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Report

Characteristics of Atmospheric Fine Particulate Matter ($PM_{2.5}$) Pollution in a Suburban Residential Area of Saitama (Japan)

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We measured the atmospheric concentrations of fine particulate matter ($PM_{2.5}$) and polycyclic aromatic hydrocarbons (PAHs) on every Tuesday from 2016 to 2019 to reveal the characteristics of $PM_{2.5}$ pollution in a suburban residential area of Saitama Prefecture, a commuter town of Tokyo, and obtained the following results. First, mean $PM_{2.5}$ concentration over four years was 15.3 µg/m³, which was slightly higher than the long-term environmental standard of 15 µg/m³. Second, extremely high $PM_{2.5}$ concentrations may have been caused by the arrival of the Yellow Sand; a high PAH concentration was also observed during this event. Third, in the area surrounding the fields, $PM_{2.5}$ as well as PAH concentrations were increased from autumn to winter owing to open burning of biomass. Finally, the $PM_{2.5}$ concentration in the suburban areas surrounding the fields was 70% of that in central Tokyo, and the PAH concentration in late winter was the same as that in central Tokyo. These findings suggest that $PM_{2.5}$ and PAH concentrations in the suburban area surrounding the fields were high despite the area's low population density because of $PM_{2.5}$ accumulation in the metropolitan area and $PM_{2.5}$ emission from biomass burning. To improve the living environment of suburban residential areas surrounding fields, it is necessary to restrict biomass burning.

Key words fine particulate matter, polycyclic aromatic hydrocarbon, suburban residential area, Saitama prefecture, open burning of biomass

INTRODUCTION

Saitama Prefecture is located north of the Tokyo Metropolitan and has a population of 7.3 million. Because 14% of the residents travel to Tokyo for work during the day, Saitama Prefecture is known as a commuter town of Tokyo.[1] Therefore, it is crucial to properly maintain the living environment of Saitama Prefecture. However, the atmosphere of Saitama Prefecture is severely polluted by photochemical oxidants and fine particulate matter ($PM_{2.5}$).[2] Since January 2013, when an extremely high concentration of $PM_{2.5}$ was observed in Beijing, China,[3] air pollution caused by $PM_{2.5}$ has become a growing concern.

$PM_{2.5}$ refers to particulate matter with diameters less than 2.5 µm suspended in the atmosphere and is produced by combustion, crushing, and chemical reactions in the atmosphere.[4] $PM_{2.5}$ comprises carcinogenic polycyclic aromatic hydrocarbons (PAHs) and various other harmful substances,[5] which can get deposited in the alveoli and cause various diseases, including lung cancer.[6] To develop appropriate measures for reducing air pollution caused by $PM_{2.5}$, it is necessary to determine the concentrations, dynamics, and sources of $PM_{2.5}$ in the atmosphere. In this study, we measured the $PM_{2.5}$ and PAH concentrations in a suburban residential area of Saitama Prefecture and evaluated the factors and sources contributing to fluctuations in their concentrations.

METHODS

$PM_{2.5}$ and particulate matter larger than 2.5 µm were collected on a PG-60 PTFE-coated composite filter (Toyo, Tokyo, Japan) and a Toyo GB-100R glass fiber filter, respectively, using a Sibata HV-500F high-volume air sampler equipped with a $PM_{2.5}$ particle size separator (Saitama, Japan).

Sampling was conducted in Ina sampling station (SS) located at Ina Town of Saitama Prefecture (35°99′06″N, 139°61′84″E; height, 60 m) and Ueno SS located at Ueno of Tokyo Metropolitan (35°71′38″N, 139°77′82″E; height, 30 m), as shown in Fig. 1 (left). Sampling at Ina SS was performed from January 2016 to December 2019, and simultaneous sampling at Ina SS and Ueno SS was conducted from February to March and August to September of 2017–2019. Sampling was started at 10:00 of every Tuesday. The flow rate and sampling time were set at 500 L/min and 24 h, respectively.

