Variation of Atmospheric PCDD/Fs in Northern Taiwan during Winter Monsoon and Local Pollution Events in 2015-2016

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Introduction

Polychlorinated dibenzo-p-dioxin and dibenzofurans (PCDD/Fs) are persistent organic pollutants are persistent organic pollutants (POPs) that are formed and released unintentionally from anthropogenic sources [1-3]. A previous study [1] has indicated that higher than 60% of PCDD/Fs in the atmosphere are essentially bounded to particles. In regions with low or no precipitation or during times of low or no precipitation, airborne PCDD/Fs are transported more effectively. Among 210 PCDD/F congeners, 17 have chlorine substitution in the 2, 3, 7, and 8 positions, are toxic to the human endocrine system, can cause adverse health effects [3,4]. Moreover, due to their susceptibility to seasonal weather changes, they undergo atmospheric long-range transport (LRT) which can lead to cross-border pollution [1]. Taiwan is an island in the subtropics that is located off the southeast coast of Mainland China. In the winter and spring (October to March), the country and its surrounding areas are often influenced by the northeasterly winter monsoon winds that originate from Central Asia [5]. The winter monsoon not only brings cold air to Taiwan, but also transports air pollutants and dust over long distances to Taiwan as well as throughout the Northwestern Pacific area [6-8]. When monsoons and cold-air masses reach Taiwan, their speeds and directions change in a manner that weakens them because of the geographical uniqueness of the island. Poor atmospheric dispersion is resulting in rapid accumulation of locally emitted pollutants in the atmosphere and bring about local pollution (LP). This study investigated the effects of the long-range transport of air pollutants by northeastern monsoons and local pollution episodes on atmospheric $PM_{2.5}$, PCDD/Fs concentrations and composition in northern Taiwan during the winters of 2015 and 2016.

Materials and methods

To investigate the chemical composition of $PM_{2.5}$ in ambient, four sampling sites, named Urban, Suburban, Rural 1 and Rural 2 (Fig. 1), were selected in northern Taiwan during the northeastern monsoon period in 2015 and 2016. During winter monsoons, Rural 1 (Fugui Cape) was in northern Taiwan are thus in the direct path of the long range transportation (LRT), whose air-current trajectory covered northern China, Japan, and Korea. The sampling procedure followed by European Union EN-14907 $PM_{2.5}$, and used high volume sampler (Analitica HVS-$PM_{2.5}$) equipped with Whatman quartz fiber filters (150 mm) and polyurethane foam(PUF) set the flow rate at 500L/min. The total volume of the air sample was more than 700 m^3 for a typical sampling duration in 24 hours. In this study, the seventeen 2,3,7,8-substituted PCDD/F congeners were analyzed by high-resolution gas chromatography (HRGC)/high-resolution mass spectrometry (HRMS) (JEOL JMS-700). The quartz fiber filter was used to determine a range of elements (Al, Fe, Na, Mg, K, Ca, Sr, Ba, Ti, Mn, Co, Ni, Cu, Zn, Mo, Cd, Sn, Sb, Tl, Pb, V, Cr, As, Y, Se, Zr, Ge, Rb, Cs, Ga, La, Ce, Nd) by inductively coupled plasma-mass spectrometry (ICP-MS) (NexIon 300X, Perkin-Elmer). About water-soluble ions (Na^+, $NH4^+$, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, PO_4^{3-}, SO_4^{2-}) was analyzed by ion chromatography (IC). Organic carbon (OC) and elemental carbon (EC) were measured with IMPROVE_A thermal/optical reflectance (TOR) protocol. Besides, positive matrix factorization (PMF) model, potential source contribution function (PSCF) and source regional apportionment (SRA) were used to identify and quantify the emission sources and potential source area.

