Impact of the COVID-19 Outbreak on the Long-range Transport of Particulate PAHs in East Asia

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ABSTRACT

Particulate polycyclic aromatic hydrocarbons (PAHs) were continuously observed at Kanazawa University Wajima Air Monitoring Station (KUWAMS), a background site located in western Japan and downwind of mainland China that is constantly under the influence of polluted air masses originating from China. The observations showed that the concentration of particulate PAHs at KUWAMS sharply dropped to the lowest level (62 pg m^{-3}) of the cold season in the last 5 years after the implementation of COVID-19 control measures in China. Despite the occasional dilution with clean air, the decrease in PAHs at KUWAMS reflected the reduction in emission intensity in China. Moreover, the PAH concentrations at KUWAMS in February, March, and April of 2020 compared with the same period in the previous year decreased by 52.6%, 36.6%, and 36.7%, respectively. These changes were consistent with the decline in SO_2 and NO_2 in northern China, which intensified during the control of COVID-19 and was slightly moderated after the resumption of work. In addition, there was little change in the composition of PAHs at KUWAMS before and during the COVID-19 outbreak and compared with previous years, which suggests a stable source composition. These findings emphasize the importance of decreased emission intensity in China for reducing long-range transport of PAHs and pollution levels in downwind areas.

Keywords: Polycyclic aromatic hydrocarbons; Air pollution; Background site; KUWAMS.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are an important component of atmospheric particulates that influence the hormonal impact of particulates and their carcinogenic and mutagenic potential (Durant *et al.*, 1996; Zhang *et al.*, 2016). PAHs primarily originate from the incomplete combustion of organic materials, such as petroleum, coal, and biomass, as well as the evaporation and escape of PAHs in raw materials and fuel (Lee *et al.*, 1995; Yang *et al.*, 1998; Lee *et al.*, 2004; Chang *et al.*, 2013). After entering the atmosphere, PAHs undergo gas-particle partitioning, transportation, conversion, and dry and wet deposition into the local environment of the receptor domain (Cheruiyot *et al.*, 2015). Compared with low-molecular-weight PAHs, high-molecular-weight PAHs that with higher toxic potential tend to bind particles, especially those with fine size (Sheu *et al.*, 1996; Lee *et al.*, 2002;

Lin *et al.*, 2008). Airborne particles carrying PAHs can participate in the short- and long-range transport of air masses (Zhang *et al.*, 2020b). Although the physical and chemical behavior of PAHs during transport may lead to the loss of PAHs in particles, the enhanced health risks caused by PAHs transported to the recipient site cannot be ignored (Lee *et al.*, 1995; Sheu *et al.*, 1997; Zhang *et al.*, 2020e).

Globally, China is a major consumer of energy. In 2018, China accounted for 24% of global energy consumption of which coal and oil accounted for 58% and 18.9%, respectively (bp p.l.c., 2019; NBSC, 2019). As a result, the concentration of atmospheric PAHs in China is much higher than that of some Asian, European and American countries (Sanderson *et al.*, 2004; Delhomme and Millet, 2012; Shen *et al.*, 2013; Tang *et al.*, 2017; Zhang *et al.*, 2019a; Hayakawa *et al.*, 2020), although strict air pollution control measures instituted by China have effectively improved air quality in recent years (Zheng *et al.*, 2018; Zhang *et al.*, 2019c).

In particular, in northern China, central heating routinely lasts up to five months per year (from November to March of the following year), and the span of unorganized coal heating lasts even longer. These activities not only increase the load of atmospheric PAHs in China but also worsen air

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pollution in downwind countries and regions (Pan *et al.*, 2015; Inomata *et al.*, 2017; Li *et al.*, 2018). The observations over 15 years at Kanazawa University Wajima Air Monitoring Station (KUWAMS), a remote background site in Japan, provide hard evidence of the transport of pollutants (Tang *et al.*, 2005; Yang *et al.*, 2007; Tang *et al.*, 2014, 2015; Zhang *et al.*, 2020e). Driven by the East Asian monsoon system, polluted air masses in northern China can be transported to Japan with Northwest winds from November to May of the following year (the cold season); between June and October (the warm season), the airflow is driven by southerly winds from the North Pacific (Tang *et al.*, 2015). Consequently, the concentration of particulate PAHs observed at KUWAMS shows regular seasonal changes (Fig. 1) (Tang *et al.*, 2015; Zhang *et al.*, 2020e), with increased PAHs in the cold season and decreased PAHs in the warm season. In addition, the mitigation of air pollution in response to control measures taken by China, for example, the interim control measures for the 2008 Beijing Olympic Games (BMEPB, 2008), the Air Pollution Prevention and Control Action Plan implemented from 2013 to 2017 (State Council, PRC, 2013), and the clean energy heating project launched in northern China since 2017 (NDRC, 2017), has been reflected in the long-term observations at KUWAMS (Tang *et al.*, 2014, 2015; Zhang *et al.*, 2020e). Although KUWAMS is sometimes affected by polluted air masses in Japan, the level of atmospheric PAHs in Japanese cities is two orders of magnitude lower than that in China (Morisaki *et al.*, 2016). Moreover, the primary contributor to PAHs in Japan is motor vehicles (Hayakawa *et al.*, 2018), while the long-term observations at KUWAMS suggest that the source of PAHs tallies with the mixed emission sources in China. Therefore, KUWAMS is clearly a sensitive receptor site for changes in air pollution on mainland China.