Filters before and after sampling were weighed at a temperature of 20–23°C and relative humidity of 30–40%. They were treated as follows. Each filter was placed in a glass tube, to which 2.5 mL of ethanol and 7.5 mL of toluene were added successively. This mixture was sonicated using a Branson 3510 ultrasonic cleaner (Yamato, Tokyo, Japan) for 15 min, and the extracted solution was evaporated to obtain a dry residue. This procedure was repeated three times. The res-

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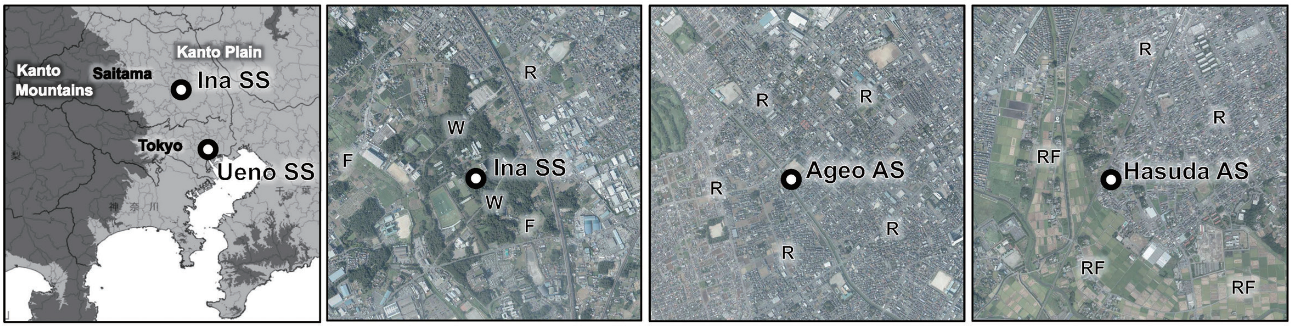

Fig. 1. Sampling Locations

Left, map showing the locations of Ina SS and Ueno SS. Light gray, altitude lower than 100 m; dark gray, altitude higher than 100 m. Second left, third left, and right, aerial photos around Ina SS, Ageo AS, and Hasuda AS, respectively. R, residence; RF, rice field; F, fields other than rice fields; W, woods. Maps and photos were obtained from the Geospatial Information Authority of Japan.

idue was then dissolved in 1 mL of ethanol and subjected to HPLC. The HPLC system consisted of a DGU-20A5 degasser, an LC-20AB binary pump, an SIL-20A auto injector, and a CTO-20AC column oven, obtained from Shimadzu (Kyoto, Japan), and two GL-7543 fluorescence detectors (GL Sciences, Tokyo, Japan). The separation column was Inertsil ODS-P (4.6 × 250 mm; GL Sciences), which was stored in a column oven set at 25°C. The mobile phase comprised a binary linear gradient of water and CH_3CN. The percentage of CH_3CN was 75% for 0–10 min, increased to 2.5%/min for 10–20 min, and maintained at 100% for 20–60 min. The flow rate was set at 1 mL/min. Excitation and emission wavelengths were 286 and 433 nm for fluoranthene, 311 and 392 nm for pyrene, 284 and 385 nm for benz[*a*]anthracene, 264 and 362 nm for chrysene, 300 and 428 nm for benzo[*b*]fluoranthene, 384 and 406 nm for benzo[*k*]fluoranthene and benzo[*a*]pyrene, 292 and 440 nm for dibenz[*a,h*]anthracene, 374 and 404 nm for benzo[*ghi*]perylene, and 294 and 482 nm for indeno[1,2,3-*cd*]pyrene, respectively.

RESULTS AND DISCUSSION

Saitama Prefecture stretches from east to west, with the Kanto Mountains to the west and the Kanto Plain to the east (Fig. 1, left). Ina Town is located almost in the center of the eastern plain, where more than 95% of the prefectural population lives. The landscape of Ina Town is mainly composed of residential buildings and agricultural fields without any heavy industries, such as thermal power plants or steel mills; therefore, automobiles and open burning of biomass are the major anthropogenic sources of $PM_{2.5}$ in the town. To reveal atmospheric $PM_{2.5}$ pollution in this suburban residential area of Saitama Prefecture, we collected particulate matter every week from 2016 to 2019 at Ina SS and measured the $PM_{2.5}$ and PAH concentrations.

