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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

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ABSTRACT

$PM_{2.5}$ concentrations in a typical residential apartment in Beijing and immediately outside of the building were measured simultaneously during heating and non-heating periods. The objective was to quantitatively explore the relationship between indoor and outdoor $PM_{2.5}$ concentrations. A statistical method for predicting indoor $PM_{2.5}$ concentrations was proposed. Ambient $PM_{2.5}$ concentrations were strongly affected by meteorological conditions, especially wind directions. A bimodal distribution was identified during the heating season due to the frequent and rapid transition between severe pollution events and clean days. Indoor $PM_{2.5}$ concentrations were significantly correlated with outdoor $PM_{2.5}$ concentrations but with 1–2 h delay, and the differences can be explained by ambient meteorological features, such as temperature, humidity, and wind direction. These results indicate the potential to incorporate indoor exposure features to the regional air quality model framework and to more accurately estimate the epidemiological relationship between human mortality and air pollution exposure.

1. Introduction

Among various air pollutants, fine particulate matter (PM) is of the greatest concern in China in terms of human health impact (He et al., 2001; Chan and Yao, 2008). PM in ambient air is either emitted primarily from various sources or formed secondarily from aerosol precursors in the atmosphere (Huang et al., 2010; Robinson et al., 2007). Beijing is among the most polluted cities in China, and the annual mean concentration of $PM_{2.5}$ from all the monitoring sites in 2013 was as high as 89.5 $\mu g/m^3$ (Huang et al., 2010; Beijing Municipal Environmental Protection Bureau, 2014).

The concentration of ambient $PM_{2.5}$ in Beijing varies widely due to variations in the emission and meteorological conditions (Huang et al., 2010; Wang et al., 2005). It has been suggested that one third of $PM_{2.5}$ at an urban background site in Beijing was from sources outside of Beijing, particularly the highly contaminated regions to south and east (Streets et al., 2007). Based on the results of daily measured size-segregated PM in Beijing in 2012, wind direction is the most important factor affecting PM concentrations in this area (Zhou et al., 2015). Severe pollution events were often associated with southeasterly winds, whereas westerly and northwesterly winds usually brought in relatively clean air mass (Zhou et al., 2015).

Robust associations between inhalation exposure to ambient PM and some respiratory diseases, cardiovascular diseases, and cancer have been established (Pope et al., 2002, 2006), especially true for fine particles such as $PM_{2.5}$ (Brook et al., 2010; Dockery et al., 1993). According to the latest estimation, the population-attributable fraction of the total mortality from ambient air PM pollution in China was as high as 16.2%, ranking fourth among the 67 major factors leading to death (World Health Organization, 2015). Inhalation is the dominant pathway of exposure to $PM_{2.5}$. According to a report that was issued last year, Chinese urban residents (adults) spend approximately 21 h indoor on average (Duan, 2013). Although the monitoring of ambient air pollutants, including $PM_{2.5}$, is a routine practice in most Chinese cities; it is hard to find data for indoor air quality, resulting in technical difficulty in exposure and risk analysis. Moreover, outdoor measurements are almost exclusively used in epidemiological studies to

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establish a relationship between exposure and health effects (Hänninen et al., 2011). The indoor PM concentrations indoors were generally lower than but significantly correlated with the ambient concentrations (Zhou, 2014). Chen and Zhao (2011) have reviewed studies on relationship between indoor and outdoor particles and found that indoor-to-outdoor ratio (I/O) very extensively and penetration factor is the most relevant parameters the penetration mechanism. They also noted the influences of thermophoresis force and airflow (Chen and Zhao, 2011).

The $PM_{2.5}$ concentrations in indoor air are either from the emissions of indoor sources or from the infiltration of ambient air into the indoor environment. Cooking, smoking, and human disturbance are the major indoor sources (Abt et al., 2000; Chao and Cheng, 2002). Infiltration contribution depends on the ambient air concentrations, penetration efficiency, deposition rates, and air exchange rates between the indoor and outdoor environment (Hänninen et al., 2011). Chithra and Nagendra (2014) have investigated the effect of meteorological parameters on indoor coarse and fine particles in a naturally ventilated classroom and found that the indoor air quality is affected significantly by outdoor meteorological parameters.