At the beginning of 2020, COVID-19 broke out in China and rapidly spread throughout the country. To limit the spread of the COVID-19 virus, Wuhan and other cities in Hubei Province announced lockdowns on January 23 and 24, 2020, respectively. Other provinces in China had started

their emergency response systems by January 29. As a result, industrial, traffic, and social activities were largely suspended in China, effectively reducing the intensity of pollution emissions and air pollution levels (Wang *et al.*, 2020; Xu *et al.*, 2020; Zhang *et al.*, 2020d). According to the data released by the Ministry of Ecology and Environment of the People's Republic of China (MEE, 2020a), PM₁₀, PM_{2.5}, SO₂, and NO₂ in 337 cities in China decreased by 30.0%, 27.3%, 23.1% and 28.0%, respectively, compared with the values for February of the previous year, and the decline was more significant than the other winter months. Therefore, the reduced pollution emissions in China during COVID-19 are expected to reduce the pollutants involved in long-range transportation and to mitigate air pollution in East Asia.

The purpose of this study was to document the impact of measures implemented in China to control the COVID-19 virus on the long-range transport of PAHs in East Asia. In this work, particulate PAHs were measured at KUWAMS and the partition of PAHs in the gas phase was estimated by models. Changes in particulate PAHs at KUWAMS during the 2020 cold season and during COVID-19 were investigated by comparison with the previous 15 years of observations. In addition, the driving factors were evaluated by estimating the airflow trajectory to KUWAMS during COVID-19 control measures and by analyzing changes in the composition of PAHs. The results of this study contribute to a better understanding of the transport and distribution of PAHs in the atmosphere of East Asia and provide an important basis for the improvement of air quality.

METHODS

Air Particulate Sampling

KUWAMS (Nisifutamata-machi, Wajima City, Ishikawa Prefecture, Japan; 37.4°N, 136.9°E; 60 m above sea level) is located on the Noto Peninsula in western Japan (Fig. 2), 2.1 km from the coastline of the Sea of Japan. A detailed description of KUWAMS is provided elsewhere (Yang *et al.*, 2007; Tang *et al.*, 2014). The collection of total suspended

Fig. 1. Concentration profile of particulate polycyclic aromatic hydrocarbons (PAHs) at Kanazawa University Wajima Air Monitoring Station (KUWAMS) from Sep. 2004 to Aug. 2019 (Tang *et al.*, 2015; Zhang *et al.*, 2020e), showing an approximately sinusoidal trend. FR: fluoranthene; Pyr: pyrene; BaA: benz[*a*]anthracene; Chr: chrysene; BbF: benzo[*b*]fluoranthene; BkF: benzo[*k*]fluoranthene; BaP: benzo[*a*]pyrene; BgPe: benzo[*ghi*]perylene; IDP: indeno[1,2,3-*cd*]pyrene.

Fig. 2. Location of KUWAMS (Tang *et al.*, 2014).

particulates (TSP) was performed using a high-volume air sampler (AH-600, Shibata Sci. Tech. Ltd., Saitama, Japan) on a quartz fiber filter (8 in. $\times$ 10 in., 2500QAT-UP, Pallflex Products, Putnam, CT, USA) at a flow rate of 700 L min⁻¹. The filter was replaced once a week from November 2, 2019, to April 25, 2020. Based on the prevailing period of the East Asian winter monsoon, this sampling period was defined as the cold season, which extended from November to May in the previous observations (Tang *et al.*, 2015; Zhang *et al.*, 2020e).

Before and after sampling, the filters were held in a desiccator with a constant temperature ($21.5 \pm 1.5^\circ\text{C}$) and humidity ($50 \pm 5\%$) and were protected from light for 24 h. These filters were then weighed. The difference in the weight of each filter before and after sampling was used to calculate the TSP concentration. After gravimetric analysis, the sampled filters were stored at -20°C until analysis. During sampling, the blank and sample filters were pretreated in the same way and delivered to KUWAMS to check the background variance. It was documented that no contamination occurred during storage, weighing, and transportation.

In addition, local meteorological conditions were automatically monitored by the weather Transmitter WXT530 during the sampling period. Meteorological parameters included temperature, relative humidity (RH), pressure, wind speed, and rainfall. The details of meteorological monitoring are provided elsewhere (Zhang *et al.*, 2020e).