Results and discussion

The results showed the hourly air quality and meteorology data from Taiwan Air Quality Monitoring Network in Table 1. Compared with normal periods, the concentration of $PM_{2.5}$, NO_x, CO, O_3 and wind speed were higher and ambient temperature was lower during LRT event. In Table 2, the highest $PM_{2.5}$ concentration were measured in Urban (27.7±19.6 μg/m^3) and Suburban site (46.4±6.62 μg/m^3) during LRT and LP, respectively. However, the results in Urban site were not only with the highest PCDD/F concentration measured during both LRT (55.2±36.3 fg I TEQ/m^3) and LP (95.1±48.1 fg I TEQ/m^3), but also with the highest PCDD/F content in $PM_{2.5}$ (2,178-3,779 pg I-TEQ g^{-1}) during all period. According to the distribution of PCDD/F congeners in Figure 2, the significantly higher distribution of PCDFs were measured at all sampling site. However, the higher contribution of PCDDs were observed at Rural 1 and 2 sites during normal periods. At urban site, there is not any significantly different of the PCDD/F congener distribution between LRT, LP and normal period. Therefore, the impact of air quality in rural and suburban sites was quite serious than urban site during the LRT and LP events. All the measurements indicated that the atmospheric PCDD/Fs measured in this study were all lower than the air quality standards for PCDD/Fs in Japan (0.6 pg-TEQ/m^3). The composition of water-soluble ions (WSI) measured in $PM_{2.5}$ collected in this study were shown in Figure 3. The main component were SO_4^{2-}, NO_3^-, NH_4^+ and Na^+ in WSI and crustal element such as Al, Fe, Na, Mg, Ca and K in trace metal, especially for urban site. The result of PMF analysis indicated that the atmospheric PCDD/Fs in northern Taiwan around 7.8%, 32.6%, 8.5%, 45.8% and 5.3% were provided from crematorium, coal-fired power plant and sinter plant, traffic emission, long range transport event and waste incinerator, respectively. In addition, the results of SRA analysis (Table 3) suggested around 80% of PCDD/Fs transported from Mongolia and China during LRT period. The results of this study serve as a reference for further research exploring the effect of LRT and LP on air quality impact and human health.

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Table 1. Statistics of weather and air quality condition (hourly data) measured in northern Taiwan for LRT, LP and normal periods.

	n	**$PM_{2.5}$** ($\mu g/m^3$)	**WS** (m/s)	**RH** (%)	**Temperature** (℃)	**SO_2** (ppb)	**NO_x** (ppb)	**CO** (ppm)	**O_3** (ppb)
LRT	697	19.8±13.0	5.43±2.90	79.1±11.0	17.2±4.57	2.01±2.12	5.35±3.22	0.31±0.11	45.5±9.51
LP	361	15.8±13.1	2.21±1.38	81.7±10.0	18.5±3.25	2.50±1.57	9.53±6.59	0.30±0.10	35.9±15.1
Normal	169	6.25±4.46	3.81±2.50	84.2±7.35	21.9±4.07	2.51±1.81	5.78±3.88	0.16±0.06	34.4±14.1

Table 2. $PM_{2.5}$ and atmospheric PCDD/F concentrations measured at different sampling site.

	$PM_{2.5}$ ($\mu g/m^3$)			**PCDD/Fs** (fg I-TEQ/m^3)		
	LRT	**LP**	**Normal**	**LRT**	**LP**	**Normal**
Rural 1	22.4±10.1	23.8±12.8	9.11±4.86	29.1±19.1	66.6±47.9	7.65±5.32
Rural 2	15.4±9.12	27.9±11.7	5.92±1.55	10.6±6.08	8.9±1.96	3.41±1.66
Suburban	18.4±13.3	**46.4±6.62**	8.01±4.87	12.5±8.27	27.2	22.7±21.3
Urban	**27.7±19.6**	29.1±13.2	7.26	**55.2±36.3**	**95.1±48.1**	20.8

Table 3. Regional contributions for each emission source via SRA analysis during LRT event.