I/O of a pollutant is a simple indicator for describing the relationship between indoor and outdoor concentrations of the pollutant and is often derived from a series of field measurements (Liu et al., 2001, 2004). When all of the pollutants in the indoor environment are from outside, the indoor-to-outdoor ratio is equivalent to the infiltration factor, which is defined as the equilibrium fraction of ambient particles that penetrate indoors and remain suspended (Hänninen et al., 2011). To derive the infiltration factor, the indoor and outdoor air exchange rate, penetration efficiency of the target pollutant, and deposition rate need to be quantified (Hänninen et al., 2011). The air exchange rate depends mostly on the ventilation characteristics (Long et al., 2001). Strong seasonality was observed with the highest infiltration rates of $PM_{2.5}$ in summer and the lowest ones in winter (Hänninen et al., 2011). An important reason for the high variation in infiltration rates is the difference in building construction, which depends on both physical and socio-economic factors (Hänninen et al., 2011). In this study, the indoor and ambient air $PM_{2.5}$ concentrations were measured simultaneously for a typical residential apartment in Beijing during both heating and non-heating periods. The relationship between the indoor and ambient air $PM_{2.5}$ concentrations and the influence of meteorological conditions on the relationship were addressed. With the limited data from a single site, we did not attempt to develop a predicting model at this stage. Instead, key factors controlling the indoor air $PM_{2.5}$ concentrations were identified, and a statistical method was developed and tested to simulate the indoor $PM_{2.5}$ concentrations in this particular case. There was no solid fuel use in the studied household. Further improvement based on large datasets from the field is necessary before a predicting model can be practically operational.

2. Methods and materials

2.1. Study site

A three-bedroom apartment at the 15th floor of a 20-story apartment building was chosen for this study. There are six apartments on each floor, and the one studied is located at the northeast corner of the floor with most of the windows facing north and east. There are other tall buildings nearby except for to the north, in which direction is a university campus with only 6-floor buildings and many bungalows near the site (Fig. S1). The residents living in these bungalows rely on coal for heating in winter. A busy street (Chengfu Rd.) is 140 m away to the south. Only two non-smoking residents live in this apartment with limited activities. One simple meal was cooked each day using either pipelined gas or electricity. Only some windows were opened during the non-heating period. With a high-efficiency ventilator that was operated in the kitchen, the main emission of $PM_{2.5}$ within the apartment was human disturbance.

2.2. On-line measurement

Two sets of SidePak (model AM510, TSI, NM, USA) personal aerosol monitors that were coupled with $PM_{2.5}$ impactors were used to measure the $PM_{2.5}$ concentrations in indoor and outdoor (immediately outside of a window facing north that was not opened during the study) air simultaneously. The measurements were conducted at 5-min intervals for both heating (from Dec. 27, 2013, to Jan. 20, 2014) and non-heating (from March 28 to May 2, 2014) periods.

2.3. Calibration

As a particle counter, the monitors cannot report the true $PM_{2.5}$ concentrations without systematic error (Liu et al., 2001). Therefore, the measurements were calibrated against the data that were reported by the Beijing Municipal Government Monitoring Center for the area in order to report more realistic concentration data. Even with the calibration, relatively high uncertainty is expected in the concentrations reported in this study because the measurements derived in this study were different from those reported by the monitoring center in terms of location and time. Fortunately, this study focused on relationship between indoor and outdoor. If there was any systematical error, the error occur synchronously for both indoor and outdoor measurements, resulting limited influence on their relationship. Because only hourly average $PM_{2.5}$ concentrations were available from the center, the 5-min measurements were pooled to generate hourly means to match the officially reported data. The comparison for both study periods is shown in Fig. S2. Although slight differences were found between the two periods, a unified model can fit the data reasonably well, with 71% of the variation explained. All of the directly measured $PM_{2.5}$ concentrations were calibrated based on the model.

2.4. Meteorological conditions

The meteorological parameters were collected using a weather station (DT80, Rainroot, China) that was set up on top of an office building that is approximately 900 m away from the study site. The parameters that were recorded every 5 min included temperature, relative humidity, wind direction, wind speed and precipitation. The hourly and daily averages were derived for various purposes. The wind speeds were broken into eight direction components: north (WS_N), northeast (WS_{NE}), east (WS_E), southeast (WS_{SE}), south (WS_S), southwest (WS_{SW}), west (WS_W), and northwest (WS_{NW}). There was no precipitation during the study period. The indoor and outdoor temperatures were measured on-line using thermometers WHOT1 (Wangyunshan, China) at 5-min intervals. The means and standard deviations of outdoor temperature, relative humidity, and wind speed were 2.81 ± 3.66 °C, 36.9 ± 17.4%, and 0.52 ± 0.80 m/s, respectively.

2.5. Statistical analysis

STATISTICA (StatSoft, USA) was used for significant testing at a significance level of 0.05. A bimodal distribution was tested and fitted using PeakFit v.4.12 (Rundel, 1991). Paired or non-paired two sample t-test were used to compare means, coefficient of variation,

and slop. Pearson correlation analysis was applied to test correlation.