PAH Analysis

Nine PAHs, fluoranthene (FR), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), benzo[ghi]perylene (BgPe), and indeno[1,2,3-cd]pyrene (IDP), were analyzed in each TSP sample. The U.S. EPA 610 PAH mix, including the nine target PAHs, was purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Two internal standards (Pyr-*d*₁₀ and BaP-*d*₁₂) were purchased from Wako Pure Chemicals (Osaka, Japan). All additional reagents were of analytical reagent grade.

The methods of pretreatment, analysis and quality control are consistent with those described in a previous study (Tang *et al.*, 2005). Each filter was cut into small pieces and placed

in a conical flask. After adding internal standards (Pyr-*d*₁₀, 0.14 mM and BaP-*d*₁₂, 0.11 mM), the compounds were extracted ultrasonically twice with benzene/ethanol (3:1, v/v). The extract was washed successively with sodium hydroxide and sulfuric acid solutions, and twice with ultrapure water. After that, 100 μL of dimethyl sulfoxide (DMSO) was added to the solution, which was then concentrated with rotation evaporator and dissolved to 1.0 mL in acetonitrile. After filtering the organic solution with an HLC-DISK membrane (pore size 0.45 μm , Kanto Chemical Co., Tokyo, Japan), an aliquot (20 μL) of the solution was injected into a high-performance liquid chromatography (HPLC) system (LC-20A series, Shimadzu Inc., Kyoto, Japan) for quantitative determination.

The nine PAHs were determined by an HPLC coupled with fluorescence detection. A reversed-phase column (Inertsil ODS-P, 4.6 i.d. $\times$ 250 mm, GL Sciences Inc., Tokyo, Japan) was employed for separating the PAH compounds. The mobile phase was an acetonitrile/water gradient with the flow rate at 1 mL min⁻¹. The time program of the fluorescence detector was set to detect at the optimum excitation and emission wavelengths for each PAH, which is consistent with the condition in the previous study (Tang *et al.*, 2005). Internal standards were used to calibrate the recovery and quantification of each PAH. The recoveries ranged from 87 to 104%. The limit of determination and quantification of each PAH was FR (70 and 240 pg mL⁻³), Pyr (6.3 and 21 pg mL⁻³), BaA (6.8 and 23 pg mL⁻³), Chr (19 and 63 pg mL⁻³), BbF (61 and 200 pg mL⁻³), BkF (15 and 49 pg mL⁻³), BaP (11 and 37 pg mL⁻³), BgPe (100 and 330 pg mL⁻³), and IDP (460 and 1550 pg mL⁻³), respectively.

Data Analysis

Statistics were performed using SPSS version 25.0 (IBM Co., Armonk, NY, US). Differences in the concentrations of TSP and PAHs between this study and previous observations were evaluated via a *t*-test. A *p*-value of less than 0.05 indicated that the results were statistically significant.

The backward trajectory model (HYSPLIT-WEB, Internet-based, the NOAA Air Resources Laboratory, https://www.ready.noaa.gov/HYSPLIT_traj.php) is widely used in environmental analysis to track air masses (Yang *et al.*, 2019;

Zhang *et al.*, 2019b). In this work, the trajectories of airflow arriving at KUWAMS during the sampling periods (between January 18 and 25, between January 25 and February 1, and between February 1 and 8, 2020) were estimated by using the HYSPLIT model and the GDAS one-degree archive. The start time was set to 0:00 UTC on January 18, January 25, and February 1, 2020, which corresponds to the local time (9:00 a.m.) when the filter was replaced. A new trajectory was calculated every 24 h. Based on the previous reports (Tang *et al.*, 2015; Zhang *et al.*, 2020e), the backtracking time was set at 72 h, and the initial height was set at 1000 m above ground level.

Gas-particle Partitioning

The concentration of PAHs in the gas phase was estimated by the gas-particle partition model. Firstly, the partition coefficient K_p (unit: $\text{m}^3 \mu\text{g}^{-1}$) for each PAH between TSP and gas phase was calculated by Eq. (1), which is based on the adsorption of PAHs by particles (Pankow, 1987) and the absorption of PAHs by organic matter in the particle (Pankow, 1994, 1998). This model is widely used for evaluating the gas-particle partitioning of semi-volatile organic compounds in the atmosphere (Kishida *et al.*, 2011; Huang *et al.*, 2014; Liu *et al.*, 2014).

$$\lg K_p = \lg K_{OA} + \lg f_{om} - 11.91 \quad (1)$$

where K_{OA} is the octanol/air partition coefficient of each PAH, $(\text{g mL}^{-1})_{\text{octanol}} (\text{g mL}^{-1})^{-1}_{\text{air}}$; f_{om} is the mass fraction of organic matter in TSP. The K_{OA} used in this study refers to the report of Odabasi *et al.* (2006); the f_{om} was estimated to be 0.1 based on previous measurements at KUWAMS (Zhang *et al.*, 2020e). The concentration of PAHs in the gas phase was then calculated by Eq. (2) (Yamazaki *et al.*, 1982).

$$K_p = \frac{C_p / C_{TSP}}{C_g} \quad (2)$$

where C_p and C_g are the concentration of each PAH in TSP and gas phase, respectively, pg m^{-3} ; and C_{TSP} is the

concentration of TSP, $\mu\text{g m}^{-3}$.

