



Ambient air pollution of particles and gas pollutants, and the predicted health risks from long-term exposure to PM_{2.5} in Zhejiang province, China

Yuanchen Chen¹ · Lu Zang¹ · Wei Du² · Da Xu³ · Guofeng Shen² · Quan Zhang¹ · Qiaoli Zou³ · Jinyuan Chen¹ · Meirong Zhao¹ · Defei Yao³

Received: 25 January 2018 / Accepted: 25 May 2018 / Published online: 6 June 2018

© Springer-Verlag GmbH Germany, part of Springer Nature 2018

Abstract

In recent years, ambient air has been severely contaminated by particulate matters (PMs) and some gas pollutants (nitrogen dioxide (NO₂) and sulfur dioxide (SO₂)) in China, and many studies have demonstrated that exposure to these pollutants can induce great adverse impacts on human health. The concentrations of the pollutants were much higher in winter than those in summer, and the average concentrations in this studied area were lower than those in northern China. In the comparison between high-resolution emission inventory and spatial distribution of PM_{2.5}, significant positive linear correlation was found. Though the pollutants had similar trends, NO₂ and SO₂ delayed with 1 h to PM_{2.5}. Besides, PM_{2.5} had a lag time of 1 h to temperature and relative humidity. Significant linear correlation was found among pollutants and meteorological conditions, suggesting the impact of meteorological conditions on ambient air pollution other than emission. For the 24-h trend, lowest concentrations of PM_{2.5}, NO₂, and SO₂ were found around 15:00–18:00. In 2015, the population attributable fractions (PAFs) for ischemic heart disease (IHD), cerebrovascular disease (stroke), chronic obstructive pulmonary disease (COPD), lung cancer (LC), and acute lower respiratory infection (ALRI) due to the exposure to PM_{2.5} in Zhejiang province were 25.82, 38.94, 17.73, 22.32, and 31.14%, respectively. The population-weighted mortality due to PM_{2.5} exposure in Zhejiang province was lower than the average level of the whole country—China.

Keywords Particulate matters · Nitrogen dioxide · Sulfur dioxide · Ambient air pollution · Meteorological conditions · Health risks

Responsible editor: Gerhard Lammel

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s11356-018-2420-5>) contains supplementary material, which is available to authorized users.

✉ Defei Yao
yaodefei@zjemc.org.cn

- ¹ Key Laboratory of Microbial Technology for Industrial Pollution Control of Zhejiang Province, College of Environment, Research Center of Environmental Science, Zhejiang University of Technology, Hangzhou 310032, China
- ² Ministry of Education Laboratory of Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China
- ³ Zhejiang Province Environmental Monitoring Center, Hangzhou 310012, China

Introduction

With the rapid economic development in China, emissions of particulate matters (PMs) and various gas pollutants including nitrogen oxide (NO_x), sulfur dioxide (SO₂), and ozone (O₃) have been increasing in the last several decades. In recent years, more and more haze incidences have been reported and gained high public attention (Chan and Yao 2008; Zhang et al. 2012; Meng et al. 2016). High concentration of PMs in the air, also of nitrogen dioxide (NO₂) and SO₂, generates strong adverse health impacts (Miller et al. 2007; Pope and Dockery 2006; Englert 2004; Khaniabadi et al. 2017; Kampa and Castanas 2008; Bravo et al. 2016), except from causing low visibility of the air. According to the latest report, the population attributable fraction (PAF) of the total mortality from ambient PM pollution in China was at the level of 16.2%, ranking fourth among the 67 risk factors (WHO 2015).

Numbers of studies have been conducted in China, measuring PMs, NO_x, and SO₂ in the ambient air (Lei et al. 2011; Zhou et al. 2015a; Yang et al. 2017). It was noted that severe ambient air pollution was attributable to emissions from industrial activities, vehicles, solid fuel combustion, power station, and so on. Especially, during winter, air pollution is much severer in northern China owing to heating activities, and due to large consumption of coal, it contributes to higher SO₂ levels in ambient air in northern China compared with southern China. Besides, the atmosphere in southeast China/Yangtze River Delta (like Shanghai, Jiangsu and Zhejiang) is also severely polluted now due to increasing energy consumption and emissions from various sources (Huang et al. 2014b; Tang et al. 2016), while seldom of them has been provided.

In addition to emission amounts, the concentrations of particles, NO₂, and SO₂ in the air were influenced by many other factors, such as meteorological conditions, including wind speed, temperature, and relative humidity (Huang et al. 2014a; He et al. 2017; Laña et al. 2016; Calkins et al. 2016). Investigating the relationship among contamination levels, emission amounts, and meteorological parameters can be used to predict ambient air pollution and the personal exposure concentrations for health risk assessments (Dominici et al. 2006; Delfino et al. 2005). During a whole day, air pollutants are also influenced by the meteorological conditions and vary in a large range between daytime and nighttime (Han et al. 2015). Thus, better understanding on the real-time 24-h concentration trend could be utilized for more accurate daily personal exposure prediction and the corresponding health outcomes (Chen et al. 2016; Chen et al. 2017a).

In this study, the hourly concentrations of PM_{2.5} (PM with aerodynamic diameter lower than 2.5 μm), PM₁₀ (PM with aerodynamic diameter lower than 10 μm), NO₂, and SO₂, and the corresponding levels of several meteorological parameters were simultaneously measured in 13 sites of three typical cities of Zhejiang province, China, during summer and winter time from 2015 to 2016. The objectives were as follows: (1) comparing the contamination level of various pollutants in different sites and relating concentrations of particles with emission inventory, (2) analyzing the influence of meteorological conditions on airborne pollutants in different sites, (3) illustrating the trend of 24-h pollutant concentrations and the influence from meteorological parameters, and (4) calculating relative risks (RRs) and population attributable fractions (PAFs) for the mortalities of diseases—ischemic heart disease (IHD), cerebrovascular disease (stroke), chronic obstructive pulmonary disease (COPD), lung cancer (LC), and acute lower respiratory infection (ALRI) (for lower than 5 years old children).

Methodology

Studied areas

Thirteen sites were involved in this study, which were in three typical cities in Zhejiang province. Zhejiang province was located in the southeast of China (Yangtze River Delta), which is a well economic-developed area in southeast China. The three cities were Hangzhou, Shaoxing, and Taizhou, which represent metropolis/capital city, industry city, and mountain area/light industry city, respectively. Hangzhou held the G20 Summit in 2016, which was now considered one of the largest cities in China. All the locations of the sites are marked in Fig. 1 (regional maps) and listed in Table A1. Among the 13 sites, some of them are downtown areas (like Hangzhou-1), some of them are industry areas (like Shaoxing-1), and some of them are in mountain or natural reserve areas (like Hangzhou-6 and Taizhou-6). The two natural reserve areas (Hangzhou-6 and Taizhou-6) have no factory or other emission source nearby.