3. Results and discussion

3.1. $PM_{2.5}$ in ambient and indoor air

Based on the calibrated 5-min data, the mean and standard deviation of the measured $PM_{2.5}$ concentrations in ambient air during the study period were 84 ± 59 μg/m^3. The mean values for the heating (82 ± 67 μg/m^3) and non-heating (86 ± 53 μg/m^3) periods were not significant ($p > 0.05$), whereas the coefficients of variation were significant between the heating (82%) and non-heating (62%) ($p < 0.05$) periods, indicating a relatively large fluctuation in the former. In Beijing, the winter features both heavy emission due to heating demand (Wang et al., 2014) and a large meteorological influence of strong northwest wind as the passage of cold fronts, which often bring in clean air from the highland of the Inner-Mongolia plateau and Siberia (Zhou et al., 2015; Song et al., 2006; Zhu et al., 2011). In fact, both the minimum (10 μg/m^3) and maximum (396 μg/m^3) values occurred during the first period, whereas the range of the second period was only 16–273 μg/m^3. For the simultaneously measured indoor $PM_{2.5}$ concentrations, the mean concentrations during both the heating and non-heating periods were lower than those in ambient air. The means and standard deviations were 59 ± 33 and 55 ± 30 mg/m^3, which were 22% and 36% lower than the values in ambient air, respectively. Additionally, the maximum $PM_{2.5}$ concentration in indoor air (253 μg/m^3) appeared during the heating period, corresponding to the heaviest pollution event in ambient air.

It is generally accepted that $PM_{2.5}$ concentrations that are measured in ambient air are log-normally distributed (World Health Organization, 2015). In this study, however, they were neither normally nor log-normally distributed based on skewness and kurtosis t-tests ($p > 0.05$ for all tests). Fig. 1 shows histograms at the log-scale for both ambient and indoor $PM_{2.5}$ during the two study periods.

During the heating season, a typical bimodal distribution can be seen for both the ambient and indoor air $PM_{2.5}$ concentrations. The two populations for ambient air $PM_{2.5}$ correspond to heavily contaminated and relatively clean times with a fast transition in between. The geometric means and standard deviations of the two populations for ambient air were 1.36 ± 0.14 and 2.03 ± 0.20 μg/m^3, respectively, indicating almost one order of magnitude difference between them. Correspondingly, the geometric means and standard deviations of the two clearly distinguished populations for the indoor $PM_{2.5}$ concentrations were 1.54 ± 0.07 and 1.89 ± 0.14 μg/m^3, respectively. For the non-heating period, however, the histogram appeared to be plat kurtosis, with coefficients of kurtosis that were significantly less than 0 ($p > 0$) (could also be two populations with less difference compared with the heating period). Compared with the heating period, both the emission and dispersion effects of the northwest wind during the non-heating period were weaker. The air quality in Beijing features a frequent interchange of severely polluted episodes and relatively clean days.

3.2. Influence of meteorological conditions on the ambient air $PM_{2.5}$ concentrations

Immediately next to Hebei (the province with extremely high emissions of various air pollutants) and Tianjin in the south and east and surrounded by the Yanshan Mountains to the north and west, the ambient air quality of Beijing is strongly affected by the wind direction and windspeed (Streets et al., 2007; Zhou et al., 2015). Based on daily observations over a one-year period and a multiple regression model, it was found that heavy ambient air pollution is often associated with southeasterly winds, which bring in various pollutants from Hebei, whereas air masses from the west and northwest are usually clean (Zhou et al., 2015). It was also found that PM concentrations in ambient air are also affected by other meteorological parameters, including temperature, humidity, and precipitation (Zhou et al., 2015). According to the ambient air $PM_{2.5}$ concentrations and the meteorological conditions that were observed in this study, it was revealed that the $PM_{2.5}$ concentration was positively correlated with the relative humidity and negatively correlated with the wind speed during both the heating and non-heating periods ($p < 0.05$) (Fig. S3). It was well accepted that high humidity is favorable for the formation of secondary aerosols (Wang et al., 2012b; Zhang et al., 2012) and that strong wind may facilitate the dispersion of the pollutants in the air. For temperature, however, a negative correlation between $PM_{2.5}$ concentration and temperature was found ($p < 0$) for the heating period, during which more fuels were combusted on colder days. On the other hand, temperature was significantly ($p < 0$) positively correlated with the $PM_{2.5}$ concentrations during the non-heating season.