RESULTS AND DISCUSSION

Overview of TSP and PAHs during the Cold Season

The concentration of TSP collected at KUWAMS during the sampling period is shown in Table 1. For the purpose of comparison, Table 1 summarizes the data from previous long-term observations at KUWAMS, including those during the cold season from 2004 to 2014 (Campaign 1; Tang *et al.*, 2015) and from 2014 to 2019 (Campaign 2; Zhang *et al.*, 2020e). The average TSP concentration $\pm$ standard deviation at KUWAMS during the present sampling period was $9.7 \pm 2.8 \mu\text{g m}^{-3}$, which was significantly lower than that during Campaign 1 ($21 \pm 14 \mu\text{g m}^{-3}$; $p < 0.001$) and Campaign 2 ($20 \pm 11 \mu\text{g m}^{-3}$; $p < 0.001$) (Tang *et al.*, 2015; Zhang *et al.*, 2020e).

Table 2 presents the gas-particle partitioning of PAHs in the atmosphere at KUWAMS during the sampling period calculated by models. The sum of the concentration of PAHs in the TSP and gas phase at KUWAMS was estimated at $25100 \pm 11000 \text{ pg m}^{-3}$ with around 1.1% of PAHs bound with TSP. These results indicate that the concentration of PAHs transported from mainland China during the sampling period was much higher than the concentration of PAHs in the TSP detected in this study. In addition, 4-ring PAHs (FR, Pyr, BaA, and Chr) mostly existed in the gas phase, while 5- and 6-ring PAHs were mostly bound with TSP. This gas-particle partition pattern is consistent with the observations of many studies (Ohura *et al.*, 2004; Kishida *et al.*, 2011; Huang *et al.*, 2014; Liu *et al.*, 2014). However, because of the influence of environmental conditions and the nature of particles, using the documented parameters to estimate K_p can lead to an uncertainty of up to 95% (Yamazaki *et al.*, 1982; Araki *et al.*, 2009; Salthammer and Schripp, 2015). Considering the high uncertainty of the calculated PAH concentrations in the gas phase, the following sections focus on particulate PAHs for further discussion. On the other hand, compared with model calculations, the measured results have higher reliability (Salthammer and Schripp, 2015). Therefore, the monitoring of gas-phase PAHs will be considered in our future research to characterize PAHs more comprehensively.

Table 1. Mean concentration $\pm$ standard variation of total suspended particulates (TSP) and PAHs at KUWAMS in the cold season from 2004 to 2020.

Sampling period	Campaign 1 ^a 2004–2014	Campaign 2 ^b 2014–2019	This study ^c 2019–2020
TSP ($\mu\text{g m}^{-3}$)	21 ± 14	20 ± 11	9.7 ± 2.8
ΣPAHs ^d (pg m^{-3})	670 ± 360	550 ± 300	270 ± 160
FR	170 ± 110	150 ± 92	71 ± 40
Pyr	110 ± 71	94 ± 57	49 ± 27
BaA	24 ± 15	21 ± 13	11 ± 7
Chr	62 ± 36	49 ± 25	24 ± 14
BbF	85 ± 54	80 ± 43	37 ± 23
BkF	31 ± 19	29 ± 15	13 ± 8
BaP	39 ± 28	37 ± 19	17 ± 10
BgPe	48 ± 30	41 ± 23	23 ± 13
IDP	49 ± 29	45 ± 24	23 ± 13

^a Tang *et al.*, 2015; The number of samples $n = 280$. ^b Zhang *et al.*, 2020e; $n = 94$. ^c $n = 27$. ^d Total PAHs.

Table 2. Gas-particle partitioning of PAHs at KUWAMS during the sampling period calculated by models.

PAH	C_g^a	$C_g + C_p^b$	$C_p/(C_g + C_p)$
ΣPAHs	24900 ± 11000	25100 ± 11000	1.1% ± 0.29%
FR	15400 ± 6800	15500 ± 6900	0.45% ± 0.12%
Pyr	9200 ± 3900	9300 ± 3900	0.53% ± 0.14%
BaA	49 ± 25	60 ± 31	18% ± 4.3%
Chr	100 ± 44	120 ± 57	19% ± 4.5%
BbF	11 ± 5.1	48 ± 28	76% ± 5.4%
BkF	3.6 ± 1.7	17 ± 9.6	77% ± 5.1%
BaP	2.8 ± 1.3	19 ± 11	85% ± 3.8%
BgPe	0.32 ± 0.14	23 ± 13	98% ± 0.45%
IDP	0.42 ± 0.19	23 ± 13	98% ± 0.59%

^a The concentration of each PAH in the gas phase, pg m^{-3} . ^b The sum of the concentration of each PAH in the TSP and gas phase, pg m^{-3} .