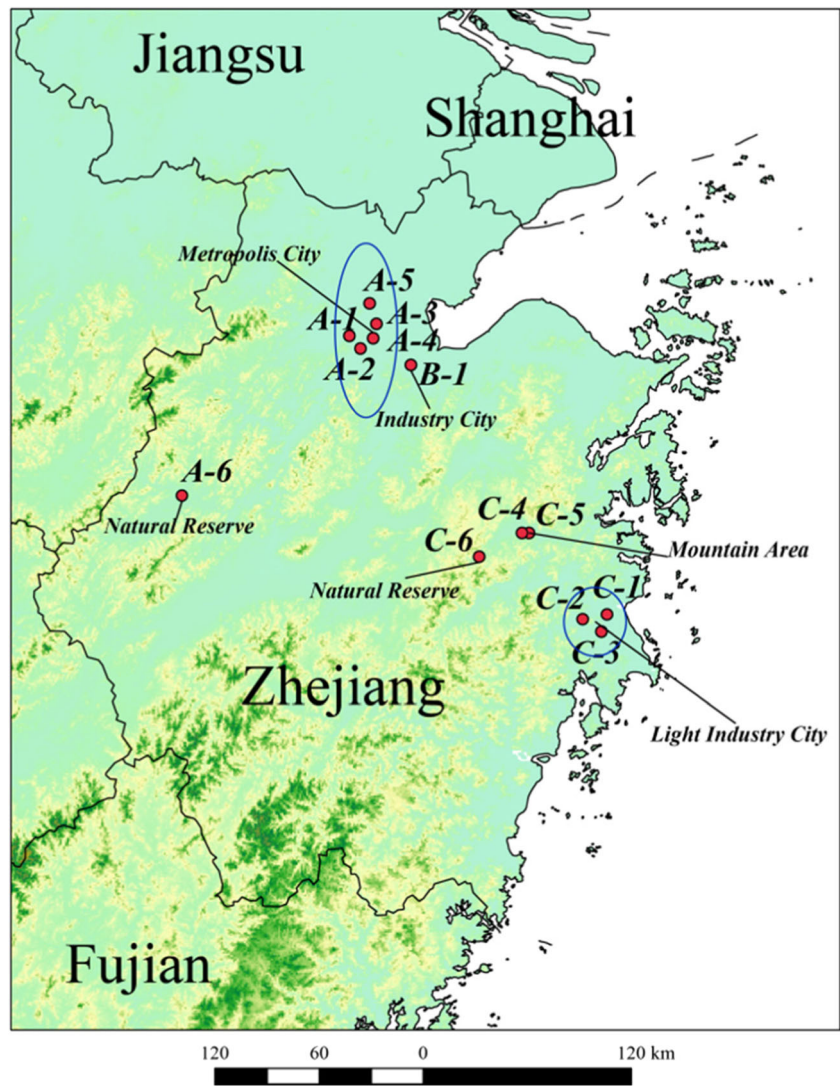
Equipment and measurement

The concentrations of PMs (including PM₁₀ and PM_{2.5}) were measured continuously with synchronized hybrid ambient real-time particulate monitor (Model 5030 SHARP monitor, ThermoFisher Scientific Inc., USA) in different sites mentioned above. And, the concentrations of NO₂ and SO₂ were measured by Model 42i-NO₂ analyzer (ThermoFisher Scientific Inc., USA) and Model 450i-SO₂ analyzer (ThermoFisher Scientific Inc., USA), respectively. Meanwhile, meteorological parameters (including temperature, relative humidity, and wind speed) were also measured (weather stations, called as Swarco-lufft WS500-UMB (Sutron Corporation, Germany)). The time intervals for the concentrations of PMs, NO₂, SO₂, and meteorological parameters were all 1 h. Daily and seasonal averages were derived for various purposes. The data covered four periods: (1) December 1, 2014–January 31, 2015; (2) June 1, 2015–August 31, 2015; (3) December 1, 2015–January 31, 2016; and (4) June 1, 2016–August 31, 2016, during winter and summer, respectively.

Data analysis and uncertainties

Kolmogorov-Smirnov *Z* statistical test was used for the comparison between two samples, and one-factor variance analysis was used to compare the concentrations of PMs in the air under different sites and meteorological conditions. A “delayed correlation” method was used to determine the time lag among temporal trends of pollutants and meteorological conditions. Pearson test was used for the linear correlation among various pollutants and meteorological parameters, after

Fig. 1 The basic information of the monitoring sites. “A,” “B,” and “C” in the map represent Hangzhou, Shaoxing, and Taizhou, respectively



log-transforming the PM, NO₂, and SO₂ data, because they are under logarithmic normal distribution (Chen et al. 2017b). The software of SPSS 13.0 (SPSS Inc., USA) was used for the statistical analysis. 0.05 was set as the significant level during all the analyzing processes. The energy consumption and the amounts of PM_{2.5} emission for Zhejiang province were calculated, respectively. Four emission sources were considered during the calculation, including power station, industry, residential source, and vehicle emission. The calculating method with data of emission factors and energy consumption was derived from previous emission inventory studies (Huang et al. 2014b; IEA 2012; Wang et al. 2012).

The risk from long-term exposure to particles was assessed in this study according to the methods provided in a previous study by Burnett et al. (2014). The method named as Integrated Exposure-Response (IER) model was developed for the Global Burden Disease (GBD) study on RRs and PAFs by integrating various cohort studies on ambient air pollution, second-hand smoke, active smoking, and

household air pollution from solid fuel use in the USA, Europe, and some developing countries, such as China. According to some previous studies, the assumption was made that yearly concentration of PM_{2.5} can represent the long-term exposure concentration when calculating the all-cause mortality during a whole year (Burnett et al. 2014; Lelieveld et al. 2015; Liu et al. 2016; Feng et al. 2017; Song et al. 2017). Thus, in this study, annual average concentration of PM_{2.5} derived from the 24-h data for different seasons and various sites was used. Based on the IER model, the RRs for the mortalities of all-aged population for IHD, stroke, COPD, LC, and incidence of ALRI were calculated by the mass exposed particles (PM_{2.5}). The IER model has been found to be the best method for calculating RR compared with several other models. The predicting model can be described as:

$$RR(z) = \begin{cases} 1 & , \text{ if } z \leq z_0 \\ 1 + \alpha [1 - e^{-\gamma(z-z_0)^\delta}] & , \text{ if } z > z_0 \end{cases}$$

where z represents exposure concentration to $PM_{2.5}$ in the unit of $\mu\text{g}/\text{m}^3$, and z_0 presents the counterfactual concentration below which no additional risk is assumed. The parameters— α , γ , δ , and z_0 —were derived from the fitting process using available RR information from previous studies (Burnett et al. 2014). One thousand sets of the joint parameter distributions were used in the calculation for various diseases. Besides, the mean levels of $PM_{2.5}$ concentrations in different sites were applied in the model. Furthermore, PAF for every outcome due to exposure to $PM_{2.5}$ can be qualified as $1-1/\text{RR}$ (Smith et al. 2014). Furthermore, the premature mortality or excess death (ED) for a specific disease attributable to $PM_{2.5}$ inhalation exposure can be calculated by multiplying its PAF value with total mortality for corresponding disease (Anenberg et al. 2010; Evans et al. 2013; Lelieveld et al. 2015). MATLAB software (MathWorks, USA) was used for the Monte Carlo simulation method with about 100,000 times during every calculation, resulting in energy consumption for various sources, emission amounts of $PM_{2.5}$.

Results and discussion

Contamination levels of various pollutants

Based on the results in a previous study, the concentrations of PMs, NO_2 , and SO_2 were all log-normally distributed (Chen et al. 2017a). Thus, geometric means and geometric standard deviations were used in the comparison for concentrations in different sites and seasons. From the measurements (seasonal average data), the geometric mean (geometric standard deviation) concentrations of $PM_{2.5}$ during winter were 71.79 (1.57), 78.32 (1.55), and 52.33 (1.62) $\mu\text{g}/\text{m}^3$, and during summer were 30.29 (1.55), 31.30 (1.66), and 22.08 (1.59) $\mu\text{g}/\text{m}^3$ for Hangzhou, Shaoxing, and Taizhou, respectively. Apparently, relatively higher concentrations were found in Shaoxing during winter ($p < 0.05$). The ambient air quality standard of daily concentration of $PM_{2.5}$ in China is 75 $\mu\text{g}/\text{m}^3$ (Ministry of Environmental Protection of P.R.C. 2012). The comparing results showed that 56, 61, and 15% days for Hangzhou, Shaoxing, and Taizhou exceeded the standard during winter; none of the daily concentration exceeded the standard during summer. When compared with the national ambient air quality standards (NAAQS) for daily concentrations of $PM_{2.5}$ in ambient air (35 $\mu\text{g}/\text{m}^3$) from the U.S. Environmental Protection Agency (USEPA) (USEPA 2008), much more percent days in these three cities exceeded the limits (97, 98, and 92% for Hangzhou, Shaoxing, and Taizhou during winter, and 42, 47, and 8.7% during summer, respectively). For PM_{10} , the concentration during winter were 105.70 (1.57), 108.12 (1.56), and 84.46 (1.57) $\mu\text{g}/\text{m}^3$, and during summer were 48.99 (1.51), 45.70 (1.57), and 37.66 (1.44) $\mu\text{g}/\text{m}^3$ for

Hangzhou, Shaoxing, and Taizhou, respectively. The contamination level of PM_{10} during winter was highest in Shaoxing and lowest in Taizhou ($p < 0.05$).

