A Comparison Study of Indoor and Outdoor Air Quality in Nanjing, China

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ABSTRACT

Because humans spend much of their time in indoor microenvironments, the air quality indoors has attracted significant attention. This study simultaneously conducted real-time indoor observations and comprehensive outdoor measurements of O₃, CO and PM_{2.5} in suburban Nanjing in eastern China from September to December 2018. The O₃, CO and PM_{2.5} exhibited median indoor concentrations of 3.2 ppb, 395.8 ppb and 17.3 µg m⁻³, respectively, based on measurements in a room with closed windows and relatively regular daily indoor human activity, and median outdoor values of 30.5 ppb, 386.1 ppb and 37.2 µg m⁻³, resulting in median indoor-outdoor (I/O) ratios of 0.14, 1.01 and 0.46. Moreover, the indoor concentrations traced the outdoor variations with correlations of approximately 0.68, 0.82 and 0.82 for the O₃, CO and PM_{2.5}, respectively. During pollution episodes, the indoor O₃ and PM_{2.5} concentrations reached about 30 ppb and 130 µg m⁻³, respectively. Both meteorological conditions (e.g., wind speed or relative humidity) and human activity indoors influenced the relationships between the indoor and outdoor concentrations, including the I/O ratios and time lag. A simplified indoor-outdoor mass-balance equation was developed to simulate the indoor concentrations, and the predictions fitted the observed data for most of the testing period, especially after considering the human activity indoors and limited penetration of particles. This study enhances our understanding of the indoor-outdoor relationships for gaseous and particulate matter concentrations in polluted areas such as the Yangtze River Delta and highlights the urgent need for improving indoor air quality in the megacities of China.

Keywords: Air quality; Indoor measurements; I/O ratio; Yangtze River Delta.

INTRODUCTION

Indoor air quality has been debated and investigated widely in recent years because of the potential detrimental impact of air pollutants on human health (Passarelli, 2009; Choo and Jalaludin, 2015; Wolkoff, 2018). Numerous epidemiological studies have reported that air pollutants were associated with short- and long-term negative impact on health, including higher morbidity of cardiovascular and respiratory diseases (Donaldson *et al.*, 2002; Pope III *et al.*, 2002; Pope III and Dockery, 2006; Sousa *et al.*, 2013), increased mortality (Samet *et al.*, 2000; Bell *et al.*, 2005; Chen *et al.*, 2012), and even adverse birth outcomes (Astrup, 1972; Nieuwenhuijsen *et al.*, 2013). As people spend more time in indoor microenvironments (Spalt *et al.*, 2016), indoor air quality must be concerned owing to its potential risks to human health.

Research on indoor air quality has been conducted in a variety of indoor microenvironments in different countries and regions (Chen and Zhao, 2011; Fadeyi, 2015; Liu and Zhang, 2019). Factors that have significant influences on indoor concentrations have been elucidated, among which outdoor concentration is a predominant factor dominating indoor concentration for most investigated gases and particles (Weschler and Shields, 1997; Abt *et al.*, 2000; Loupa *et al.*, 2006; Walker and Sherman, 2013; Zhong *et al.*, 2013). Other factors include indoor sources and deposits, the building infiltration, natural and mechanical ventilation and recirculation of air etc. (Hayes, 1991; Lunden *et al.*, 2003; Chen and Zhao, 2011; Cros *et al.*, 2012). Presence of indoor sources, such as re-suspension of particles due to walking, emission from smoking and other domestic or office equipment, can impose remarkable effects on the concentrations of indoor air pollutants (Stranger *et al.*, 2007; Fazlzadeh *et al.*, 2015). Surfaces of indoor materials can remove some gases like ozone (Lamble *et al.*, 2011; Gall *et al.*, 2013) and are meanwhile where particulate matter deposits (Liu *et al.*, 2014). Moreover, ventilation rate significantly affects the indoor-outdoor relationships when

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the ventilation system is on (Koponen *et al.*, 2001). Besides factors associated with building structure and indoor layout, outdoor meteorological parameters, such as wind speed, wind direction, temperature and relative humidity, also play critical parts (Riain *et al.*, 2003; Elbayoumi *et al.*, 2013; Chithra and Nagendra, 2014; Zhao *et al.*, 2015). In particular, based on the studies of the influential factors on indoor-outdoor relationships, mass-balance models were established in some research to predict the indoor concentrations of pollutants (Hayes, 1989; Du *et al.*, 2018; Tang *et al.*, 2018).

In China, rapid urbanization and industrialization have brought about serious issues of air pollution (Chan and Yao, 2008; Liu *et al.*, 2015). However, currently the majority of research mainly focuses on outdoor air pollution. Some indoor measurements have been conducted in some cities of China, e.g., Guangzhou, Dalian and Chengdu, focusing on pollutants like PM_{2.5} and formaldehyde (Zhao *et al.*, 2004; Huang *et al.*, 2007; Liu *et al.*, 2018). In Yangtze River Delta of China, some indoor measurements on PM_{2.5} and gases have been carried out, e.g., in Nanjing, Shanghai and Hangzhou (Weng *et al.*, 2010; Zhong *et al.*, 2013; Hu *et al.*, 2015; Wang *et al.*, 2016; Shao *et al.*, 2017; Bi *et al.*, 2018). Zhong *et al.* (2013) investigated the indoor and outdoor CO concentrations in Shanghai and found they were comparable and had good correlation in a single-side natural ventilating room without internal source. Shao *et al.* (2017) and Bi *et al.* (2018) measured indoor and outdoor concentrations of fine particulate matter in Yangtze River Delta and found the averaged indoor PM_{2.5} concentration was higher than the indoor air quality guideline published by the World Health Organization. Additionally, the relationships between indoor and outdoor PM_{2.5} concentrations were found to be different depending on the various indoor microenvironments (Bi *et al.*, 2018). Hu *et al.* (2015) found that indoor acidic and oxidizing gaseous pollutants, i.e., SO₂, NO₂ and O₃, had potential physical and chemical damage to objects indoors. However, many of them collected data merely for short periods (e.g., < 1 month) with relatively coarse time resolution. Compared with the increasing outdoor air pollution measurements, continuous indoor air quality measurements with high time resolution are still significantly fewer. Moreover, the discussions on the relationships between indoor air quality and meteorological conditions are lacking.