As shown in Table 1, the average level of PAHs in the TSP during the present sampling period ($270 \pm 160 \text{ pg m}^{-3}$) was significantly lower than that during Campaign 1 ($670 \pm 360 \text{ pg m}^{-3}$; $p < 0.001$) and Campaign 2 ($550 \pm 300 \text{ pg m}^{-3}$; $p < 0.001$) (Tang *et al.*, 2015; Zhang *et al.*, 2020e), which was similar to the variation of TSP. As mentioned above, the Chinese government has been committed to controlling air pollution in recent years and has effectively improved air quality nationwide (Zheng *et al.*, 2018). Therefore, the downward trend of TSP and PAHs at KUWAMS from Campaign 1 to Campaign 2 has been attributed to the reduction of pollutants transported from the Chinese mainland (Zhang *et al.*, 2020e). This is also considered to be an important reason for the lower concentrations of TSP and PAHs during the sampling period in this study.

After the input of air pollutants into the atmosphere, the development of air pollution will be influenced by local meteorological conditions. Unfavorable meteorological conditions such as light air, calm winds, and high RH can lead to the accumulation of pollutants and heavy pollution. In contrast, an improvement in meteorological conditions will facilitate the diffusion of local pollutants (Zhang *et al.*, 2015; Zhong *et al.*, 2018). Based on 15 years of observations at KUWAMS during Campaign 1 and Campaign 2, the particulate PAHs were significantly negatively correlated with temperature during the cold season, but no significant correlation was found between PAHs and wind speed or RH (Tang *et al.*, 2015; Zhang *et al.*, 2020e). Due to the semivolatility of PAHs, the increase in temperature promotes the transition of particulate PAHs into the gas phase, whereas low temperature is conducive to the accumulation of PAHs into particles (Araki *et al.*, 2009). Based on the comparison in Table 3, the monthly meteorological conditions at KUWAMS during the present sampling period were comparable to those during Campaign 2 (Zhang *et al.*, 2020e). This finding suggests that the effects of local meteorological conditions on TSP and PAHs during the two sampling periods were similar. Moreover, long-range transport was the primary factor affecting the variation in TSP and PAHs at KUWAMS. On the other hand, since temperature is an important factor affecting the gas-particle partitioning of PAHs, the relatively stable monthly temperature at KUWAMS (Table 3) indicates that the gas-particle partition pattern of

PAHs changed a little in the past long-term observation. These results emphasize the comparability between the concentration of particulate PAHs during the present sampling period and the previous campaign.

PAHs are typically released from pollution sources as a mixture, and the combustion temperature and fuel type play an important role in the composition of the resulting PAHs (Tang *et al.*, 2005; Ravindra *et al.*, 2008; Phoungthong *et al.*, 2017). Thus, the molar ratio of characteristic PAHs is widely used to identify their major sources (Tang *et al.*, 2017; Xing *et al.*, 2020). Table 4 compares the characteristic ratios of PAHs at KUWAMS in this study with previous observations. The ratios of the five PAH pairs during the present sampling period were consistent with those during Campaign 1 and Campaign 2 (Tang *et al.*, 2015; Zhang *et al.*, 2020e). Due to the lack of emission sources near KUWAMS and the relatively stable meteorological conditions, it is inferred that the composition of the source of PAHs has not changed substantially. The ratios of $[\text{FR}]/([\text{Pyr}] + [\text{FR}])$, $[\text{BaA}]/([\text{Chr}] + [\text{BaA}])$ and $[\text{IDP}]/([\text{BgPe}] + [\text{IDP}])$ (Table 4) were closer to the range indicating vehicle emissions (Sicre *et al.*, 1987; Rogge *et al.*, 1993; Tang *et al.*, 2005), whereas the ratios of $[\text{BbF}]/([\text{BbF}] + [\text{BkF}])$ and $[\text{BaP}]/([\text{BaP}] + [\text{BgPe}])$ indicated an origin of coal combustion (Daisey *et al.*, 1979; Masclet *et al.*, 1987; Huang *et al.*, 2014). These results are consistent with previous observations that particulate PAHs at KUWAMS are jointly affected by vehicle exhaust and coal combustion (Tang *et al.*, 2015; Zhang *et al.*, 2020e). Although a large fraction of 4-ring PAHs was distributed in the gas phase (Table 2), the characteristic ratios obtained after considering the gas-phase concentration were consistent with those of particulate PAHs (Table 4). This result emphasizes the stable source of atmospheric PAHs at KUWAMS.

Changes in TSP and PAHs during COVID-19

Fig. 3 shows the weekly variation in PAHs and TSP at KUWAMS during the cold season over the last 5 years. The concentration of TSP (represented by hollow squares) showed a similar trend in each cold season from 2015 to 2019 (Figs. 3(a)–3(d)). The TSP concentration was approximately $20 \mu\text{g m}^{-3}$ between November and February of the following year. The TSP then rose to a relatively high level until the end of April, which can be attributed to the frequent invasion

Table 3. Comparison of monthly meteorological conditions (mean $\pm$ standard variation) at KUWAMS in this study and during Campaign 2 (Zhang *et al.*, 2020e).