The concentrations of NO_2 were 57.61 (1.36), 60.87 (1.30), and 32.71 (1.41) $\mu\text{g}/\text{m}^3$ during winter, and 27.87 (1.35), 24.58 (1.35), and 11.58 (1.58) $\mu\text{g}/\text{m}^3$ during summer for Hangzhou, Shaoxing, and Taizhou, respectively. Besides, the concentrations of SO_2 were 19.99 (1.49), 36.98 (1.48), and 11.65 (1.69) $\mu\text{g}/\text{m}^3$ during winter, and 7.99 (1.31), 13.20 (1.43), and 4.85 (1.52) $\mu\text{g}/\text{m}^3$ during summer for Hangzhou, Shaoxing, and Taizhou, respectively. According to the NAAQS for NO_2 (80 $\mu\text{g}/\text{m}^3$), 10, 11, and 0% days exceeded during winter in Hangzhou, Shaoxing, and Taizhou, but none of the days exceeded the limit during summer (Ministry of Environmental Protection of P.R.C. 2012). For SO_2 (150 $\mu\text{g}/\text{m}^3$), during both seasons, none of the days were higher than the limit (Ministry of Environmental Protection of P.R.C. 2012). Similarly, compared with the standard limits from USEPA (NO_2 69 $\mu\text{g}/\text{m}^3$; SO_2 97 $\mu\text{g}/\text{m}^3$), which were lower than the national standard in China, more percent days were expected to exceed the limits (USEPA 2008).

Owing to substantially solid fuels using during winter time for cooking and space heating activities (Zhang and Smith 2007), some studies reported severer air pollution in northern China compared with that in southeast China. For example, the daily concentration of $PM_{2.5}$ during the wintertime in Baoding, Hebei province, was at the average level of 119 and 110 $\mu\text{g}/\text{m}^3$ in Xingtai which is also in Hebei province (Zhang and Cao 2015); the concentrations were almost twice as those in this study. In a previous study, the concentration of SO_2 during winter in Jing-jin-ji area was 161 $\mu\text{g}/\text{m}^3$, which was five to eight times higher than that in this study (Jiang et al. 2015). And, for NO_2 , the concentration was found at the level of 78 and 69 $\mu\text{g}/\text{m}^3$ during winter in Jing-jin-ji area and North China plain respectively, both of which were much higher than that in Zhejiang province (Jiang et al. 2015).

For comparison of the ambient pollutant concentrations among cities, regions, and seasons, the seasonal average data were also employed. The means and standard deviations of $PM_{2.5}$, PM_{10} , SO_2 , and NO_2 for different seasons of different sites were graphed in Fig. 2. For all the sites, concentrations of $PM_{2.5}$ and PM_{10} during winter were about two times higher than summer, although not as high as those in northern cities in China (Zhang and Cao 2015). The similar results were also obviously found for NO_2 and SO_2 . The concentrations of NO_2 and SO_2 were significantly higher during winter compared with those during summer ($p < 0.05$). It is similar with previous reports due to more heating activities taken place (Li et al. 2013; Chafé et al. 2015), and also due to the meteorological condition which can restrict the diffusion of ambient air pollutants during winter (Ji et al. 2014).

In addition, Fig. 2 apparently illustrates that the two rural areas (Hangzhou-6 and Taizhou-6), both of which were

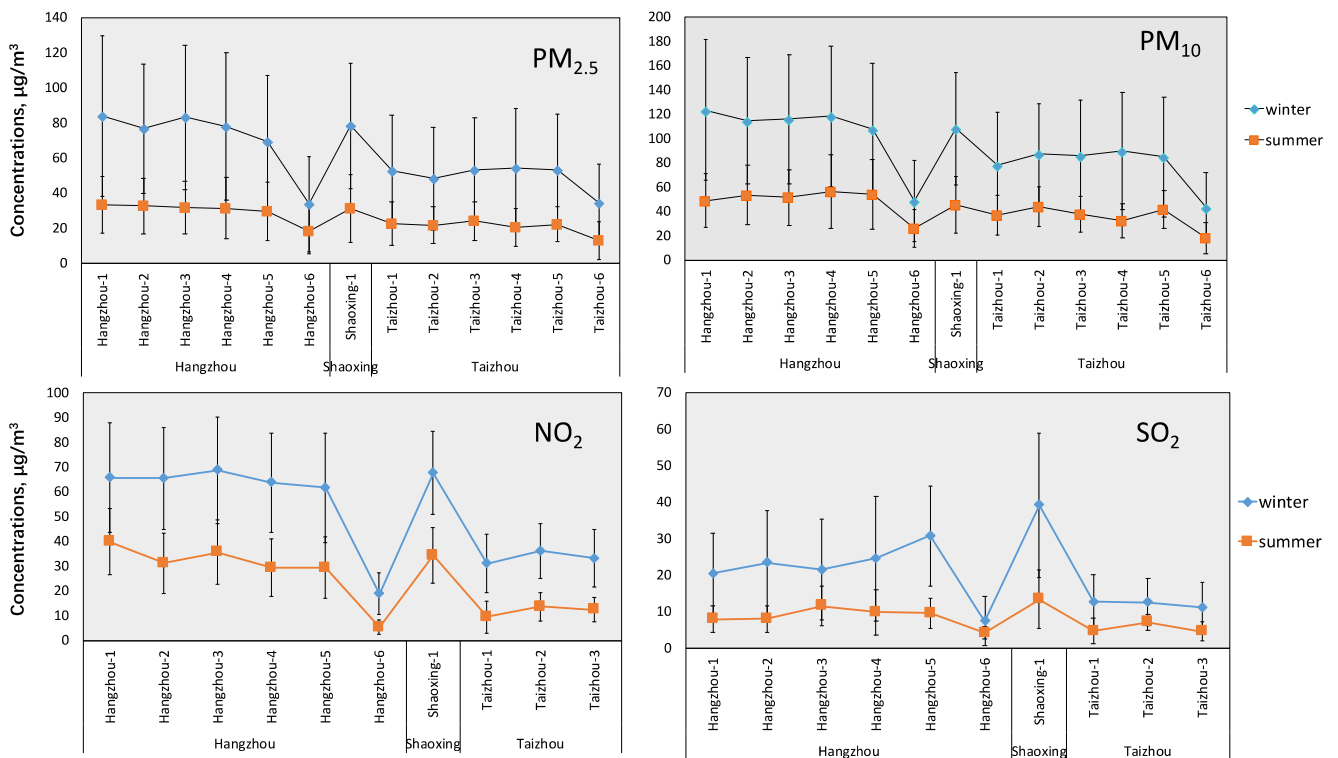


Fig. 2 Concentrations of PM_{2.5}, NO₂, and SO₂ of different sites in winter and summer from 2014 to 2016. Means and standard deviation are also shown. NO₂ and SO₂ were not measured in Taizhou-4, Taizhou-5, and Taizhou-6

natural reserve areas, had the lowest concentrations of PM_{2.5} and PM₁₀ ($p < 0.05$). In addition, for the gas pollutants (NO₂ and SO₂), the lowest concentrations were also found in Hangzhou-6 in both seasons ($p < 0.05$). It was because there is no industrial or seldom residential emissions in the areas of Hangzhou-6 (Qiandao Lake) and Taizhou-6 (Lishimen Reservoir). Most seasonal concentrations of PM_{2.5}, NO₂, and SO₂ from daily data of the sites in Hangzhou and Shaoxing were statistically higher than those in Taizhou ($p < 0.05$). As the results shown above, comparing the PM, NO₂, and SO₂ concentrations among the three cities based on the average concentration for a city, Shaoxing had the highest levels, and Taizhou had the lowest concentrations levels of various pollutants for each period ($p < 0.05$). The results suggested that metropolis city (Hangzhou) and industry city (Shaoxing) own large energy consumption would have severer ambient air pollution compared with that in mountain areas or light industry city (Taizhou).