Figure 2 shows the atmospheric concentrations of $PM_{2.5}$ and sum of 10 PAHs (ΣPAH) at Ina SS over the four years and their four-year mean concentrations. The four-year mean concentration of $PM_{2.5}$ was 15.3 μg/m³, and considering that the long-term environmental standard (annual mean) is 15 μg/m³, 15.3 μg/m³ is not low enough to not cause any adverse health effects. The daily $PM_{2.5}$ concentration exceeded the short-term environmental standard (daily mean) of 35 μg/m³ on five days. The highest $PM_{2.5}$ concentration (66.0 μg/m³) was recorded on April 3, 2018, and the third highest (50.1 μg/m³) was recorded on March 27, 2018. The particulate matter collected on these days was dark yellow. Further, these dates coincided with the arrival of the Yellow Sand (Asian Dust), as confirmed by the Japan Meteorological Agency,[7] which may be the main reason behind these high-concentration events. Onishi *et al.* reported that air pollutants such as heavy metals, which adversely affect human health, are transported over long distances by the Yellow Sand.[8,9] In the present study, we observed high concentrations of ΣPAH (1.25 and 2.33 ng/m³, respectively) on these days, indicating that high $PM_{2.5}$ pollution caused by the Yellow Sand can severely impact human health. Additionally, on February 7, 2017 and January 29, 2019, high $PM_{2.5}$ concentrations (60.1 and 42.8 μg/m³, respectively) were observed with strong wind (maximum instantaneous wind speed, 16.8 and 16.7 m/s, respectively),[10] indicating that these high $PM_{2.5}$ events were caused by lifted soil. In these days, ΣPAH concentrations were not high (0.48 and 0.47 ng/m³, respectively), suggesting that high $PM_{2.5}$ pollution caused by windy lifted soil is relatively less serious.

Next, to reveal the characteristics of $PM_{2.5}$ pollution at Ina SS, the monthly mean concentration of $PM_{2.5}$ at Ina SS was compared with those at the nearby Ageo (35°97′70″N, 139°57′12″E) and Hasuda (35°97′27″N, 139°64′95″E) public air monitoring stations (AS) installed by Saitama Prefectural Government. Because Ageo AS started to monitor $PM_{2.5}$ from 2017, hourly mean concentration data over three years (2017–2019) were downloaded from the website of Saitama Prefectural Government,[11] and the monthly mean concentration at Ina SS was calculated during the same sampling period. Figure 3 (top) shows the monthly and three-year mean concentrations of $PM_{2.5}$ at Ina SS, Hasuda AS, and Ageo AS. Three-year mean concentration was highest at Ina SS (13.1 μg/m³), followed by Hasuda AS (12.2 μg/m³), and was lowest at Ageo AS (10.8 μg/m³). Although the population density of Hasuda City (2,257 individuals/km²) is lower than that of Ageo City (4,966 individuals/km²), the $PM_{2.5}$ concentration at Hasuda AS was higher than that at Ageo AS. Furthermore, $PM_{2.5}$ concentration at Ina SS, which is surrounded by forests and fields (Fig. 1, second left), was unexpectedly higher than those at Hasuda AS and Ageo AS. Therefore, we compared the monthly mean $PM_{2.5}$ concentrations among these stations. In August, the $PM_{2.5}$ concentration was the same at all three stations; however, from