Month		T ^a (°C)	RH ^b (%)	Press (hPa)	WS ^c (m s ⁻¹)	Rain ^d (mm)
Nov.	This study	9.78 $\pm$ 2.40	76.3 $\pm$ 6.63	1010 $\pm$ 5.90	0.98 $\pm$ 0.28	0.25 $\pm$ 0.42
	Campaign 2	9.77 $\pm$ 2.80	80.6 $\pm$ 5.00	1010 $\pm$ 3.81	0.95 $\pm$ 0.20	0.27 $\pm$ 0.38
Dec.	This study	6.74 $\pm$ 2.26	76.4 $\pm$ 6.91	1010 $\pm$ 5.22	1.04 $\pm$ 0.23	0.19 $\pm$ 0.26
	Campaign 2	4.78 $\pm$ 2.79	80.1 $\pm$ 6.85	1010 $\pm$ 5.49	1.06 $\pm$ 0.26	0.31 $\pm$ 0.35
Jan.	This study	5.12 $\pm$ 1.95	77.9 $\pm$ 6.82	1010 $\pm$ 7.52	1.01 $\pm$ 0.30	0.27 $\pm$ 0.26
	Campaign 2	3.44 $\pm$ 2.14	75.1 $\pm$ 7.66	1010 $\pm$ 5.19	1.09 $\pm$ 0.29	0.17 $\pm$ 0.30
Feb.	This study	5.11 $\pm$ 2.77	76.3 $\pm$ 5.51	1010 $\pm$ 6.12	1.06 $\pm$ 0.26	0.22 $\pm$ 0.33
	Campaign 2	4.96 $\pm$ 2.14	73.7 $\pm$ 7.56	1010 $\pm$ 5.51	1.08 $\pm$ 0.25	0.15 $\pm$ 0.27
Mar.	This study	7.13 $\pm$ 2.60	69.7 $\pm$ 6.68	1010 $\pm$ 5.87	1.12 $\pm$ 0.24	0.16 $\pm$ 0.33
	Campaign 2	6.18 $\pm$ 2.66	74.7 $\pm$ 6.95	1010 $\pm$ 5.53	0.99 $\pm$ 0.20	0.23 $\pm$ 0.48
Apr.	This study	9.40 $\pm$ 2.09	69.1 $\pm$ 7.66	1010 $\pm$ 5.09	1.10 $\pm$ 0.26	0.14 $\pm$ 0.24
	Campaign 2	11.8 $\pm$ 3.36	74.8 $\pm$ 8.47	1010 $\pm$ 5.37	1.01 $\pm$ 0.24	0.17 $\pm$ 0.34

^a Temperature. ^b Relative Humidity. ^c Wind Speed. ^d Monthly rainfall.

Table 4. Characteristic ratios of PAH pairs at KUWAMS in the cold season from 2004 to 2020 and the range indicating different sources.

Ratio	[FR]/([Pyr] + [FR])	[BaA]/([Chr] + [BaA])	[BbF]/([BbF] + [BkF])	[BaP]/([BaP] + [BgPe])	[IDP]/([BgPe] + [IDP])
Campaign 1	0.59 ^a	0.28	0.74	0.44	0.50
Campaign 2	0.61 ^b	0.29	0.73	0.45	0.52
This study	0.58 $\pm$ 0.02 ^c	0.32 $\pm$ 0.02	0.74 $\pm$ 0.01	0.42 $\pm$ 0.02	0.50 $\pm$ 0.02
This study	0.62 $\pm$ 0.02 ^d	0.32 $\pm$ 0.02	0.74 $\pm$ 0.01	0.46 $\pm$ 0.02	0.50 $\pm$ 0.02
Vehicle emission	0.6–0.7 ^e	0.36 ^f	0.52–0.60 ^g	0.23–0.28 ^h	0.35–0.7 ⁱ
Coal combustion	0.36–0.53 ^{fj}	0.5 ^f	0.78–0.95 ^{h,j}	0.47–0.86 ^k	0.14–0.33 ^{fj}

^a Median ($n = 280$). ^b Mean ($n = 94$). ^c Mean $\pm$ standard variation ($n = 27$). ^d The ratio was calculated based on the sum of the concentration of each PAH in the TSP and gas phase. ^e Sicre *et al.*, 1987. ^f Tang *et al.*, 2005. ^g Dickhut *et al.*, 2000.

^h Masclat *et al.*, 1987. ⁱ Rogge *et al.*, 1993. ^j Huang *et al.*, 2014. ^k Daisey *et al.*, 1979.

of Asian dust (Yang *et al.*, 2007; Zhang *et al.*, 2020c). In contrast to the trend in TSP, the concentration of PAHs (represented by solid circles) between November and February was higher than that between March and April (Figs. 3(a)–3(d)). This trend was essentially coincident with the central heating period in northern China as mentioned previously. After the central heating period, the levels of PAHs at KUWAMS decreased, and the occasional increase in PAHs might be associated with the presence of unorganized coal burning in rural areas (MEE, 2020b).

