

PCDD/F Measurement at a High-Altitude Station in Central Taiwan: Evaluation of Long-Range Transport of PCDD/Fs during the Southeast Asia Biomass Burning Event

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Recent biomass burning in Southeast Asia has raised global concerns over its adverse effects on visibility, human health, and global climate. The concentrations of total suspended particles (TSPs) and other vapor-phase pollutants (CO and ozone) were monitored at Lulin, an atmospheric background station in central Taiwan in 2008. To evaluate the long-range transport of persistent organic pollutants (POPs) during the Southeast Asia biomass burning event, the atmospheric polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) were also measured at Lulin station. The atmospheric PCDD/F and TSP concentrations measured at Lulin station ranged from 0.71–3.41 fg I-TEQ/m³ and 5.32–55.6 µg/m³, respectively, during the regular sampling periods. However, significantly higher concentrations of PCDD/Fs, TSPs, CO, and ozone were measured during the spring season. These high concentrations could be the result of long-range transport of the products of Southeast Asia biomass burning. During the Southeast Asia biomass burning event (March 18–24, 2008), an intensive observation program was also carried out at the same station. The results of this observation program indicated that the atmospheric PCDD/F concentration increased dramatically from 2.33 to 390 fg I-TEQ/m³ (March 19, 2008). The trace gas (CO) of biomass burning also significantly increased to 232 ppb during the same period, while the particle-bound PCDD/Fs in the TSP increased from 28.7 to 109 pg I-TEQ/g-TSP at Lulin station during the burning event. We conclude that there was a significant increase in the PCDD/F concentration in ambient air at a high-altitude background station in central Taiwan during the Southeast Asia biomass burning event.

1. Introduction

It is well-known that the main areas of wild biomass burning lie in the tropical and subtropical areas of Africa, South America, Southeast Asia, Indonesia, and Australia. During the past decade, Southeast Asia biomass burning has raised global concerns over its adverse effects on visibility, human health, and global climate, due to the emission of particulate matter and other gaseous pollutants such as CO, SO_x, NO_x, and volatile organic compounds (1). The impact of biomass burning on carbon monoxide (CO) is easier to measure. Biomass burning CO emissions amount to 500–700 Tg/year, or ~25% of the global CO budget, according to inverse model results by Bergamaschi et al. (2). In the summer of 1995 large forest fires in northern Canada caused 52–74% of the variance in ground level CO measurements and were responsible for 10 to 30 ppb ozone enhancements throughout the southeastern United States over a two week period (3, 4). Additionally, forest fires in Canada caused 58% of the CO enhancement in Mace Head, Ireland in 1998 (5).

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) are classified as persistent organic pollutants (POPs). PCDD/Fs are formed during combustion processes in the presence of carbon source, oxygen, chlorine, and catalysts (especially copper and iron). Limited data suggest that field burning of agricultural crops and woods can result in formation and emission of PCDD/Fs (6, 7). The draft United States inventory of PCDD/F sources in 2000 indicated that about 6% of the estimated total annual emissions come from forest fires (8). Different combustion conditions, such as wind speed conditions and fuel moisture, can also affect the emissions (9). A previous study (10) also indicated that the PCDD/F emission factor during biomass burning ranged from 15 to 25 ng I-TEQ/kg burned. The mass-specific emissions are about 20 times higher than the concentration in the extracted biomass, suggesting that PCDD/F emissions are not simply a result of vaporization of cuticle-bound PCDD/Fs but are formed predominantly during biomass combustion. These particles are introduced into the atmosphere through wind blowing and eventually settle to water bodies or other receptors in the environment via either dry or wet deposition mechanisms (11). In this study, we monitored the concentrations of particulate matter and 17 2,3,7,8-substituted PCDD/Fs in central Taiwan using high-volume ambient air samplers at Lulin atmospheric background station. The objective of this study was to evaluate the effects of Southeast Asia biomass burning on the atmospheric concentrations of PCDD/Fs.