In this study, the wind directions were also broken into eight components, and the $PM_{2.5}$ concentrations were correspondingly classified into eight groups according to the dominant wind directions during the measurements. The calculated means and standard deviations of the $PM_{2.5}$ concentrations of the eight

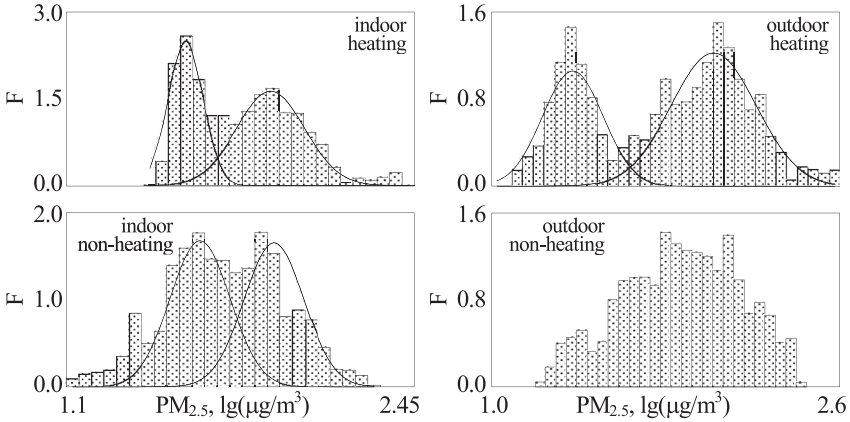

Fig. 1. Frequency distributions of the indoor (left panels) and outdoor (right panels) $PM_{2.5}$ concentrations during the heating (top panels) and non-heating (bottom panels) periods. The histograms were fitted with a bimodal distribution except for ambient air during the non-heating period.

direction groups varied from 40 ± 40 μg/m^3 (NW) to 94 ± 73 μg/m^3 (W) during the heating period and from 57 ± 35 μg/m^3 (N) to 96 ± 59 μg/m^3 (NE) during the non-heating period. The results of every direction are shown in Fig. S4. For the non-heating period, the concentrations were relatively high when the winds were mainly from the east and south and relatively low when the west and north winds dominated, which is similar to those reported previously (Zhou et al., 2015). However, different results were observed during the heating period, during which the highest mean $PM_{2.5}$ concentration occurred when the wind was from the west, whereas the major emission sources are distributed in Hebei, Shandong, and Tianjin to the east and south. One likely explanation is the influence of local emission from bungalows immediately to the west and northwest of the site (Fig. S1). Emissions from coal burning can affect the site when the wind is from the west.

3.3. Temporal trends of $PM_{2.5}$ in indoor and outdoor air

Fig. 2 shows the time series of directly measured $PM_{2.5}$ concentrations in both ambient and indoor air at 5-min intervals during both heating and non-heating periods. Strong and synchronized variations occurred in both the indoor and ambient environment during both periods. For 87% of the measurements, the indoor air $PM_{2.5}$ concentrations were lower than those in outdoor air, whereas the remaining indoor air $PM_{2.5}$ concentrations were higher than the outdoor ones, when the ambient $PM_{2.5}$ concentrations were relatively low. Relatively high concentrations of $PM_{2.5}$ in indoor air of a classroom in comparison with outdoor air during day time was also reported (Chithra and Nagendra, 2014).

It is interesting to note that $PM_{2.5}$ concentrations tend to be higher at night (18:00–5:00, roughly times for sunset and sunrise for the two periods) than during the day (5:00–18:00), and such differences are particularly true during the heating period (Fig. 3). Statistically, the means and standard deviations of the ambient air $PM_{2.5}$ concentrations were 107 ± 85 μg/m^3 during the daytime and 219 ± 195 μg/m^3 at night, with average daily ratios (night/day time) of 2.2 during the heating period. During the non-heating period, the values were 94 ± 48 μg/m^3 at night and 48 ± 67 μg/m^3 during the daytime, with an average daily ratio (night/day time) of 1.6. During both periods, the differences between the daytime and night are significant ($p < 0.05$). Such differences are further exploited by calculating multi-day average hourly $PM_{2.5}$ concentrations for the non-heating and heating periods in Fig. 2. During the heating period, the ambient air $PM_{2.5}$ concentrations began to increase in the early afternoon, peaked at approximately 20:00, and did not begin to decrease until the middle of the night. The main reason could be the shift in the wind direction between day and night. With the increased wind at this site (Fig. 3, day and night patterns of wind directions are significant at $p < 0.05$ based on a χ^2-test), the north-northwest wind from the mountain area was stronger during the daytime, whereas south-southeast-southwest wind was dominant at night. As previously discussed, the north-northwest winds usually bring in clean air mass. Although a similar daily variation pattern was found for indoor air, the difference between the indoor and outdoor was larger at night than during the daytime, which could be partly due to the indoor contribution of human disturbance. It was reported in the literature that the levels of the indoor air $PM_{2.5}$ in a classroom are strongly affected by activities of occupants (Chithra and Nagendra, 2014).