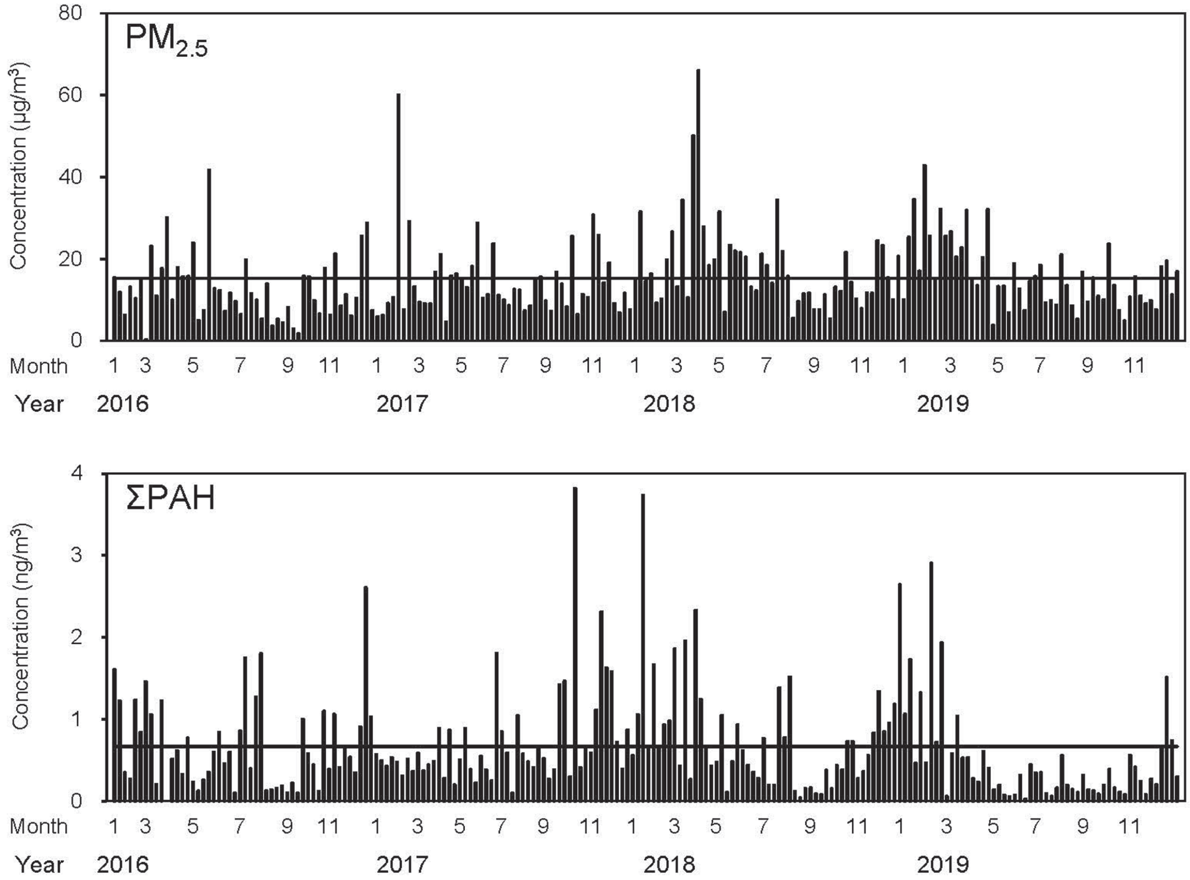

Fig. 2. Atmospheric Concentrations of $PM_{2.5}$ and ΣPAH at Ina SS.

The vertical bars and horizontal lines represent the daily and four-year mean concentrations, respectively.

October, the concentrations at Ina SS and Hasuda AS were higher than that at Ageo AS, and this trend continued till May. Hasegawa *et al.* reported that open burning of biomass is the main cause of high $PM_{2.5}$ concentrations from autumn to winter owing to the increasing atmospheric concentration of levoglucosan produced by the incomplete combustion of cellulose in chaff and straw during this period.[12–14] A similar phenomenon has been reported in Ibaraki Prefecture, which is adjacent to Saitama Prefecture.[15] Because Hasuda City and Ina Town are closer to farmlands than Ageo City (Fig. 1), biomass burning was believed to be the cause of the high $PM_{2.5}$ concentrations at Ina SS and Hasuda AS from autumn to winter. Figure 3 (bottom) shows monthly concentration of ΣPAH, which are derived from combustion of organic compounds, calculated from the data in Fig. 2 (bottom). ΣPAH concentration at Ina SS was also high from autumn to winter, suggesting that open combustion of biomass may elevate carcinogenic risk to humans. Furthermore, high $PM_{2.5}$ concentration at Ina SS was observed during spring. $PM_{2.5}$, which is formed in the atmosphere from volatile organic chemicals derived from plants,[16] may be present at a high concentration at the Ina SS in spring because it is surrounded by forests.

Finally, to compare atmospheric $PM_{2.5}$ and PAH pollution in Ina Town and central Tokyo, we sampled airborne particulates simultaneously for 28 days in Ina Town and Ueno, a major city in Tokyo, and measured their $PM_{2.5}$ and PAH concentrations. Figure 4 shows the atmospheric concentrations of $PM_{2.5}$ and