2. Experimental Section

2.1. Sampling Sites. In order to measure the long-range transport of PCDD/Fs, a high-altitude sampling site was selected based on the meteorological information and location relative to the biomass burning in Southeast Asia (12). The Lulin atmospheric background station (http://lulin.tw/index_en.htm) is located at the peak of Mt. Lulin (23.51°N, 120.92°E; 2862 m above mean sea level). Mt. Lulin is surrounded by other mountainous areas of relatively low altitude, and only Mt. Jade (3954 m above mean sea level), situated to the northeast and several kilometers away, is higher than this site. Its high elevation means it is generally free from local pollution. The Lulin station is unique because its location and altitude can enhance the global network of Global Atmosphere Watch (GAW) in the Southeast Asian region where no high-elevation baseline station is available. The Lulin station generally lies in the free troposphere

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(especially during the winter months) and is free from boundary layer pollution. Frontal mechanisms, which frequently occur in winter and spring of Taiwan, have been reported to be able to transport pollutants from the boundary layer to the free troposphere (12). Since April 16, 2006, the continuous operations included atmospheric radiation, meteorological parameters, trace gases (CO and ozone), and aerosol (PM_{10}) had been measured on Lulin station. In this study, CO (APMA-360, Horiba, Japan) and Ozone (EC-9810B, Ecotech, Australia) instruments were operated on the second floor of Lulin station. Eight Dekoron tubings (1/4" O.D., Synflex, USA) were bundled together as the air intake line extruding to the roof and the inlet point at about 10 m above the ground. An oil-free sirocco fan (JSD-30S, Jouning, Taiwan) provided a gas flow rate of approximately $2.0 \text{ m}^3/\text{min}$ to the gas analyzers.

2.2. Trajectory and Tracer/Dynamic Models. In order to identify the sources of the Southeast Asia biomass burning episode that occurred on March 2008 and to examine how transport paths could affect the atmospheric PCDD/F concentration in central Taiwan, the HYSPLIT (Hybrid Single-Particle Lagrangian-Integrated Trajectory) model (13) was used to trace the origins of the air masses. Five-day back trajectory analyses using HYSPLIT at the altitudes of 3 km from the location of Lulin station in central Taiwan. Furthermore, we employed the WRF/Chem (Ver.3.0) (14) modeling system with a tracer module (15) to identify the long-range transport associated with biomass burning over Indochina in our case study. The tracers were assigned to the fire locations derived from MODIS satellite data over Indochina ranging from 5 to 25°N and 90 to 110°E . The Yonsei University (YSU) (16) planetary boundary layer scheme was selected in this study. The horizontal resolution for our simulations was 27 km, and the grid box had 200×200 points in both the east–west and north–south directions. There were 35 vertical levels, and the lowest level was about 20 m above the surface. To analyze the effects of the Southeast Asia biomass burning event in Taiwan during the period of March 2008, on the levels of ambient PCDD/F compounds, five-day back trajectory analyses are calculated using HYSPLIT at the altitudes of 3 km from the location of Lulin station in central Taiwan for the event of March 19, 2008 (Figure 1) suggested that the air masses in central Taiwan originated from Indochina on March 15–17, at a lower elevation than the trace layer. Moderate Resolution Imaging Spectroradiometer (MODIS) satellite (1 km resolution) data (Figure 1) show significant active fire detections occurred during March 15–17.

2.3. Sample Collection and Analysis. In this study, all PCDD/F and TSP samples were taken at Lulin station during the regular sampling periods (March, June, September, and November 2008) for the analysis of PCDD/Fs with 6 to 11 samples taken each month. During a special long-range transport event (2008/3/18–3/24), one sample was taken everyday for 24 h. Ambient air samples for the both vapor and solid phases of PCDD/F compounds were collected using high-volume sampling trains (Shibata HV-1000F). The HV-1000F samplers were equipped with Whatman quartz fiber filters (20.3 cm \times 25.4 cm) for collecting particle-bound compounds, and polyurethane foam (PUF) plugs were used to retain PCDD/F compounds in the vapor phase. The PUF and filter samples were then Soxhlet extracted with toluene for 24 h, treated with concentrated sulfuric acid, and then passed through a series of cleanup columns containing sulfuric acid-silica gel, acidic aluminum oxide, and Celite/carbon. Finally, the dioxin congeners were analyzed using high resolution gas chromatography (HRGC) (Thermo Trace GC) /high resolution mass spectrometer (HRMS) (Thermo DFS) using a fused silica capillary column DB-5 MS (60 m \times 0.25 mm \times 0.25 μm , J&W). The Thermo DFS used in this study is a high resolution magnetic sector mass spectrometer. The DFS has completely new, state-of-the-art electronics. It