For the most severe pollution events, $PM_{2.5}$ in ambient air often took days to build up but decreased quickly within hours. Three pollution events continuously occurred from 15:31 on Jan. 1 to 12:51 on Jan. 7, 2014, and three separately occurring episodes (from 13:10 on April 10 to 19:50 on April 15, 2014; from 15:20 on April 22 to 2:20 on April 26, 2014; and from 11:50 on April 27 to 6:30 on May 2, 2014) are shown in Fig. 4 as typical examples. In addition to the $PM_{2.5}$ concentrations, the north and northwest wind components are also shown. When the wind from the north and northwest was weak, $PM_{2.5}$ and its precursors from either local sources or neighboring provinces had accumulated. The buildup processes were not terminated until the meteorological condition suddenly changed with strong and frequent north or northwest winds, which brought clean air mass to the area. The sudden shift in wind direction led to the rapid disappearance of air pollutants, often from hundreds to tens μg/m^3 within hours. The overall gradual accumulation and rapid cleanup processes were also quantified by calculating the number of increasing and decreasing "runs", which were defined as a segment of measurements with a continuously increasing or decreasing trend. During the heating period, there were 1566 increasing runs with an average concentration difference of 3.91 μg/m^3. In comparison, there were 1463 decreasing runs with an average difference of 4.21 μg/m^3. Similarly, the increasing and decreasing runs during non-heating period were 2314 and 1969, with average differences of 2.96 and 3.50 μg/m^3, respectively. Fewer runs and a larger distance suggest sharper changes.

3.4. Influence of ambient air $PM_{2.5}$ on indoor air $PM_{2.5}$

The air qualities indoors and outdoors are closely related through air exchange and the penetration of air pollutants from the outdoor to indoor environment or vice versa (Viana et al., 2011). In this study, no indoor emission source was identified, except for

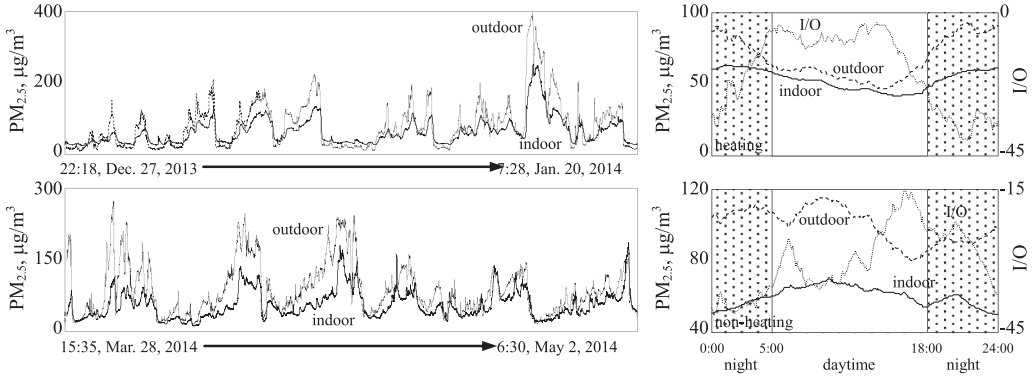

Fig. 2. Relationship of $PM_{2.5}$ concentrations between the ambient air and delayed indoor air during the heating (top-left panel) and non-heating (bottom-left panel) periods based on 5-min-interval measurements. Time trends of 5-min data are shown in left panels, while the average daily variations of the indoor and outdoor $PM_{2.5}$ concentrations as well as the I/O ratios are shown in the right panels.

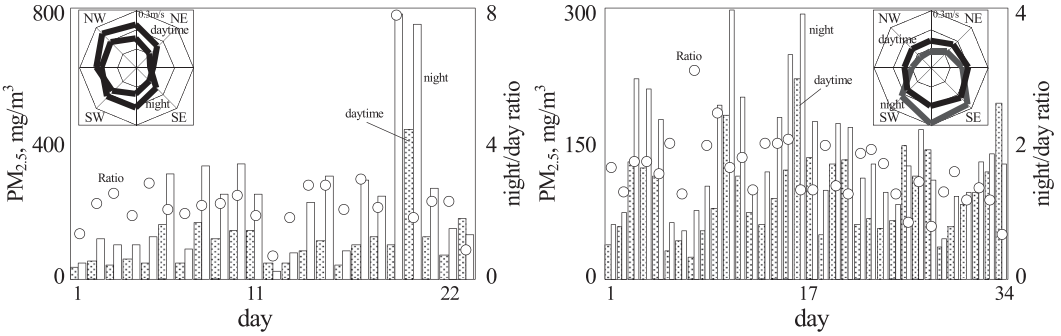

Fig. 3. Comparison between the daytime and nighttime average $PM_{2.5}$ concentrations in ambient air for individual days during the heating (left panel) and non-heating (right panel) seasons. The concentration ratios of nighttime/daytime are also shown. Inserted are the daytime and nighttime wind increases during the heating (left) and non-heating (right) periods, showing a typical mount-valley wind pattern at the site.