Furthermore, it can be demonstrated by the PM_{2.5} emission results from emission inventory for these three cities. For example, based on a newly released global fuel data product (PKU-FUEL-2014) and an emission factor database, the emission density of primary PM_{2.5} (ton/km²/year) was calculated and illustrated in Fig. 3 with the high resolution of 0.1° × 0.1° (10 km × 10 km) for one grid. It reveals that in the area of Hangzhou and Shaoxing except the site of Hangzhou-6, the emission density was very high (dark red areas), and those

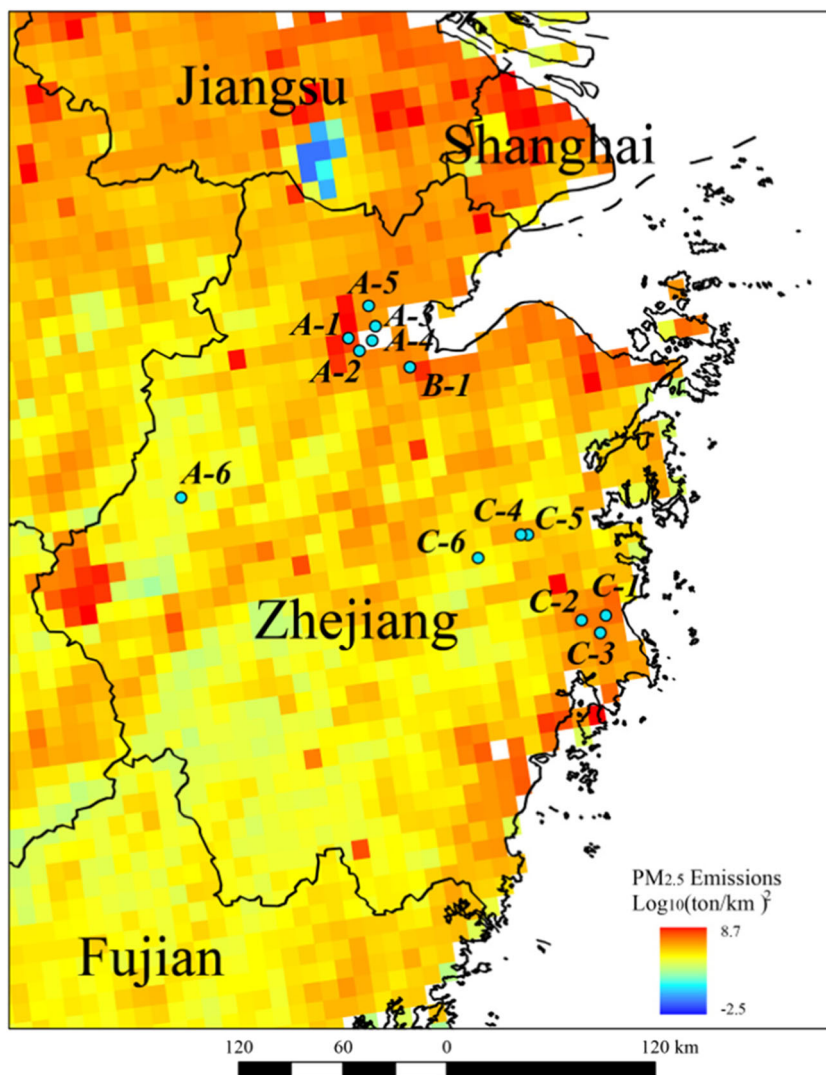
were relatively low in Taizhou. Apparently, the results of the emission inventory were comparable to the concentrations of PM_{2.5} in the air for the individual 13 sites. Moreover, the relationship between emissions and concentrations was studied, also graphed in Fig. A1, suggesting significantly positive linear correlation between them ($p < 0.05$). Therefore, it could be concluded that the emission amount can dominate the air PM pollution.

Sometimes, the ratio of PM_{2.5} to PM₁₀ can indicate the emission source. In the comparison between different seasons, though most ratios of PM_{2.5} to PM₁₀ for winter were higher than those during summer, no significant difference was found ($p > 0.05$) (Fig. 4). In addition, for the comparison of PM_{2.5}/PM₁₀ among different sites during the same season, no significant difference has been found ($p > 0.05$). It indicated that the emission sources of particles were similar among different sites and seasons in the studied areas.

Temporal trends and influencing factors on ambient air pollution

As discussed above, we found that owing to large fuel consumption and emission, the ambient air was contaminated by various ambient air pollutants in Zhejiang province, such as particles, NO_x, and SO₂. In addition, large differences were also found among different sites and seasons. Moreover, to obtain and reveal the influencing factors on the concentrations

Fig. 3 The emission density (ton/km²/year) of PM_{2.5} in the monitoring area. The letters “A,” “B,” and “C” in the graph represent Hangzhou, Shaoxing, and Taizhou, respectively

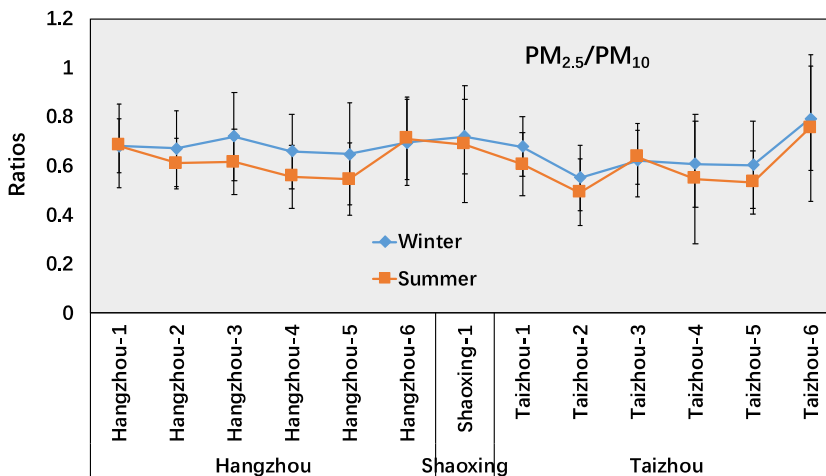


of ambient air pollutants, the temporal trends from hourly data were employed in this section.

The relationship among the ambient air pollutants is expected to help us to understand the characteristics of

various pollutants in ambient air in southeast of China, and the influencing effects of one pollutant on the others. The temporal trends of PM_{2.5}, NO₂, and SO₂ are shown in Fig. A2 with the hourly concentrations from 2015 to 2016.

Fig. 4 Ratios of PM_{2.5} to PM₁₀ (PM_{2.5}/PM₁₀) of the 13 monitoring sites during winter and summer from 2014 to 2016



The shape of the curves is similar with each other, though $PM_{2.5}$ has the highest concentration and that of SO_2 is the lowest during the same time. However, from the similar shape of the different temporal trends, lag time was found among the three curves. Then, a delayed correlation method was used to verify and determine the lag time among the temporal trends of the three kinds of pollutants. It was found that the concentrations of NO_2 and SO_2 in ambient air delayed with approximate 1 h after that of $PM_{2.5}$, and no lag was found between NO_2 and SO_2 . It demonstrated that though particle and gas pollutant pollution is mainly derived from emissions from fuel combustion, there are some underlying influencing factors on the ambient air pollution from particles and gas pollutants. Therefore, the temporal trends of some meteorological parameters (including temperature, relative humidity, and wind speed) are taken into account. On the contrary, the lag time of temperature and wind speed was both -1 h to $PM_{2.5}$, and no lag was found between $PM_{2.5}$ and relative humidity. That is, the trend of $PM_{2.5}$ delayed 1 h after temperature and wind speed, and the trends of NO_2 and SO_2 delayed 2 h after the meteorological parameters.

To further illustrate the influencing effect of different factors, the correlations among $PM_{2.5}$, NO_2 , SO_2 , temperature, relative humidity, and wind speed are plotted respectively for winter and summer seasons in Fig. 5. Owing to the log-normal distribution found for the concentrations of the pollutants, they were all log-transferred for the correlation analysis. Besides, the data were all replaced backward or forward with 1–2 h to eliminate the lag time among $PM_{2.5}$, NO_2 , SO_2 , temperature, and wind speed. For the relationship among the pollutants, obviously, the concentrations of $PM_{2.5}$, NO_2 , and SO_2 were positively linearly correlated with each other ($p < 0.05$), verifying the interacting effect among these typical air pollutants.

It was previously found that ambient air pollution is affected by some meteorological conditions, such as temperature, relative humidity, wind, and precipitation (Xu et al. 2011; Zhou et al. 2015b). The association between the air pollutants and meteorological conditions is also demonstrated here. For temperature, significantly negative linear correlation was found with all the air pollutants ($PM_{2.5}$, NO_2 , and SO_2) ($p < 0.05$), that is, under lower temperature, even during winter, the concentrations of the pollutants were turned out to be higher. It was because the thermal inversion layer is easy to be formed during lower troposphere temperature, which would be a barrier for the diffusion of the pollutants.