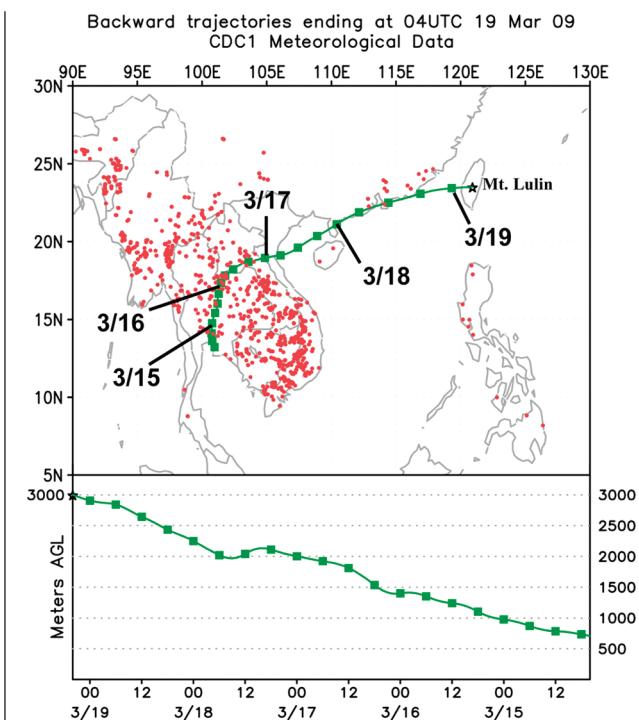


FIGURE 1. Five-day backward trajectory of Lulin station started at 04 UTC March 19, 2008 and active hotspot/fire observation (red dots) by MODIS during air masses passed through Indochina on March 15–17, 2008.

uses dedicated microcontrollers on all boards, enabling efficient and fast computer control and read-back of all important parameters and voltages through a common interconnect bus. The mass spectrometer was operated with a resolution greater than 10,000 under positive EI conditions, and the data of 17,2,3,7,8-substituted PCDD/F congeners were analyzed in the selected ion monitoring (SIM) mode. A laboratory blank and matrix spike sample (2.0–20 $\text{pg}/\mu\text{L}$ PCDD/Fs) were used in the analytical procedure for every eight samples for quality control. Method detection limits (0.02–0.10 fg/m^3) were determined from the blanks and quantified as three times the standard deviation of the concentration in the blanks. In this study, the concentrations of all laboratory blank samples were <1.15 pg for PCDD/Fs. The mean recoveries of standards for all $^{13}\text{C}_{12}$ -2,3,7,8-substituted PCDD/Fs ranged from 48–111%. The analyzed results were all within the acceptable 40–130% range for the U.S. EPA Method 23.

3. Results and Discussion

3.1. Measurement of Atmospheric PCDD/F Concentration during the Regular Sampling Periods. To investigate the variation of atmospheric PCDD/F concentration at Lulin atmospheric background station during different seasons. Based on the meteorological condition observed at Lulin station, the regular sampling periods in this study were set up at spring (March 2008), summer (June 2008), fall (September 2008), and winter (November 2008). During the regular sampling periods, the atmospheric PCDD/F and TSP concentrations measured at Lulin station range from 0.71–3.41 $\text{fg I-TEQ}/\text{m}^3$ and 5.32–55.6 $\mu\text{g}/\text{m}^3$, respectively (Figure 2). The lowest concentrations were measured during the summer season (June 10–17, 2008). In some developed countries including the U.S. and U.K., the PCDD/F concentrations in ambient air in the urban area were about 10 to 50 $\text{fg I-TEQ}/\text{m}^3$ (17, 18). In some Asian countries (like Korea and Japan), the atmospheric PCDD/F concentrations in the urban area ranged from 28 to 120 $\text{fg I-TEQ}/\text{m}^3$ (19, 20). Our

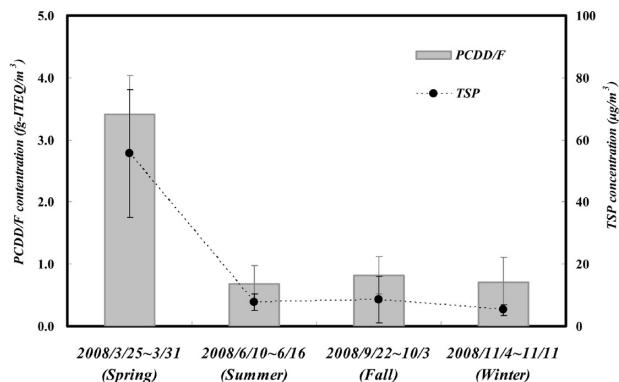


FIGURE 2. Atmospheric PCDD/F and total suspended particle (TSP) concentrations measured at Lulin station during different periods.