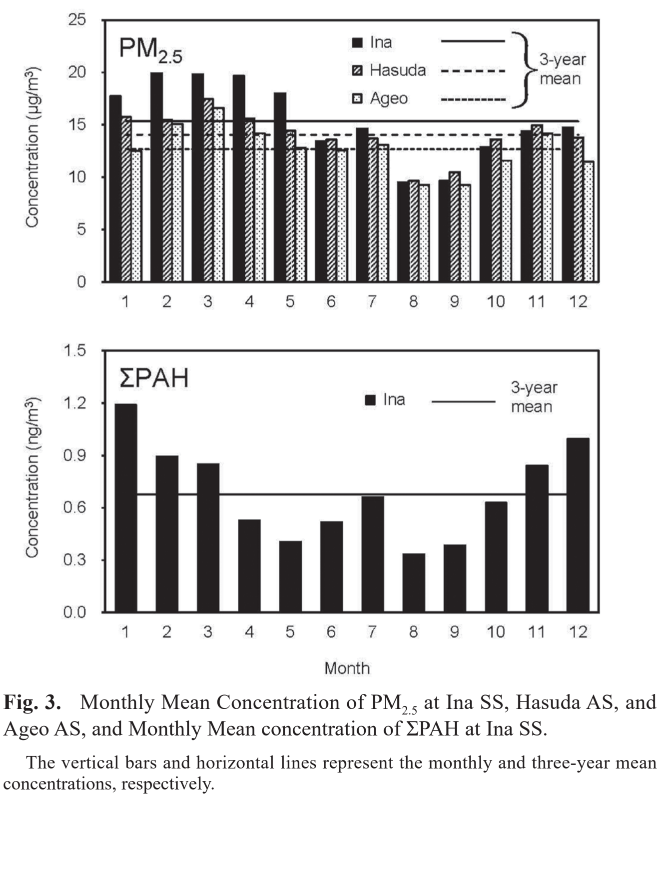

Fig. 3. Monthly Mean Concentration of $PM_{2.5}$ at Ina SS, Hasuda AS, and Ageo AS, and Monthly Mean concentration of ΣPAH at Ina SS.

The vertical bars and horizontal lines represent the monthly and three-year mean concentrations, respectively.