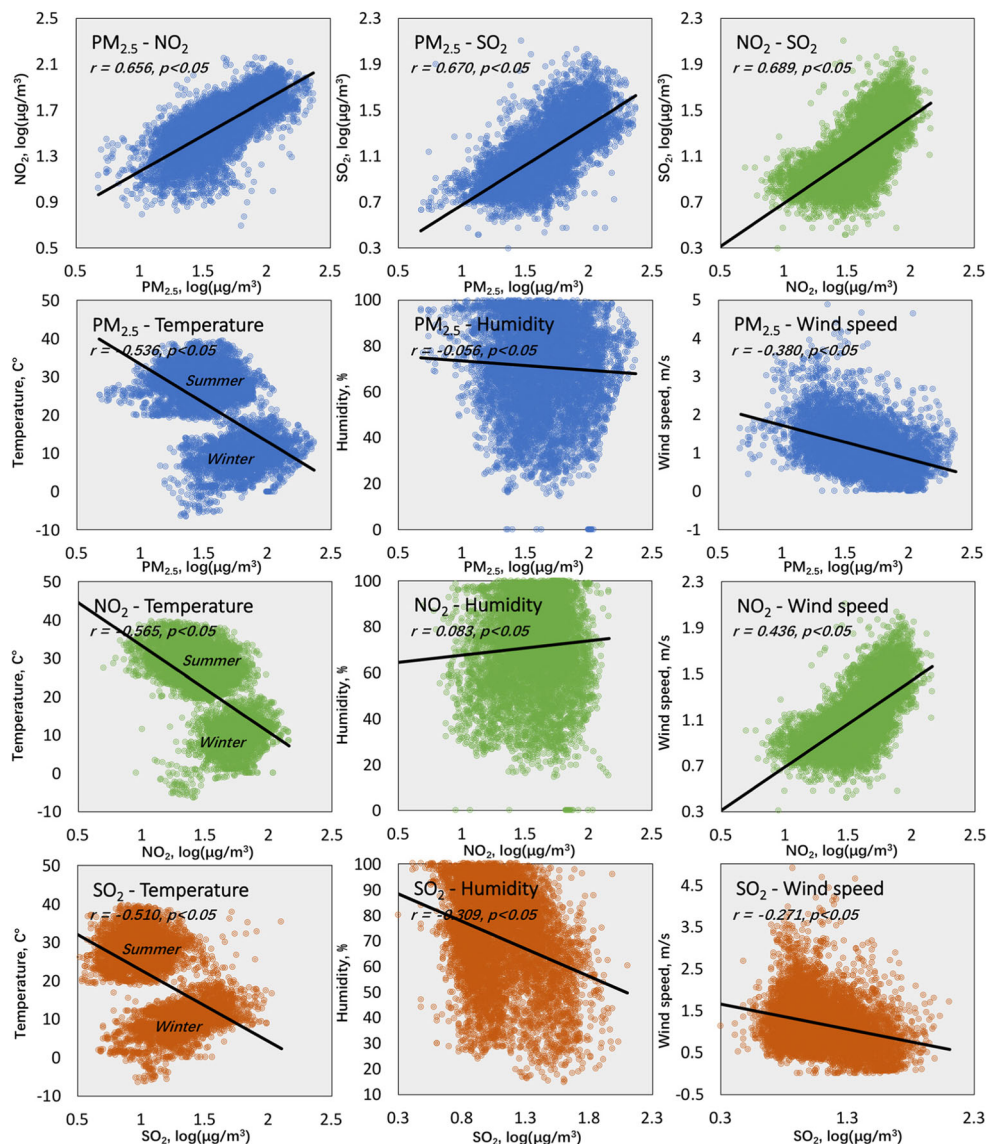
For relative humidity, the concentrations of $PM_{2.5}$ and SO_2 were found negatively correlated with it ($p < 0.05$). It is because somehow high relative humidity can indicate more wet precipitation, which is conducive to particle removal in the ambient air (Tai et al. 2010). In addition, the correlation between $PM_{2.5}/SO_2$ and wind speed is also illustrated in Fig. 5. It

could be clearly seen that the concentrations of $PM_{2.5}$ and SO_2 were significantly negatively linearly correlated with wind speed ($p < 0.05$), suggesting higher wind speed can accelerate their removal from ambient air. However, the correlation results for NO_2 with relative humidity and wind speed were just opposite to those for $PM_{2.5}$ and SO_2 , because higher relative humidity is beneficial for the oxidation from NO to NO_2 . The opposite results for the association between NO_2 and wind speed (positive linear correlation) suggested some underlying reasons which should be further studied in the near future.

For further study on the influence of meteorological conditions on ambient air pollution, the daily trend (24 h) of the concentrations of particles and gas pollutants and meteorological conditions was studied through the hourly data. The concentrations can be divided into two groups—daytime (5:00–18:00, roughly based on the sunrise and sunset times) and nighttime (roughly from 18:00 to 5:00 in the next day)—during both of the seasons—winter and summer. The daytime concentrations of $PM_{2.5}$, NO_2 , and SO_2 were 74 ± 5.7 , 49 ± 3.3 , and $25 \pm 2.4 \mu\text{g}/\text{m}^3$ during winter and 30 ± 1.1 , 23 ± 4.8 , and $9.2 \pm 1.4 \mu\text{g}/\text{m}^3$ during summer, and the nighttime concentrations of $PM_{2.5}$, NO_2 , and SO_2 were 75 ± 1.2 , 58 ± 6.9 , and $23 \pm 1.0 \mu\text{g}/\text{m}^3$ during winter and 31 ± 0.36 , 29 ± 2.0 , and $8.8 \pm 0.88 \mu\text{g}/\text{m}^3$ during summer, respectively. Obviously, significantly higher concentration of SO_2 was found during daytime in both seasons ($p < 0.05$), which could be explained by the reason of more emission amount during daytime. For $PM_{2.5}$ and NO_2 , there was no significant difference between daytime and nighttime ($p > 0.05$) owing to large variations. Some studies conducted in northern China found much higher concentrations of ambient air pollutants during nighttime (Han et al. 2015), which was not consistent with that in this study. The little difference between day-night temperatures in southeast of China (this studied areas) leads to the close contamination levels of $PM_{2.5}$ and NO_2 between daytime and nighttime.

For further discussion of the influence of meteorological conditions on ambient air pollution, Fig. 6 illustrates the daily trends (24 h) of concentrations of $PM_{2.5}$, NO_2 , and SO_2 , and meteorological conditions of temperature, wind speed, and relative humidity. It clearly reveals that during winter, the concentrations of $PM_{2.5}$ dramatically decreased from the noon (about 9:00–10:00) to the lowest level of approximate $65 \mu\text{g}/\text{m}^3$ (15:00–16:00), and peak of the concentration happened before the noon due to the accumulation of the particles from combustion processes and the accumulation through the nighttime under the influence of meteorological conditions. However, the trend of the concentrations of $PM_{2.5}$ was relatively stable during nighttime, since seldom human activities happen during this period. During summer, the trend was more stable (coefficient of variation (CV) 2.8%) compared with that during the winter time (CV 5.5%), although the shape was similar

Fig. 5 The correlation among concentrations of PM_{2.5}, NO₂, and SO₂, and meteorological conditions (temperature, relative humidity, and wind speed) during winter and summer periods in 2015–2016. The hourly data are used, and the concentrations of PM_{2.5}, NO₂, and SO₂ are log-transformed



with each other (valley 15:00–16:00; peak 9:00–10:00). It could further demonstrate the influencing impact of meteorological conditions on the PM pollution. It illustrates that the trend of temperature was opposite to PM_{2.5} trend, which was consistent with the results of the relationship between PM concentration and temperature (negative linear correlation). When the concentration of the PM_{2.5} reached the bottom level (15:00–16:00), the temperature was highest during a day. The reason has been discussed above that higher temperature is conducive to the dispersion of the particles in the air compared with lower temperature. Similarly, the trend of the wind speed was comparable with that of temperature, though the levels for winter and summer time were very close. Therefore, it also indicated that stronger wind speed is conducive to the dispersion of particles in the air, which was also consistent with the results discussed in the relationship between PM

pollution and wind speed. In addition, the similar trend was also found for relative humidity (lowest during 15:00–16:00). It also demonstrated the association between PM_{2.5} and relative humidity discussed above.

For SO₂, the temporal trend during winter was similar with that of PM_{2.5}, though it had a lag time (about 2 h) to the trend of PM_{2.5}. And the more stable trend during summer was attributable to fewer emission. It also revealed that the meteorological conditions along with emission have great impacts on the temporal trend of SO₂. Moreover, though the trend of NO₂ also reached the bottom in the afternoon, the highest level was found during nighttime—around 18:00–19:00 during winter and 21:00–22:00 during summer. It could be explained by the high relative humidity level during nighttime, because the relative humidity is positively correlated with the NO₂ pollution, which has been demonstrated in the last section.