previous study (21) also indicated that the atmospheric PCDD/F concentrations measured in the urban area in Taiwan ranged from 20 to 110 fg I-TEQ/m³. Recent studies further indicate that around 20–790 fg I-TEQ/m³ PCDD/Fs are measured in the coastal province of China (22, 23). The atmospheric PCDD/F concentration measured at Lulin station were considerably lower than those measured in other countries. Compared to other relevant studies measured at the background station, the atmospheric PCDD/F concentrations (0.71–3.41 fg I-TEQ/m³) measured at Lulin Mountain in central Taiwan were significantly lower than the measurements in the primeval forest in Denmark (6.15–24.0 fg I-TEQ/m³) significantly lower than (24) and a regional park of Simbruini Mountains in Italy (1.48–6.57 fg I-TEQ/m³) (25). The low atmospheric PCDD/F concentration can be attributed to the lack of dioxin emissions and combustion

sources within almost 50 km of the station at Lulin Mountain. However, a significant increase of PCDF compounds in ambient air is measured during the spring, and the highest concentration of atmospheric PCDD/Fs and TSPs was observed at Lulin station during the spring season.

Figure 3 shows the average concentrations of CO, ozone, and PM₁₀ measured at Lulin station in 2008 are about 122 ± 33 ppb, 33 ± 10 ppb, and 9.4 ± 5.1 µg/m³, respectively. The lower concentrations of CO, ozone, and PM₁₀ about 84 ± 13 ppb, 24 ± 3.0 ppb, and 7.0 ± 1.4 µg/m³, respectively, were measured during the summer season (June, July, and August). The background concentrations of CO, ozone, and PM₁₀ are estimated 82 ppb, 28 ppb, and 6.0 µg/m³, respectively. However, the concentrations of above three pollutants (CO: 200 ppb, ozone: 55 ppb, PM₁₀: 24 µg/m³) observed in March show twice higher than their background values. A previous study (12) also indicated that a similar K⁺ concentration, as a biomass burning tracer, was measured at Malaysia and Lulin station in the spring season. The concentrations of anthropogenic species such as nonsea-salt SO₄²⁻, NO₃⁻, and NH₄⁺ tend toward the high side when they are compared with those of Asian measurements. Hence, the concentrations of chemical species were elevated in the spring months at Lulin station, due to emissions from south/southeast Asia and peak biomass burning activities.

3.2. Variation of Atmospheric PCDD/F Concentration during a Southeast Asia Biomass Burning Event. The satellite data and air mass paths (Figure 1) revealed that the air masses possibly came from biomass-burning regions for this episode. The air mass of Southeast Asia biomass burning moved across central Taiwan in high altitude between March 18 and 21, 2008. An intensive observation program was carried out at the same time at Lulin station. The TSP concentrations

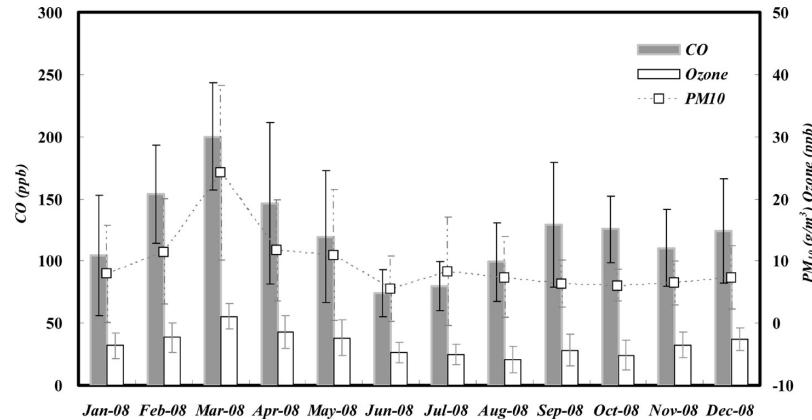


FIGURE 3. Average concentrations of CO, ozone, and PM₁₀ measured at Lulin station in 2008.