disturbance caused by the daily activity of the residents. Therefore, penetration is the dominant process of the exchange. The correlations between log-transformed indoor and ambient $PM_{2.5}$ concentrations are shown in Fig. S5 for the two periods. Significantly positive correlations ($p < 0.05$) were revealed, and the coefficients of correlation were 0.911 and 0.848 during the heating and non-heating seasons, respectively. By measuring the size-segregated ambient and indoor air PM in Beijing, a significantly linear relationship was found between the indoor and ambient measurements for a number of size fractions, including $PM_{2.5}$ (Zhou et al., 2015). Because indoor air $PM_{2.5}$ is mainly from ambient air, it is expected that there should be a lag time. Such a delay is demonstrated by a "delayed correlation" method here (to repeatedly calculate the correlation coefficients between the indoor and ambient $PM_{2.5}$ by displacing the indoor air $PM_{2.5}$ data points backwards in 5-min intervals). The results based on the log-transformed $PM_{2.5}$ concentrations are shown in Fig. 5 for both the heating and non-heating periods. It appears that when the indoor air $PM_{2.5}$ concentrations were displaced backwards, the coefficients of correlation increased first and then decreased after the peak values were reached. During the heating period, the original coefficient of correlation was 0.911 and peaked at 0.949 (0.92 and 0.96 for data without log-transformation) at 75 min backward displacement. Similarly, during the non-heating period, the original coefficient of correlation increased from 0.848 without displacement to a peak value of 0.902 with a 115-min displacement during the non-heating period (079 and 0.85 for data without log-transformation). The lag time for the heating period was slightly shorter than that of the non-heating period, likely due to heavier ambient air pollution and a larger temperature difference between indoor and outdoor air (also see the discussion on the unified model) in the former.

Although the delay occurred generally over the entire study period, the phenomenon was mostly obvious during severe air pollution episodes. Three such examples are shown in Fig. S6 as 1) from 17:56 on Jan. 1 to 13:41 on Jan. 3, 2014; 2) from 14:16 on Jan. 15 to 18:11 on Jan. 17, 2014; and 3) from 19:00 on April 12 to 21:25 on April 15, 2014. Clearly, when the $PM_{2.5}$ concentrations increased during the first period of the cycles, the indoor air $PM_{2.5}$ concentrations responded more or less slowly and peaked later than the ambient air concentrations. For example, the occurrences of the three peak $PM_{2.5}$ concentrations in indoor air were 100, 40, and 15 min later than the corresponding peaks in ambient air during the first period. Similarly, the time to reach the two valleys of the indoor air $PM_{2.5}$ concentrations were 55 and 35 min behind those in ambient air for this period. The lag times that were identified by the displacement correlation are average values with large variations.

The relationship between the indoor and outdoor concentrations of a given pollutant can be quantified by field observation

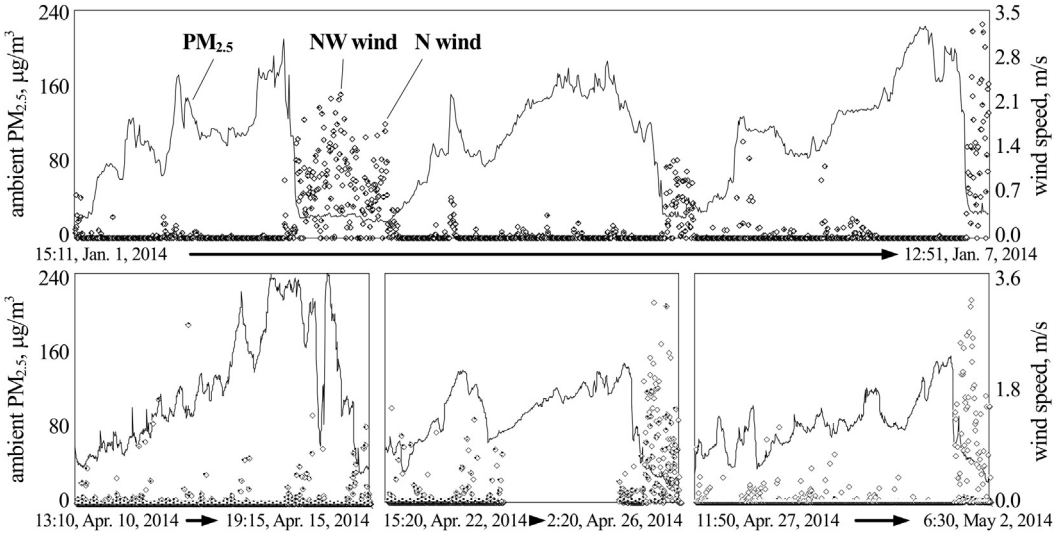

Fig. 4. Typical pollution episodes during the two periods, showing the strong influence of the northwest and west winds on the accumulation and decrease of $PM_{2.5}$.