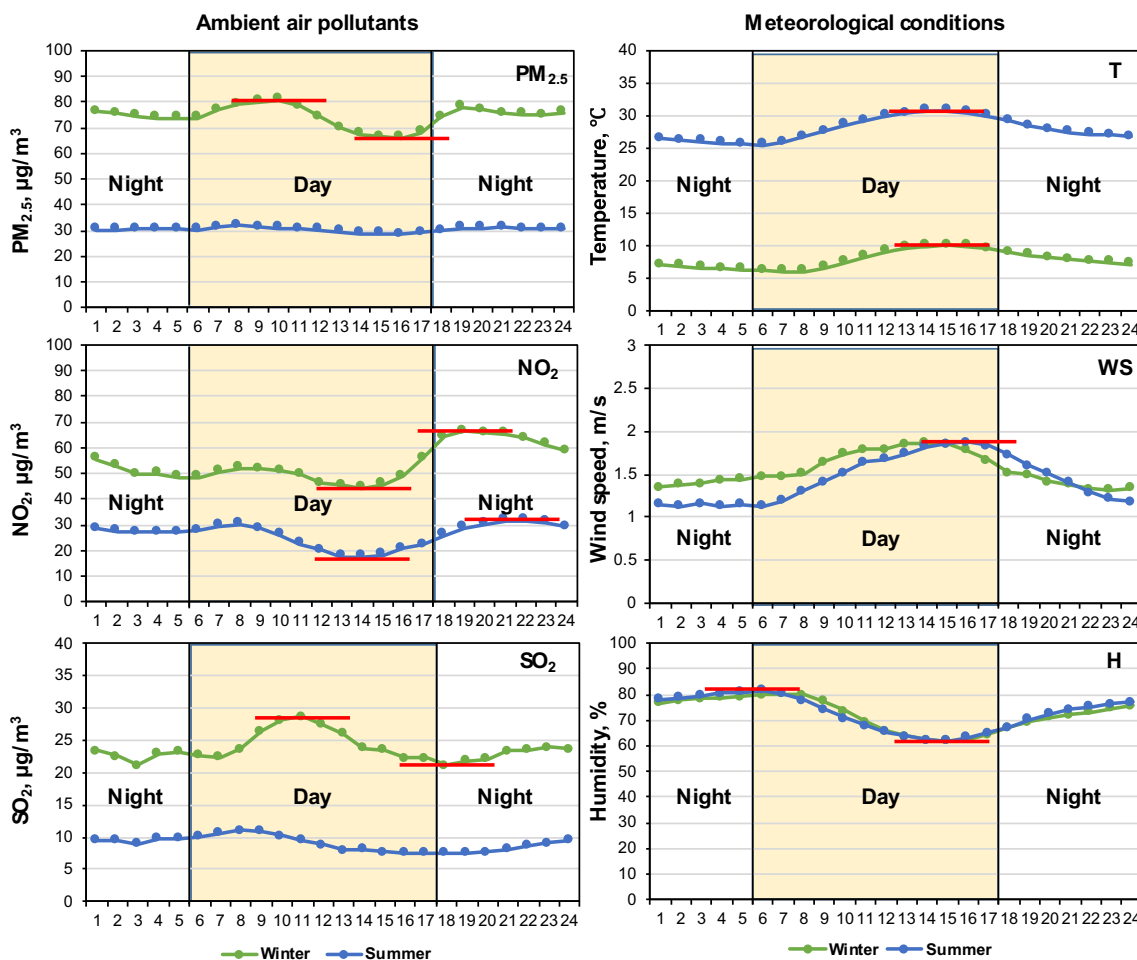


Fig. 6 The daily trends (24 h) of the pollutants (PM_{2.5}, NO₂, and SO₂) and meteorological conditions (temperature (T), wind speed (WS), and relative humidity (H))

Risk prediction on exposure to PM_{2.5} and its limitation

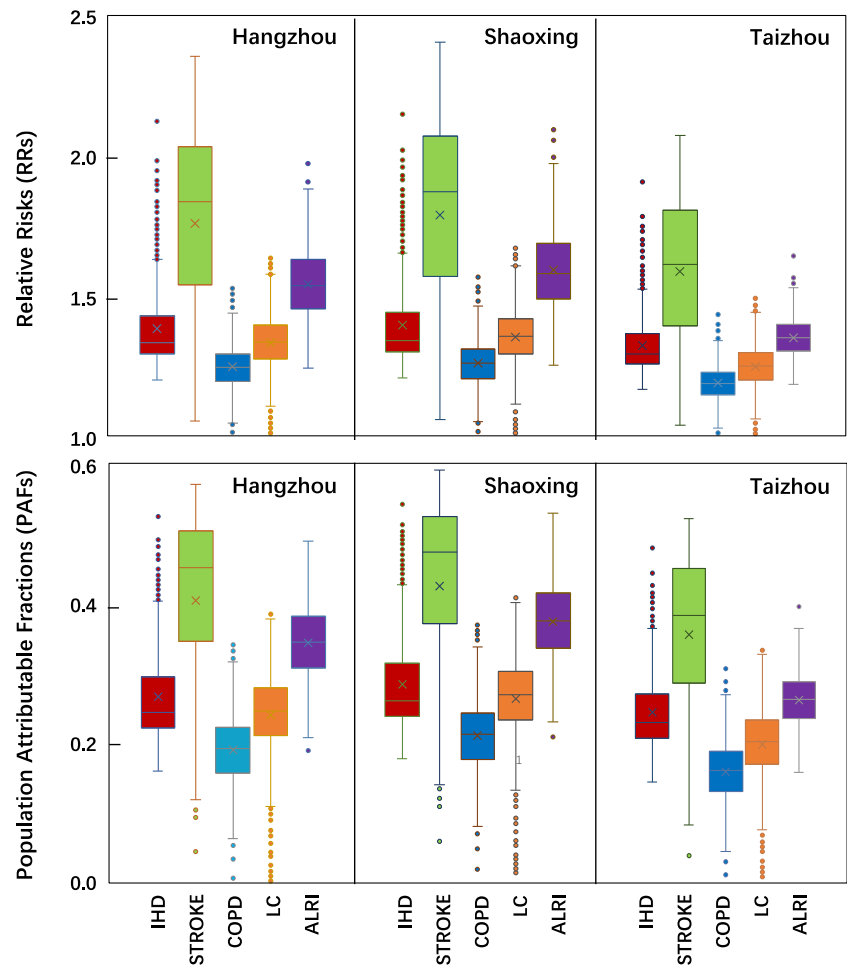
Since the exposure to PMs can induce adverse health impacts on human health, in this study, the RRs and PAFs for various diseases (including IHD, stroke, COPD, LC, and ALRI) in these three cities and 13 sites were calculated based on IER model with 1000 times simulation from the variation of the model parameters (Burnett et al. 2014). The RRs of IHD, stroke, COPD, LC, and ALRI due to inhalation exposure to PM_{2.5} in Hangzhou, Shaoxing, and Taizhou are illustrated in Fig. 7. Apparently, for each of the diseases, the RR was found statistically highest in Shaoxing (an industry city), and lowest in Taizhou (a mountain area or light industry city) ($p < 0.05$). For the comparison of the RRs of different diseases in the same city, it was noted that the RR of stroke induced by the exposure to PM_{2.5} was significantly highest among these five diseases, followed by ALRI, IHD, and LC, and the statistically lowest level was found for COPD ($p < 0.05$). Besides, RRs of the five diseases in the 13 sites are listed in Table A2, separately. It was found that the residents in Hangzhou-6 (Qiandao Lake) and Taizhou (Lishimen Reservoir) had the lowest risks

due to the inhalation exposure to PM_{2.5} ($p < 0.05$), because these two sites are both natural reserve areas owning lower ambient air PM_{2.5} pollution levels as discussed above.

In addition, based on the equation— $PAF = 1 - 1/RR$ —PAFs for different diseases in Hangzhou, Shaoxing, and Taizhou were calculated and graphed in Fig. 7. Similarly, the comparison results among different sites were comparable to those of RRs, illustrating statistically highest levels in Shaoxing and lowest levels in Taizhou ($p < 0.05$). Besides, for the comparison of PAFs among various diseases in the same site, stroke (cardiovascular disease) ranked as the highest one, followed by ALRI, IHD, and LC, and COPD was the lowest one ($p < 0.05$), indicating strongest association between ambient air PM_{2.5} exposure and ED of stroke. In addition, the PAFs of the five diseases in the 13 sites were also separately listed in Table A3. Like the RR comparison results, the PAF of various diseases in Qiandao Lake and Lishimen Reservoir ($p < 0.05$) indicates that the air in the natural reserve city is healthier compared with industry city which has higher emissions.