TABLE 1. PCDD/F Concentrations Measured at Lulin Station during a Southeast Asia Biomass Burning Event

date	2008/3/18	2008/3/19	2008/3/20	2008/3/21	2008/3/22	2008/3/23	2008/3/24
sampling period (start)	0000 UTC March 18, 2008	0000 UTC March 19, 2008	0000 UTC March 20, 2008	0000 UTC March 21, 2008	0000 UTC March 22, 2008	0000 UTC March 23, 2008	0000 UTC March 24, 2008
sampling period (terminate)	0000 UTC March 19, 2008	0000 UTC March 20, 2008	0000 UTC March 21, 2008	0000 UTC March 22, 2008	0000 UTC March 23, 2008	0000 UTC March 24, 2008	0000 UTC March 25, 2008
sampling volume (m ³)	838	845	847	846	846	846	850
ambient temperature (°C)*	8.9	7.2	7.0	8.4	7.8	5.5	3.3
rainfall (mm)	0.0	0.0	0.0	0.0	0.0	38.0	3.4
CO (ppb)*	225	195	232	216	213	190	199
ozone (ppb)*	67.9	61.4	54.5	52.7	53.4	59.2	58.6
TSPs (µg/m ³)	89.4	53.3	77.8	76.5	81.9	59.2	61.4
PCDD/F concentration (fg I-TEQ/m ³)	2.33	390	25.9	13.2	7.92	4.14	3.32

* Average value in 24 hours sampling data.