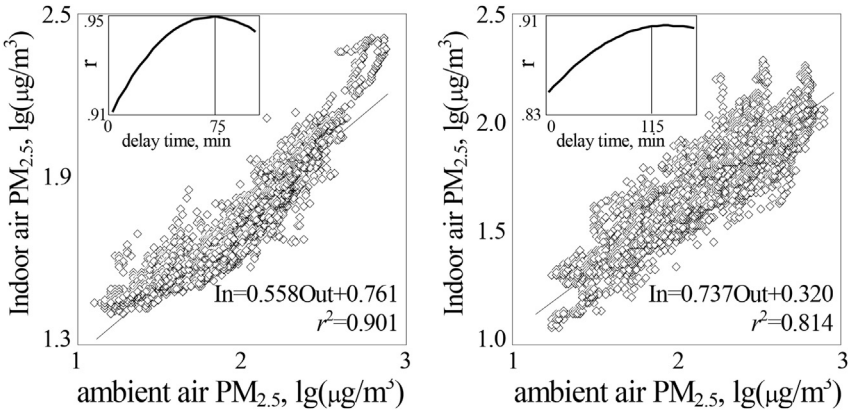

Fig. 5. Relationship of $PM_{2.5}$ concentrations between ambient air and delayed indoor air for the heating (left panel) and non-heating (right panel) periods based on 5-min-interval measurement. The optimal lag times were derived by "delayed correlation" and are shown in the inserted curves.

based I/O (Liu et al., 2004; Pekey et al., 2010). Fig. S7 shows a non-linear dependence of I/O on the ambient air $PM_{2.5}$ concentrations. For most of the data, the indoor air $PM_{2.5}$ concentrations were lower than the outdoor concentrations when relatively high concentrations occur outdoors, resulting relatively low I/O values (<1). This result occurs because ambient air is the major source of $PM_{2.5}$ in the indoor environment without an obvious indoor source except for human disturbance, and $PM_{2.5}$ in the indoor environment can be removed by deposition or adsorption (Riley et al., 2002; Yoon et al., 2009). On the other hand, the I/O values were greater than 1 when the ambient air $PM_{2.5}$ concentrations were relatively low, and the penetration of $PM_{2.5}$ from outdoor air was overruled by the contribution of the disturbance of residents in the apartment.

3.5. Simulation modeling

The significant correlation between the indoor and outdoor concentrations suggests that it is possible to simulate indoor air $PM_{2.5}$ concentrations based on ambient measurements with reasonable uncertainty. The quantitative relationship that was revealed here may guide future modeling efforts on a regional scale. The linear regression models based on log-transformed data and the optimum lag times derived from the correlation analysis are shown in Fig. 5 for the heating and non-heating periods (5-min intervals). The majority of the variations (81–90%) in indoor air $PM_{2.5}$ concentrations can be explained well by the ambient air $PM_{2.5}$ concentrations, and the differences between the two periods are obvious. The slopes of the two models were significantly different, at 0.558 and 0.737 for the heating and non-heating periods, respectively. With a higher slope, a correspondingly stronger influence of ambient air quality on indoor air was found during the non-heating period, when more windows were opened, leading to stronger natural ventilation. Meantime, the model interception during the heating period (0.761) was higher than that during the non-heating period (0.320), suggesting higher background $PM_{2.5}$ concentrations in indoor air in the former. Such a difference can likely be explained by the fact that heating radiators that were operated during the heating period facilitate air circulation in the indoor environment, creating stronger disturbance.

The daily average concentrations are often reported by government agencies and are readily available for risk analysis. In Fig. S8, similar results were obtained based on the daily average $PM_{2.5}$ concentrations for the two periods. With the 5-min data that were polled into daily concentrations, the regression parameters differ from those based on the 5-min data. No delay was found at the daily scale, and the slopes were approximately doubled because detailed fluctuations within days were smoothed, whereas such a smoothing effect was more effective on ambient air $PM_{2.5}$ concentrations.

For the purpose of prediction, a unified model instead of two separated ones for the two periods would be more meaningful. Therefore, a single regression model for both periods was developed with a coefficient of determination of 0.768, lower than the two individual ones (0.901 and 0.814), and a lag time of 90 min between those of the two individual models (75 and 115 min). One major difference between the two periods was temperature. The outdoor temperatures of the study site varied from −6 to 14 °C during the heating period and from 12 to 29 °C during the non-heating periods. The influence of temperature was explored by plotting the residues of the unified regression model (the indoor air $PM_{2.5}$ as a function of the ambient air $PM_{2.5}$ concentrations) against temperature in Fig. S9. A small but significantly ($p < 0.05$) positive slope was identified, suggesting an influence of temperature on the relationship between the indoor and ambient air $PM_{2.5}$ concentrations. Because the indoor temperatures of the two periods were relatively constant and similar to one another (18.9 ± 1.0 and 18.2 ± 4.8 °C for the heating and non-heating periods, respectively), the differences in the indoor–outdoor temperature between the two periods were similar to ambient temperatures differences between the two periods. The positive slope suggests that the indoor–outdoor temperature differences could be a driving force for air exchange and PM penetration and that large gradients are favorable for the penetration of $PM_{2.5}$ from an ambient to indoor environment (Hänninen et al., 2011).