Combining the data for three cities together (typical cities in Zhejiang province), the mean levels of PAFs for IHD,

Fig. 7 Box plots of the relative risks (RRs) and population attributable fractions (PAFs) for the five diseases (IHD, stroke, COPD, LC, and ALRI) in Hangzhou, Shaoxing, and Taizhou of Zhejiang province



stroke, COPD, LC, and ALRI due to the exposure to $PM_{2.5}$ were 25.82, 38.94, 17.73, 22.32, and 31.14%, respectively. Obviously, for the whole PAFs due to the inhalation exposure to $PM_{2.5}$, the stroke (cardiovascular disease) contributed to 29% among the various diseases. According to the mortalities for different diseases from the report of Global Burden of Disease (GBD) (GBD Mortality Causes of Death Collaborators 2015), the total mortalities for IHD, stroke, COPD, LC, and ALRI in 2015 were 24,359, 53,921, 37,610, 2961, and 9997, respectively. Thus, in Zhejiang province, the premature mortalities/EDs in 2015 for IHD, stroke, COPD, LC, and ALRI due to the exposure to $PM_{2.5}$ were 6289, 20,995, 6668, 5125, and 3113, respectively, and the total ED due to ambient $PM_{2.5}$ pollution was 42,191, only contributing 3% of the total ED for exposure to $PM_{2.5}$ in China (1367000) (Liu et al. 2016). Considering the population in Zhejiang province (accounting for about 4% of the total population in China), it can be seen that the risk from exposure to $PM_{2.5}$ for Zhejiang residents was lower than the average level for the whole country—China.

Though most studies used this method to assess to $PM_{2.5}$ exposure risks (Lelieveld et al. 2015; Liu et al. 2016; Feng et al. 2017; Song et al. 2017), it still should be considered a

rough prediction for the health outcomes from the long-term exposure to $PM_{2.5}$, seeing large variations or uncertainties owing to the distributions of the parameters in this function. In addition, the limited epidemiological studies that were involved in China are expected to induce some bias when calculating the risk. Therefore, the future work based on local epidemiological studies, such as some cohort studies, would be more helpful for the accurate prediction or understanding for the adverse health impact of $PM_{2.5}$ and supporting the principle of making air quality guidelines. Moreover, not like $PM_{2.5}$, the health outcomes from the long-term exposure to NO_2 and SO_2 were not taken into account in this study, because no acceptable dose-response model has been developed for these two kinds of gas pollutants. It is expected to be studied in the near future to obtain a comprehensive understanding on the health risks from different ambient air pollutants.

Conclusions

In this study, hourly concentrations of $PM_{2.5}$, PM_{10} , NO_2 , and SO_2 were measured in 13 sites in Zhejiang province, located

in three typical cities (Hangzhou, Shaoxing, and Taizhou) of southeast China lasting for 2 years. In addition, the meteorological conditions, including temperature, wind speed, and relative humidity, were also measured hourly simultaneously. Significantly higher contamination levels were found during winter compared with summer. It was found that the concentrations of particles and gas pollutants in Zhejiang province were much lower than those in northern China, and the ambient air pollution in industry city was much severer than mountain city or natural reserve areas, which is consistent with the emission inventory results.

Based on the hourly data, though the trends of the pollutants were close with each other, gas pollutants SO₂ and NO₂ had a lag time of 1 h to PM_{2.5}. Furthermore, compared with meteorological conditions, it was found that the concentration of PM_{2.5} delayed for 1 h after the influence from temperature and relative humidity. There was no lag time was found between PM_{2.5} and wind speed. Significantly positive linear correlation was verified among the different pollutants, and temperature was negatively linearly correlated with all the pollutants. The concentrations of SO₂ and PM_{2.5} were negatively correlated with both relative humidity and wind speed. However, positive linear correlation was found between NO₂ and wind speed. Thus, it was noted that ambient air pollution was affected by meteorological conditions other than emission. From the 24-h temporal trends, it revealed that the concentrations of PM_{2.5}, NO₂, and SO₂ were lowest in the afternoon (from 15:00–18:00) during winter, and much more stable during summer. For PM_{2.5} and SO₂, the highest levels were found before noon. However, for NO₂, the concentration reached the peak during nighttime, which was attributable to the influence of relative humidity.

In addition, according to the concentrations of PM_{2.5} and IER model, RRs, PAFs, and EDs were calculated for the evaluation on health impact (diseases including IHD, stroke, COPD, LC, and ALRI) from inhalation exposure to PM_{2.5}. The health risks for the residents in Shaoxing (industry city) were highest and lowest for the residents in Taizhou (mountain area or light industry city). Stroke (cardiovascular) dominated the total PAFs/EDs of the different health outcomes. It was found that the health risks in Zhejiang province were lower than average level for the whole country, which was consistent with the results revealed from contamination levels of PMs.

Funding information This work was supported by the National Natural Science Foundation of China (41701584 and 21337005), Natural Science Foundation of Zhejiang Province (LZ15B07000), and China Postdoctoral Science Foundation (2017 M612025).

Compliance with ethical standards

Competing interest The authors declare that they have no competing interest.

References

- Anenberg S, Horowitz L, Tong D, West J (2010) An estimate of the global burden of anthropogenic ozone and fine particulate on premature human mortality using atmospheric modeling. *Environ Health Persp* 118:1189–1195
- Bravo M, Son J, De Freitas C, Gouveia N, Bell M (2016) Air pollution and mortality in São Paulo, Brazil: effects of multiple pollutants and analysis of susceptible populations. *Journal of Exposure Science and Environmental Epidemiology* 26:150–161
- Burnett R, Pope C, Ezzati M, Olives C, Lim S, Mehta S, Shin H, Singh G, Hubbell B, Brauer M, Anderson A, Smith K, Balme J, Bruce N, Kan H, Laden F, Pruess-Ustuen A, Turner M, Gapstur S, Diver W, Cohen A (2014) An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure. *Environ Health Persp* 122:397–403
- Calkins C, Ge C, Wang J, Anderson M, Yang K (2016) Effects of meteorological conditions on sulfur dioxide air pollution in the North China Plain during winters of 2006–2015. *Atmos Environ* 147:296–309
- Chafe Z, Brauer M, Klimont Z, Dingenen R, Mehta S, Rao S, Riahi K, Dentener F, Smith K (2015) Household cooking with solid fuels contributes to ambient PM_{2.5} air pollution and the burden of disease. University of British Columbia
- Chan C, Yao X (2008) Air pollution in mega cities in China. *Atmos Environ* 42:1–42
- Chen Y, Shen G, Huang Y, Zhang Y, Han Y, Wang R, Shen H, Su S, Lin N, Zhu D, Pei L, Zheng X, Wu J, Wang X, Liu W, Wong M, Tao S (2016) Household air pollution and personal exposure risk of polycyclic aromatic hydrocarbons among rural residents in Shanxi, China. *Indoor Air* 26:246–258
- Chen Y, Du W, Shen G, Zhuo S, Zhu X, Shen H, Huang Y, Su S, Lin N, Pei L, Zheng X, Wu J, Duan Y, Wang X, Liu W, Wong M, Tao S (2017a) Household air pollution and personal exposure to nitrated and oxygenated polycyclic aromatics (PAHs) in rural households: influence of household cooking energies. *Indoor Air* 27:169–178
- Chen Y, Zang L, Chen J, Xu D, Yao D, Zhao M (2017b) Characteristics of ambient ozone (O₃) pollution and health risks in Zhejiang Province. *Environ Sci Pollut Res* 24:27436–27444
- Delfino R, Sioutas C, Malik S (2005) Potential role of ultrafine particles in associations between airborne particle mass and cardiovascular health. *Environ Health Persp* 113:934–946
- Dominici F, Peng R, Bell M, Pham L, McDermott A, Zeger S, Samet J (2006) Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases. *Jama* 295:1127–1134
- Englert N (2004) Fine particles and human health—a review of epidemiological studies. *Toxicol Lett* 149:235–242
- Evans J, van Donkelaar A, Martin R, Burnett R, Rainham D, Birkett N, Krewski D (2013) Estimates of global mortality attributable to particulate air pollution using satellite imagery. *Environ Res* 120:33–42
- Feng L, Ye B, Feng H, Ren F, Huang S, Zhang X, Zhang Y, Du Q, Ma L (2017) Spatiotemporal changes in fine particulate matter pollution and the associated mortality burden in China between 2015 and 2016. *Int J Env Res Pub He* 14:1321
- GBD Mortality and Causes of Death Collaborators (2015) Global, regional, and national age–sex specific all-cause and cause-specific mortality for 240 causes of death, 1990–2013: a systematic analysis for the Global Burden of Disease Study 2013. *Lancet* 385:117–171. [https://doi.org/10.1016/S0140-6736\(14\)61682-2](https://doi.org/10.1016/S0140-6736(14)61682-2)
- Han Y, Qi M, Chen Y, Shen H, Liu J, Huang Y, Chen H, Liu W, Wang X, Liu J, Xing B, Tao S (2015) Influences of ambient air PM_{2.5} concentration and meteorological condition on the indoor PM_{2.5} concentrations in a residential apartment in Beijing using a new approach. *Environ Pollut* 205:307–314