As shown in Fig. 3(e), the local TSP concentration has remained below 16 $\mu\text{g m}^{-3}$ from November 2019 to April 2020. This is related to the absence of Asian dust at KUWAMS during this period (JMA, 2020). Impressively, due to the implementation of strategies against COVID-19 in China from January 23, 2020, the concentration of PAHs stabilized from late January to April of 2020 (Fig. 3(e)) at a lower level than that in the previous years (Figs. 3(a)–3(d)). To better understand this trend, the year to year changes in the concentrations of TSP and PAHs at KUWAMS were calculated (Table 5) and compared with those of air pollutants in the representative region of northern China, the Beijing-Tianjin-Hebei Metropolitan Region (BTH Metropolis) (MEE, 2020a). In comparison with the previous years, there was a relative increase in PM_{2.5} in December and January. But in subsequent months, the PM₁₀, PM_{2.5}, SO₂, and NO₂

in the BTH Metropolis showed decreases in each month with the largest decline in February, particularly SO₂ and NO₂, which are primary pollutants indicative of coal combustion and vehicle exhaust, respectively. These substantial reductions reflect the widespread suspension of industrial and traffic activities in China since the implementation of control measures related to the COVID-19 pandemic (Wang *et al.*, 2020). As the COVID-19 pandemic has improved in China, the provinces outside Hubei have released strategies for a gradual return to work after February 10 (Zhang *et al.*, 2020d). To avoid a recurrence of the pandemic, industrial production and traffic flow have been resumed at a slower pace. As a result, the decrease in air pollutants in the BTH Metropolis was moderated in March and April (MEE, 2020b); as shown in Table 5, the concentration of PAHs at KUWAMS decreased by 52.6%, 36.6%, and 36.7% in a year to year comparison in February, March, and April, respectively. This trend corresponded to the changes in SO₂ and NO₂ in the BTH Metropolis, which highlights the direct effect of the reduction in industrial and traffic activities in northern China on the PAH level at KUWAMS.

It is worth noting that during the Chinese Spring Festival (a week-long holiday), the intensity of industrial emissions weakened (Tang *et al.*, 2020). Although the sampling week at KUWAMS did not perfectly correspond to the Chinese Spring Festival (indicated by a gray rectangle in Fig. 3), the

Fig. 3. Weekly variation of Σ PAHs and TSP at KUWAMS in the cold season from 2015 to 2020. The solid circle represents the concentration of Σ PAHs. The hollow square represents the concentration of TSP. The gray rectangle represents the week of the Chinese Spring Festival. The data for 2015 to 2019 were collected from Zhang *et al.* (2020e).

Table 5. Year-on-year comparison of air pollutants in KUWAMS and Beijing-Tianjin-Hebei Metropolitan Region (BTH Metropolis) in the cold season from 2019 to 2020.

Site / region	KUWAMS		BTH Metropolis ^a			
Pollutants	Σ PAHs	TSP	PM ₁₀	PM _{2.5}	SO ₂	NO ₂
Nov. 2019	–22.5% ^b	–28.0%	–10.4%	–25.9%	–20.0%	–7.1%
Dec. 2019	–1.6%	–15.5%	–16.4%	1.2%	–36.0%	–5.5%
Jan. 2020	–69.9%	–50.6%	–10.2%	10.2%	–36.7%	–19.4%
Feb. 2020	–52.6%	–27.7%	–45.7%	–42.6%	–40.0%	–41.5%
Mar. 2020	–36.6%	–59.4%	–24.1%	–16.7%	–25.0%	–17.5%
Apr. 2020	–36.7%	–35.2%	–17.8%	–25.0%	–7.1%	–5.7%

^a Monthly report for national air quality (MEE, 2020a). ^b The year-on-year change was calculated by $100\% \times (\text{present monthly mean} - \text{previous monthly mean}) / \text{previous monthly mean}$; for example, present: Nov. 2020, previous: Nov. 2019.

concentration of PAHs showed a decrease during the corresponding periods such as the samplings from February 13 to 20 in 2016, from January 28 to February 4 in 2017, and

from February 9 to 16 in 2019. In 2020, the Chinese Spring Festival coincided with the period of COVID-19 control, leading to the concentration of PAHs at KUWAMS between

January 25 and February 1 (Fig. 3(e)) falling to the lowest level (62 pg m^{-3}) of the cold seasons from 2015 to 2020. This could explain the dramatic decline in the relative PAHs and TSP at KUWAMS in January 2020 (Table 5).