Location	**Factor 1** Crematorium 7.8%	**Factor 2** Coal-fired power plant and sinter plant 32.6%	**Factor 3** Traffic emission 8.5%	**Factor 4** Industrial emission 45.8%	**Factor 5** Waste incinerator 5.3%	**Total**
Mongolia & Northern China	0.54%	2.27%	0.59%	3.18%	0.4%	**6.6%**
Western China	0.13%	0.56%	0.15%	0.79%	0.1%	1.6%
Central China	6.01%	25.3%	6.60%	35.4%	4.1%	**73.3%**
Northeast Asia	0.18%	0.74%	0.19%	1.04%	0.1%	2.2%
Southeast Asia	0.33%	1.39%	0.36%	1.95%	0.2%	**4.0%**
Taiwan	0.45%	1.90%	0.50%	2.66%	0.3%	**5.5%**
Pacific Ocean	0.12%	0.51%	0.13%	0.72%	0.1%	1.5%

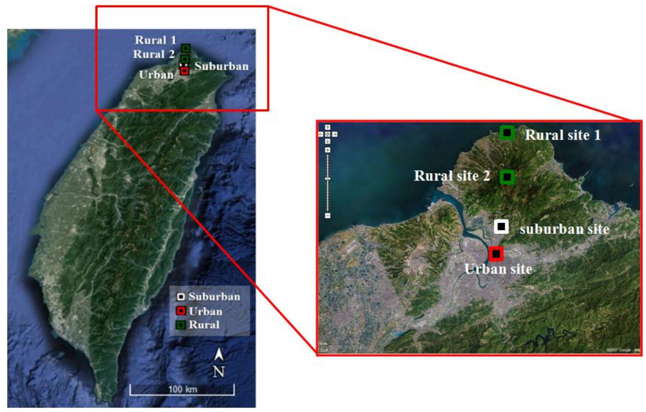

Figure 1. Locations of atmospheric sampling sites selected in Taiwan.

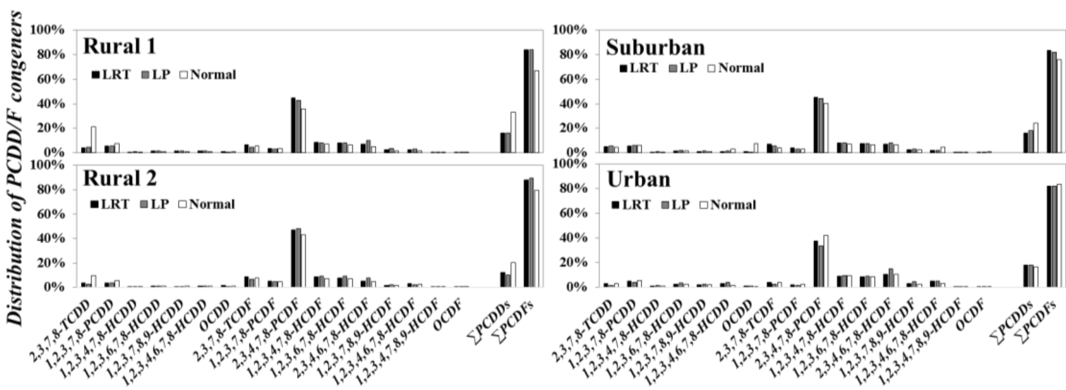

Figure 2. Distribution of PCDD/F congener measured in northern Taiwan during LRT, LP and Normal period.

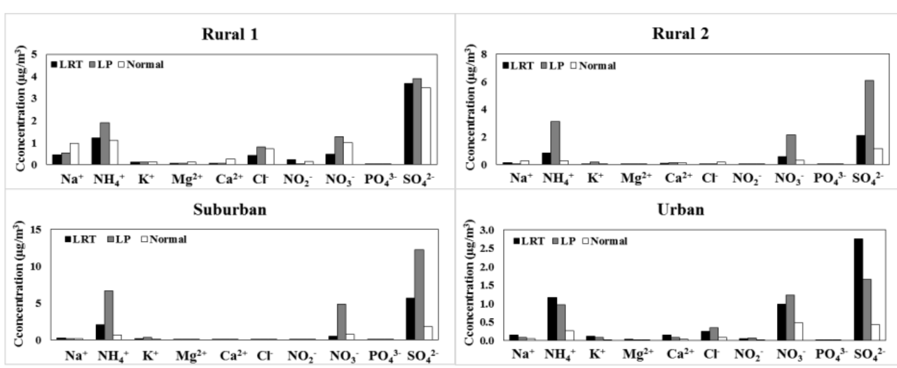

Figure 3. Composition of water-soluble ions in $PM_{2.5}$ measured in northern Taiwan during LRT, LP and Normal period.