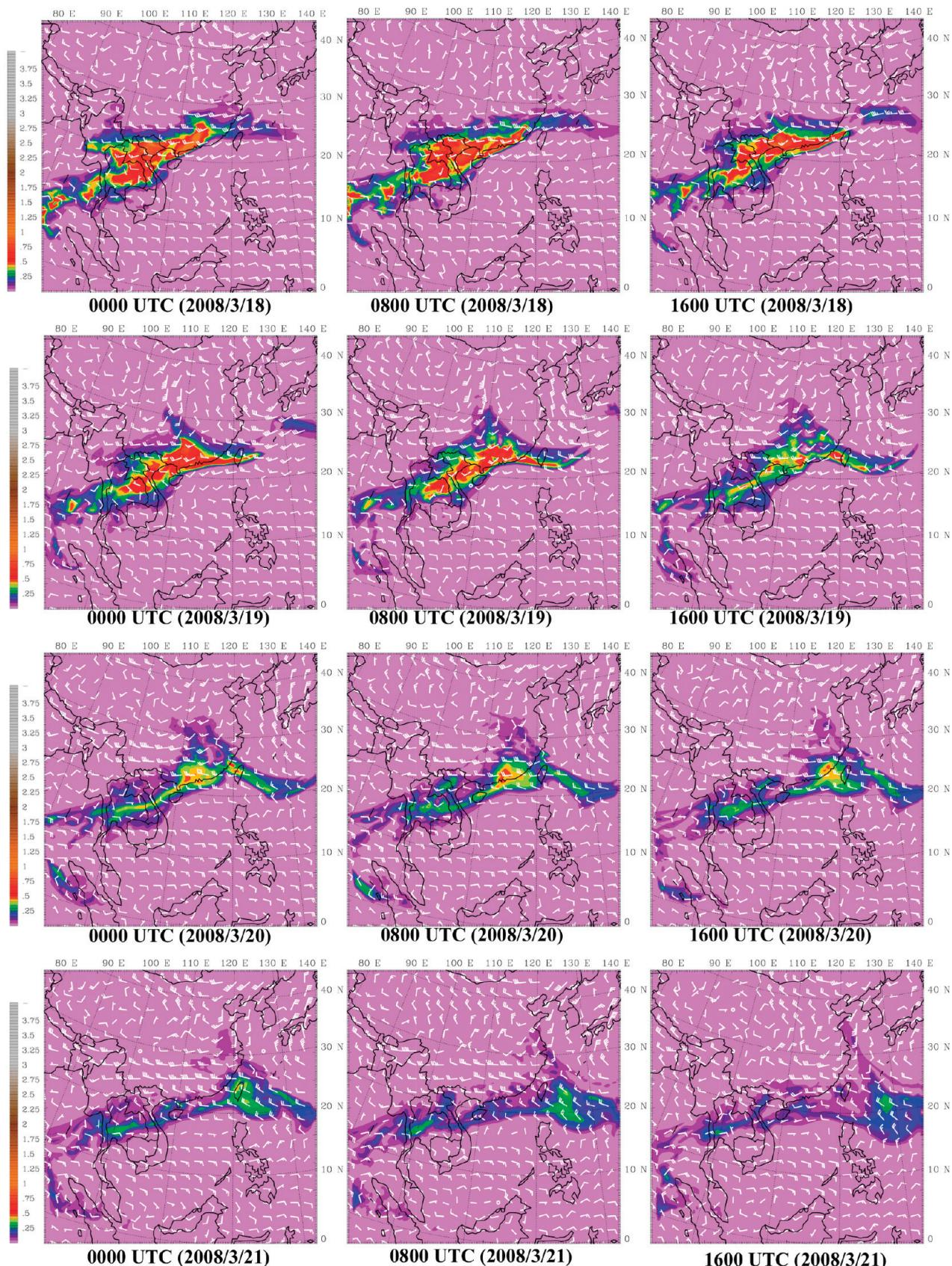


FIGURE 4. Simulated distributions of tracer concentration (colored) and wind field at 700 hPa (00:00UTC March 18, 2008 to 1600 UTC March 21, 2008).

measured at Lulin station during the Southeast Asia biomass burning event ranged from 53.3–89.4 $\mu\text{g}/\text{m}^3$ (Table 1). This was significantly higher than the TSP levels measured during the regular sampling periods (5.32–55.6 $\mu\text{g}/\text{m}^3$). In addition,

the CO and ozone concentration observed at Lulin station during the Southeast biomass burning event ranged from 190–232 ppb and 52.7–67.9 ppb, respectively (Table 1). Figure 2 also shows the area of intense fires in Southeast Asia