Characteristics of PAHs from January 25 to February 1

The week of overlap between the 2020 Chinese Spring Festival and the COVID-19 control period was identified as a period of particular interest and corresponded to the sampling week between January 25 and February 1 at KUWAMS. The trajectory of the airflow arriving at KUWAMS in the week before (W 1), during (W 2) and after (W 3) this period was estimated using the HYSPLIT model. As shown in Fig. 4, the airflow arriving at KUWAMS originated from or passed through northern China in W 1, when industrial and traffic activities in China continued as usual. The Northwest wind facilitated the transport of polluted air masses from northern China to KUWAMS, resulting in a relatively high PAH level (620 pg m^{-3}) at KUWAMS. In W 2, the airflow carrying pollutants over northern China arrived at KUWAMS on January 25, 26, and 27, during which time the intensity of industrial and traffic pollution on the Chinese mainland declined (Wang *et al.*, 2020; Xu *et al.*, 2020). Moreover, the airflow arriving at KUWAMS between January 28 and February 1 originated from the Okhotsk Sea and the Philippine Sea on the North and South sides of Japan, respectively. Pollutants from other Japanese cities might have been transported to KUWAMS with the airflow in the lower atmosphere. However, the level of PAH pollution in Japan is much lower than that in China (Tang *et al.*, 2013; Hayakawa *et al.*, 2018; Zhang *et al.*, 2020a). Therefore, the sharp decrease in PAH concentration to 62 pg m^{-3} in W 2 was attributed to the reduced input of pollutants reaching KUWAMS and the dilution with cleaner air. In W 3, the airflow largely returned to the control of the East Asian monsoon, where industrial production and traffic flow in northern China had not yet to resume. Consequently, the concentration of PAHs (250 pg m^{-3}) at KUWAMS rebounded slightly but was still lower than the level in W 1.

Fig. 5 compares the composition of particulate PAHs in W 1, W 2, and W 3 in 2020 with the same period in previous years, i.e., the week before (W 1), during (W 2) and after (W 3)

the Chinese Spring Festival from 2016 to 2019. The data for 2018 were not available due to a sampler failure and were not included in the comparison. The composition of PAHs at KUWAMS remained stable around the Chinese Spring Festival from 2016 to 2020. In the sampling weeks in Fig. 5, the PAHs were dominated by 4-ring homologs (FR, Pyr, BaA and Chr), which accounted for 54.7–62.6% of the total, followed by 5-ring (BbF, BkF and BaP) and 6-ring homologs (BgPe and IDP), which accounted for 23.3–26.3% and 14.1–20.1%, respectively. This result suggests that the reduced emission intensity in China had little effect on the composition of PAHs at KUWAMS, which supports the conclusion in section 3.1 that the main sources of PAHs did not change substantially. These findings are consistent with the assessment of the effectiveness of pollution control policies adopted in China, i.e., that the strengthening of emission standards has contributed substantially to the improvement in air quality, whereas the effect of energy structure adjustments was nonsignificant (Zhang *et al.*, 2019c).

CONCLUSIONS

The atmospheric TSP was continuously collected at KUWAMS, a remote background site in western Japan, from November 2019 to April 2020, and nine PAHs in the TSP samples were analyzed. Due to its location in the East Asian monsoon zone, the air quality at KUWAMS is greatly affected by the transport of polluted air masses from northern China during the cold season. Compared with the previous 15 years of observations at KUWAMS, the average concentration of TSP and PAHs decreased significantly during this sampling period. Based on the concentration profile, the decrease in TSP and PAHs at KUWAMS before the COVID-19 outbreak was related to the implementation of stringent air pollution control measures in China. During the period of COVID-19 control, the widespread suspension of industrial and traffic activities in China resulted in a low level of TSP and PAHs at KUWAMS from late January to April. In particular, in the week overlapping with the Chinese Spring Festival (between January 25 and February 1), the PAH concentration at KUWAMS dropped to the lowest level of the cold seasons between 2015 and 2020. This can

Fig. 4. Back trajectories at 1000 m above KUWAMS within the week before (W 1), during (W 2) and after (W 3) the Chinese Spring Festival using HYSPLIT-WEB model provided by the Air Resources Laboratory, NOAA.

Fig. 5. Composition of PAHs at KUWAMS in W 1, W 2, and W 3 from 2016 to 2020. The data for 2016 to 2019 were collected from Zhang et al. (2020e). The data for 2018 were not available due to a sampler failure.

be attributed to the reduced intensity of emissions in China and the dilution with cleaner air. In addition, it was found that the composition of PAHs at KUWAMS remained stable because the emission sources did not change substantially. These results indicate that the characteristics of PAHs at KUWAMS directly reflect the air pollution control policies adopted in China. Moreover, the observations at KUWAMS during COVID-19 emphasize that a decline in anthropogenic emissions from the Chinese mainland would reduce the air pollutants involved in long-range transport and change the distribution of PAHs and other pollutants in the atmosphere over East Asia. Therefore, the Chinese government should continue its efforts to strictly control the emission of air pollutants, which will be beneficial to the improvement of air quality in China and in downwind areas.

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