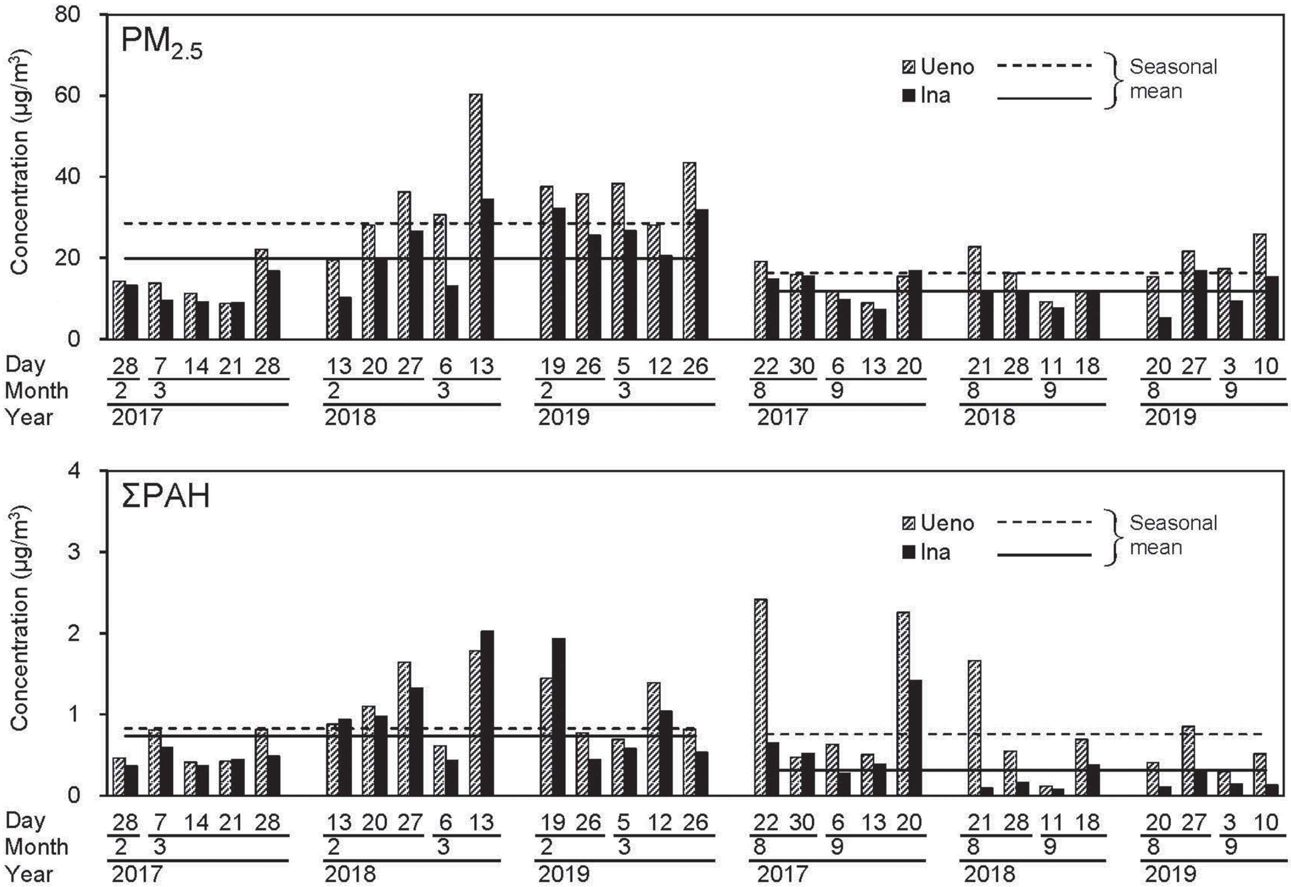

Fig. 4. Comparison of the $PM_{2.5}$ and ΣPAH Concentrations at Ueno SS and Ina SS.

The vertical bars and horizontal lines represent daily and seasonal mean concentrations, respectively.

ΣPAH at Ina SS and Ueno SS. The mean $PM_{2.5}$ concentration at Ueno SS was 28.6 μg/m^3 in late winter (February to March) and 16.3 μg/m^3 in late summer (August to September). The concentration at Ina SS during the same sampling period was 70% of that at Ueno; therefore, $PM_{2.5}$ pollution in Ina Town is serious despite its low population density (3,174 individuals/km^2), which is only 16% of that in Taito Ward, where Ueno is located (19,592 individuals/km^2). As for ΣPAH, the mean concentration in late winter at Ina SS (0.73 ng/m^3) was almost the same as that at Ueno SS (0.83 ng/m^3). This may have been due to additional sources of biomass burning around Ina SS. In late summer, without biomass burning, the mean concentration of ΣPAH at Ina SS (0.31 ng/m^3) was much lower than that at Ueno SS (0.76 ng/m^3).

In this study, we evaluated the atmospheric pollution caused by $PM_{2.5}$ in a suburban residential area and a metropolitan area. In the central area of the Kanto Plain, the accumulation of a large amount of $PM_{2.5}$ emitted from this area increases the atmospheric concentration of $PM_{2.5}$.[17,18] Therefore, the $PM_{2.5}$ concentration is not low even in suburbs with a low population density. Furthermore, atmospheric $PM_{2.5}$ concentrations in areas adjacent to fields were found to be higher than those in neighboring urban areas owing to the emission of $PM_{2.5}$ by biomass burning. To improve the environment in suburban residential areas surrounding fields, biomass burning needs to be limited. Over the last century, particulate matter emitted from automobiles, particularly diesel vehicles, has caused substantial air pollution. However, since 1999, the Tokyo Metropolitan and neighboring prefectural governments have tightened regulations on particulate matter emissions from diesel vehicles,[19] and the number of particulate matter-derived diesel vehicles has been steadily decreasing.[20,21] In recent years, the open burning of particulates has been regarded as a serious problem.[12] Nitrogen oxides and sulfur oxides, which are mainly emitted from mobile sources, are oxidized in the air to form nitrates and sulfates, respectively, and have been reported to increase $PM_{2.5}$ concentration.[22–24] Thus, the sources of $PM_{2.5}$ in the atmosphere are diverse, and the environmental dynamics of $PM_{2.5}$ are also complex. $PM_{2.5}$ exposure in humans is associated with the development of thrombosis and immune disorders, as well as cancers.[25,26] Currently, our research group is focusing on the components of $PM_{2.5}$ that cause diseases. *In vitro* experiments are in progress to elucidate the mechanism of disease development caused by the exposure to $PM_{2.5}$ and/or its extracts using macrophages and immune-related cells. Air pollution caused by $PM_{2.5}$ has continued to be a social concern, and we hope that our research will facilitate environmental protection.

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Conflict of interest The authors declare no conflict of interest.

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