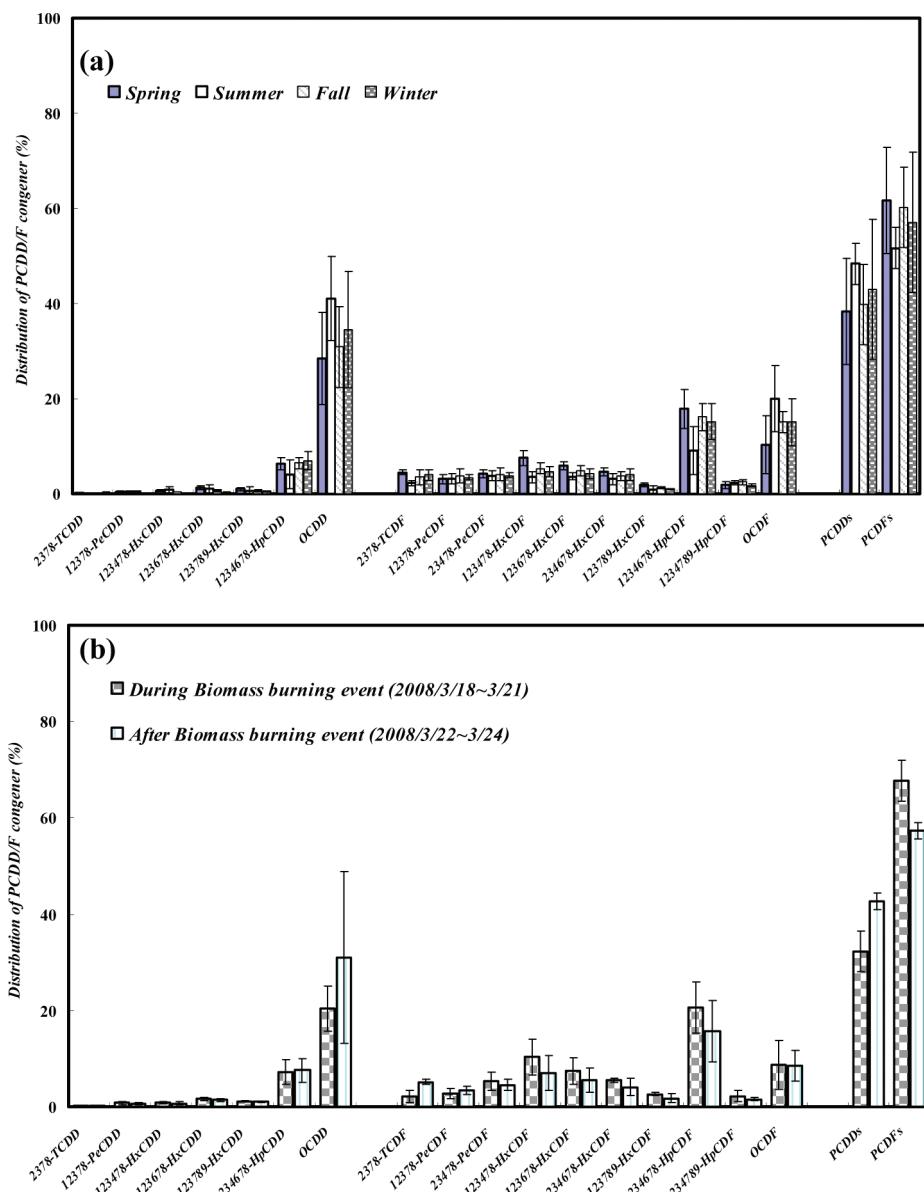


FIGURE 5. PCDD/F congener distribution in ambient air at Lulin station during (a) regular sampling periods and (b) the Southeast Asia biomass burning event.

area during March 15–17, 2008. The trajectory of the air mass during the period March 15–19 showed that the air mass originating from biomass burning areas potentially carry pollutants to the Lulin station. Hence, the high TSP and CO concentrations measured at Lulin station were attributable to the Southeast Asia biomass burning. Based on the results of the intensive observation program (Table 1), the atmospheric PCDD/F concentration increased dramatically from 2.33 to 390 fg I-TEQ/m³ (March 19, 2008) at Lulin station during the Southeast Asia biomass burning event. However, the atmospheric PCDD/F concentration decreased dramatically 2 days after the biomass burning event. Based on the observation of trace gases (Figure 3), the average concentrations of CO and ozone measured at Lulin station on February 2008 were 154 and 38.4 ppb, respectively. In general, CO has a relatively long lifetime in the atmosphere (between about 2 weeks and 2 months) (26) and thus may be used as an important pollution tracer. A previous study (27) also concluded that high CO emissions were the signature of biomass burning. In this study, the significant increases in levels of the trace gases were observed during the Southeast Asia biomass burning event. Our results also indicated that

the CO concentration fell sharply, 10 days after the Southeast Asia biomass burning event.

According to the trajectory analysis (Figure 1) the air masses passed over Indochina during March 15 and 17 before reaching Taiwan. To further investigate the impact of Southeast Asia biomass burning on East Asia, we conducted tracer simulations with WRF/Chem with tracers placed from March 15–17 over the fire locations reported by the MODIS satellite. The simulated tracers were placed at the first level above the surface at each fire location with a concentration of 10,000 units per day (416.67 units/h). The meteorological initial and boundary conditions for WRF/Chem were obtained from NCEP-FNL data sets at 6-h intervals. Figure 4 depicts the horizontal distribution of tracer concentration and wind field at the level of 700 hPa (around 3 km). The relatively strong wind belt (wind speed >10 m/s) at 700 hPa ranges from 20 to 30° N (Figure 4) and merged into a higher-latitude strong-wind belt over East Asia's marginal seas. At 00:00 UTC March 19, the high concentration tracers were transported to Taiwan following the strong wind belts and continued about the whole day. The tracer concentration gradually diluted after March 20. Our modeling study

predicted very well comparing with the sampling at the Lulin mountain station. The simulation results identified the sources of the high PCDD/F concentration are attributed to the biomass burning from the Indochina.

During the regular sampling periods (Figure 5a), major contributors of atmospheric PCDD/Fs measured at Lulin station include 1,2,3,4,6,7,8-HxCDF (9–18%), OCDF (10–20%), and OCDD (29–41%). In addition, the PCDFs account for 52–62% of total PCDD/Fs, and the highest distribution of PCDFs is observed during the spring. The distribution of PCDD/F congeners measured at Lulin station is quite different from those measured at urban and industrial area in Taiwan with high PCDF distribution (>70%) especially in 2,3,7,8-TeCDF and 2,3,4,7,8-PeCDF (21, 28). Figure 5b shows that the increase (14%) of high-chlorinated PCDFs especially in 1,2,3,4,7,8-HxCDF and 1,2,3,4,6,7,8-HxCDF is measured during the Southeast Asia biomass burning event (March 18–21, 2008). Interestingly, the distribution of PCDFs decreased from 67% to 57% after the burning event (March 22–24, 2008). After the burning event, major contributors include 1,2,3,4,6,7,8-HxCDF (16%), OCDF (9%), and OCDD (31%) measured in Lulin station are quite similar with that measured during the regular periods. Gullett et al. (29) indicated that raw biomass is dominated by PCDDs, particularly OCDD, whereas the emissions are dominated by PCDFs, primarily PeCDFs. These observations suggest that the emissions are not simply a result of target volatilization and dechlorination but represent in situ formation. Hence, the measurements conducted in this study demonstrate that the increase in PCDF compounds could be attributed the long-range transport of the Southeast Asia biomass burning event.