- He J, Gong S, Yu Y, Yu L, Wu L, Mao H, Song C, Zhao S, Liu H, Li X, Li R (2017) Air pollution characteristics and their relation to meteorological conditions during 2014–2015 in major Chinese cities. *Environ Pollut* 223:484–496
- Huang R, Zhang Y, Bozzetti C, Ho K, Cao J, Han Y, Dällenbach K, Slowik J, Platt S, Canonaco F, Zotter P, Wolf R, Pieber S, Bruns E, Crippa M, Ciarelli G, Piazzalunga A, Schwikowski M, Abbaszade G, Schnelle-Kreis J, Zimmermann R, An ZS, Szidat S, Baltensperger U, Haddad IE, Prévôt ASH (2014a) High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* 514:218–222
- Huang Y, Shen H, Chen H, Wang R, Zhang Y, Su S, Chen Y, Lin N, Zhuo S, Zhong Q, Wang X, Liu J, Li B, Liu W, Tao S (2014b) Quantification of global primary emissions of PM_{2.5}, PM₁₀, and TSP from combustion and industrial process sources. *Environ Sci Technol* 48:13834–13843
- IEA (International Energy Agency) (2012) IEA World Energy Statistics and Balances. <http://www.oecd-ilibrary.org/statistics> (accessed May 25, 2012)
- Ji D, Li L, Wang Y, Zhang J, Cheng M, Sun Y, Liu Z, Wang L, Tang G, Hu B, Chao N, Wen T, Miao H (2014) The heaviest particulate air-pollution episodes occurred in northern China in January, 2013: insights gained from observation. *Atmos Environ* 92:546–556
- Jiang J, Zhou W, Cheng Z, Wang S, He K, Hao J (2015) Particulate matter distributions in China during a winter period with frequent pollution episodes (January 2013). *Aerosol Air Qual Res* 15:494–503
- Kampa M, Castanas E (2008) Human health effects of air pollution. *Environ Pollut* 151:362–367
- Khaniabadi Y, Goudarzi G, Daryanoosh S, Borgini A, Tittarelli A, De Marco A (2017) Exposure to PM₁₀, NO₂, and O₃ and impacts on human health. *Environ Sci Pollut Res* 24:2781–2789
- Laña I, Del Ser J, Padró A, Vélez M, Casanova-Mateo C (2016) The role of local urban traffic and meteorological conditions in air pollution: a data-based case study in Madrid, Spain. *Atmos Environ* 145:424–438
- Lei Y, Zhang Q, He K, Streets D (2011) Primary anthropogenic aerosol emission trends for China, 1990–2005. *Atmos Chem Phys* 11:931–954
- Lelieveld J, Evans J, Fnais M, Giannadaki D, Pozzer A (2015) The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature* 525:367–371
- Li W, Wang C, Wang H, Chen J, Yuan C, Li T, Wang W, Shen H, Huang Y, Wang R, Wang B, Zhang Y, Chen H, Chen Y, Tang J, Wang X, Liu J, Coveney R Jr, Tao S (2013) Distribution of atmospheric particulate matter (PM) in rural field, rural village and urban areas of northern China. *Environ Pollut* 185:134–140
- Liu J, Han Y, Tang X, Zhu J, Zhu T (2016) Estimating adult mortality attributable to PM_{2.5} exposure in China with assimilated PM_{2.5} concentrations based on a ground monitoring network. *Sci Total Environ* 568:1253–1262
- Meng J, Liu J, Xu Y, Guan D, Liu Z, Huang Y, Tao S (2016) In globalization and pollution: tele-connecting local primary PM_{2.5} emissions to global consumption. *Proc R Soc A*. doi:<https://doi.org/10.1098/rspa.2016.0380>
- Miller K, Siscovick D, Sheppard L, Shepherd K, Sullivan J, Anderson G, Kaufman J (2007) Long-term exposure to air pollution and incidence of cardiovascular events in women. *N Engl J Med* 356:447–458
- Ministry of Environmental Protection of P.R.C, General Administration of Quality Supervision. 2012. Ambient air quality standards. Beijing. GB 3095–2012
- Pope C, Dockery D (2006) Health effects of fine particulate air pollution: lines that connect. *J Air Waste Manag Assoc* 56:709–742
- Smith K, Bruce N, Balakrishnan K, Adair-Rohani H, Balmes J, Chafe Z, Dherani M, Hosgood H, Mehta S, Pope D, Rehfuess E, Grp H (2014) Millions of dead: how do we know and what does it mean? Methods used in the comparative risk assessment of household air pollution. *Annu Rev Public Health* 35:185–206
- Song C, He J, Wu L, Jin T, Chen X, Li R, Ren P, Zhang L, Mao H (2017) Health burden attributable to ambient PM_{2.5} in China. *Environ Pollut* 223:575–586
- Tai A, Mickley L, Jacob D (2010) Correlations between fine particulate matter (PM_{2.5}) and meteorological variables in the United States: implications for the sensitivity of PM_{2.5} to climate change. *Atmos Environ* 44:3976–3984
- Tang L, Yu H, Ding A, Zhang Y, Qin W, Wang Z, Chen W, Hua Y, Yang X (2016) Regional contribution to PM₁ pollution during winter haze in Yangtze River Delta, China. *Sci Total Environ* 541:161–166
- USEPA (U.S. Environmental Protection Agency) (2008) Criteria Pollutants Database <http://www.epa.gov/airexplorer/>
- Wang R, Tao S, Wang W, Liu J, Shen H, Shen H, Wang B, Liu X, Li W, Huang Y, Zhang Y, Lu Y, Chen H, Chen Y, Wang C, Zhu D, Wang X, Li B, Liu W, Ma J (2012) Black carbon emissions in China from 1949 to 2050. *Environ Sci Technol* 46:7595–7603
- WHO (World Health Organization) (2015) WHO air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide. Global update 2005. http://whqlibdoc.who.int/hq/2006/WHO_SDE_PHE_OEH_06.02_eng.pdf
- Xu W, Zhao C, Ran L, Deng Z, Liu P, Ma N, Lin W, Xu X, Yan P, He X, Yu J, Liang W, Chen L (2011) Characteristics of pollutants and their correlation to meteorological conditions at a suburban site in North China plain. *Atmos Chem Phys* 11:4353–4369
- Yang X, Wang S, Zhang W, Yu J (2017) Are the temporal variation and spatial variation of ambient SO₂ concentrations determined by different factors? *J Clean Prod* 167:824–836
- Zhang J, Smith K (2007) Household air pollution from coal and biomass fuels in China: measurements, health impacts, and interventions. *Environ Health Perspect* 115(6):848–855
- Zhang Y, Cao F (2015) Fine particulate matter (PM_{2.5}) in China at a city level. *Sci Rep* 5:14884
- Zhang X, Wang Y, Niu T, Zhang X, Gong S, Zhang Y, Sun J (2012) Atmospheric aerosol compositions in China: spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols. *Atmos Chem Phys* 12:779–799
- Zhou B, Shen H, Huang Y, Li W, Chen H, Zhang Y, Su S, Chen Y, Lin N, Zhuo S (2015a) Daily variations of size-segregated ambient particulate matter in Beijing. *Environ Pollut* 197:36–42
- Zhou Y, Cheng S, Chen D, Lang J, Wang G, Xu T, Wang X, Yao S (2015b) Temporal and spatial characteristics of ambient air quality in Beijing, China. *Aerosol Air Qual Res* 15:1868–1880