Besides temperature, humidity also affects the relationship between the indoor and outdoor air $PM_{2.5}$ concentrations. As shown in Fig. S10, the residues of the unified regression model residue were negatively correlated with humidity. Such a relationship is unlikely due to indoor–outdoor exchange or penetration. Instead, the humidity was positively correlated with ambient air $PM_{2.5}$ concentrations, which were positively correlated with regression residues (Wang et al., 2012a; Zhang et al., 2012).

The predictions were also affected by wind direction. Based on the 5-min data, the regression model residues were classified into eight direction components for positive and negative residues and are shown as a rose map in Fig. S11. The positive residues tend to be caused by northwest winds, whereas the negative residues seem associated with southeast winds. Such an effect was primarily due to high ambient $PM_{2.5}$ concentrations that were associated with southeast winds and low concentrations related to northwest winds.

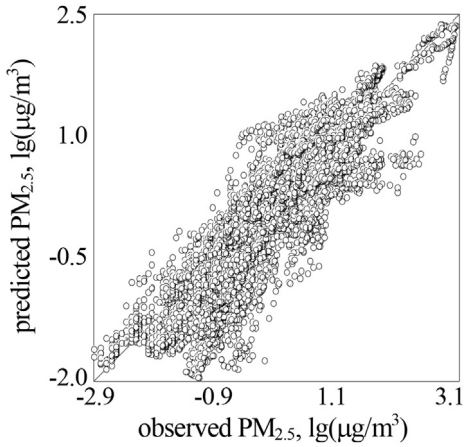

Fig. 6. Comparison between the observed and multi-regression model-predicted $PM_{2.5}$ concentrations in indoor air.

With all of the parameters, including the outdoor $PM_{2.5}$ concentrations, meteorological parameters a forward stepwise multiple regression model was developed. The equation is as follows with a coefficient of determination of 0.809:

$$PM_{2.5}\ (\text{in}) = 0.613\ PM_{2.5}\ (\text{out}-90) - 0.0047\ T + 0.009\ RH + 0.0269\ WS_E + 0.0161\ WS_{NW} + 0.616,$$

where $PM_{2.5}$ (in) and $PM_{2.5}$ (out−90) are the $PM_{2.5}$ concentrations ($\mu g/m^3$) indoors now and outdoors 90 min ago, respectively. T (°C) and RH (%) are temperature and relatively humidity, respectively. WS_E and WS_{NW} are the east and northwest wind speed components, respectively, which are only significant among the eight tested wind components.

Fig. 6 shows the relationship between the predicted model and the field indoor $PM_{2.5}$ concentrations. With more than 80% of the variations explained, the equation can be used to satisfactorily predict the indoor air $PM_{2.5}$ concentrations for the particular apartment. However, the above equation cannot be extrapolated to other sites at this early development stage. What these results provide is not a prediction model itself, but rather, these results demonstrate the possibility of predicting indoor air $PM_{2.5}$ concentrations based on outdoor air values and meteorological parameters and offer a statistical method that can be further tested when additional field data become available. It is also noted that a substantial underestimation of approximately −0.9 (lg ($\mu g/m^3$)) occurred for some data, thus further study is warranted to improve the model (Fig. 6).

Again, this study only provides a potential method that can be used for the future development of a statistical model to predict indoor air $PM_{2.5}$ concentrations on a regional scale. The importance of meteorological parameters, should be addressed in the future work on data collection and model development.

4. Conclusions

Different distribute patterns of indoor and outdoor concentrations between heating and non-heating period was found. A bimodal distribution was identified during the heating season while distinguishable for the non-heating season, suggesting more frequent and rapid transition between severe pollution events and clean days occurred during heating season. Ambient air $PM_{2.5}$ concentration are strongly meteorological condition dependent with wind direction as the dominant influencing factor. Higher $PM_{2.5}$ concentrations were observed at night than during the daytime due to the different wind direction. Strong correlations between indoor and delayed outdoor $PM_{2.5}$ concentrations were found and the lag time is longer for non-heating season. Regression model for indoor $PM_{2.5}$ concentrations reveals positive correlation between temperature and residue while negative for relative humidity.

Conflict of interest

The authors declare no competing financial interest.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2015.04.026.

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