3.3. Particle-Bound PCDD/Fs in Suspended Particle Measured at Lulin Station during Different Sampling Periods. In general, the biomass burning activities has a strong seasonal variation. In Southeast Asia, there are often agriculture cleanup activities during springtime, producing large biomass burning events (30). Previous studies also showed that spring is the major biomass burning season in Southeast Asia, which could release tremendous amount of air pollutants (31, 32). Figure 1 shows that the air mass passed over Southeast Asia before the Lulin station during the Southeast biomass burning event. Lee et al. (33) conducted the intensive aerosol sampling campaigns at Lulin station during 2003–2008. The results demonstrated that the enhancement of potassium ion in aerosols measured at Lulin station during the Southeast Asia biomass burning event (March 18–19, 2008) in contrast to nonbiomass burning period was at 341%. In general, potassium is an important plant nutrient. It is highly mobile and is absorbed into woody material through the root system and subsequently transported to all areas of the growing tree (34). A previous study (35) also concluded that high potassium emissions were a signature of biomass burning. Apparently, aerosols generated by long-range transport of biomass burning products caused wide variations in pollutant levels.

Figure 6 shows that the quantities of PCDD/Fs adsorbed onto suspended particles ranged from 22.2–65.3 pg I-TEQ/g-TSP during the regular sampling periods. Previous study (36) indicated that the PCDD/F contents of suspended particle observed in rural and urban areas of Germany were 320 and 630 pg I-TEQ/g-TSP, respectively. Those measurements were significantly higher than that measured during the regular sampling periods in this study. In addition, the PCDD/F content of suspended particle fraction measured during the winter season is higher than during other periods. Several mechanisms could be responsible for this seasonal pattern in air pollutant distribution observed in Lulin station, such lower boundary layer height and lower oxidation reaction rates in winter, higher atmospheric oxidant con-

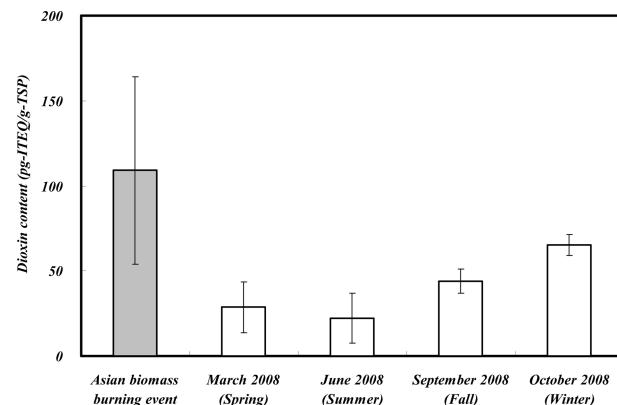


FIGURE 6. Comparison of particle-bound PCDD/Fs in total suspended particle (TSP) at Lulin station during different sampling periods.

centration in summer, and seasonal changes in meteorological conditions. Additionally, the vapor pressure of organic compounds increases with temperature, resulting in a lower fraction of PCDD/F congeners being adsorbed onto particles. During the winter season, temperatures at Lulin station were generally below 10 °C. Hence, the ambient air temperature affects the percentage of solid-phase dioxins.

The results of the current study demonstrated that significant PCDD/F mass (28.7 to 109 pg I-TEQ/g-TSP) was adsorbed onto suspended particles measured at Lulin station during the Southeast Asia biomass burning event. The large amounts of particulate matter generated by the Southeast Asia biomass burning accompanied an increase in the solid-phase PCDD/F concentrations in the atmosphere measured at Lulin station. PCDD/F congeners with lower vapor pressures are more likely to exist in the solid phase, and Southeast Asia biomass burning generated a significant mass of coarse and fine particles that were able to adsorb and transport solid-phase PCDD/Fs. We therefore conclude that the significant increase in solid-phase PCDD/Fs measured at high-altitude station in central Taiwan was attributable to the long-range transport of the products of the Southeast Asia biomass burning event.

Acknowledgments

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