To better understand the indoor air quality in the western Yangtze River Delta of China, the measurement was carried out in the room of suburban Nanjing, with relatively good air-tightness, unobvious indoor sources of gases measured, and fairly regular human activities indoors. Continuous monitoring on indoor/outdoor O₃, CO and PM_{2.5} concentrations was conducted for about three months. In this study, we studied the features of indoor and outdoor concentrations of O₃, CO and PM_{2.5} in detail. In particular, to better evaluate the impact of both human activities and meteorological factors, the O₃ and PM_{2.5} pollution episodes were selected and analyzed. Finally, a prediction method was established based on the mass-balance equation. Those findings can provide meaningful references for further research on effects of atmospheric gases and particles on

indoor microenvironments and help the development of methods for the control of indoor air pollutants.

METHODS

The Sampling Site and Indoor Environment

The measurement campaign was carried out at SORPES station (Station for Observing Regional Processes of the Earth System), the location of which is in the campus of Nanjing University (118°57'10"E, 32°07'14"N), suburban part of Nanjing 20 km away from downtown area. The station is located on a 40 m high hill and no tall building surrounds it. The SORPES station is heavily instrumented to monitor the outdoor pollutants from both the North China Plain and Yangtze River Delta. The outdoor meteorological conditions, including temperature, relative humidity, wind speed and direction etc., are observed at SORPES as well. During the measurement, the outdoor temperature and relative humidity were 5.1–28.2°C and 18.6–98.6% (mean: 16.1°C and 68.3%, respectively). Wind speed was 0.0–5.9 m s⁻¹ and prevailing wind direction was northeast (mean: 1.4 m s⁻¹ and 52.5°, respectively). Rainfall occurred occasionally with intensity from 0.1 to 4.8 mm h⁻¹ during the measurements. More details about SORPES station are described in Ding *et al.* (2016). To study the indoor air quality, continuous real-time indoor monitoring of O₃ and CO concentrations was conducted from September to November 2018. Moreover, considering the frequent PM pollution in late autumn and winter in Yangtze River Delta, intermittent but fairly regular real-time sampling of indoor PM_{2.5} mass concentration was conducted from October to December 2018.

The indoor measurements of O₃, CO and PM_{2.5} were conducted in a room of SORPES station, which is mainly used as the space for the long-term placement of measurement instruments. As shown in Fig. 1, the features of the room, such as the building structure and ventilation, are similar with the residential buildings. The room has a floor area of 25 m² with a small attic whose area is 12.5 m². A 1 m² skylight allows sunlight to enter the room through the window glass. All the windows were kept closed during the measurement period, and the door was not open for most of the time. An air conditioner without indoor-outdoor air exchange function was on constantly during the measurement period, which kept the indoor temperature relatively stable. The setting was inclined to get good air-tightness, which was equated to low indoor-outdoor exchange rate. Indoor human activities occurred routinely from 09:00 LT to 12:00 LT and from 15:00 LT to 18:00 LT every day including weekdays and weekends, although occasional exceptions may happen. During the everyday periods of occupancy, the room was generally occupied by 1–3 staff. Smoking was seriously prohibited in the room. There are no obvious O₃ and CO sources inside the room and the re-suspension of particles due to walking and some other activities can be regarded as the only indoor source of PM_{2.5} in specific periods of time as mentioned above. The sample inlets of instruments for outdoor measurements were installed on the roof of SORPES station less than 10 m from the windows of the room where indoor measurement was conducted, and simultaneously, for

Fig. 1. Indoor microenvironment of the sampling room in this study: (a) vertical view and (b) 3-D picture. Note: the positions of the instruments are shown in the figures (red: indoor observation instruments; blue: outdoor observation instruments).

indoor monitoring, the inlets of instruments were set indoors 1.0 m above the ground and 1.5 m away from the window of the western side in the room (Fig. 1).

Instrumentation

Indoor monitoring of O_3 volume fraction concentrations was carried out using a photometric ozone analyzer (Model T400; Teledyne API) while outdoor measurement relied on a Thermo Scientific Model 49i. The measurement principles of two instruments were the same based on the Beer-Lambert law and the comparison between gas stream containing O_3 and a stream that had been scrubbed of O_3 . The resolutions were both 0.1 ppb and the counting intervals were both set to be 5 minutes during the sampling period. For CO volume fraction concentrations, indoor and outdoor monitoring was conducted by a CO analyzer with auto-reference (Model T300U; Teledyne API). The measurement principle was based on Beer-Lambert law and comparison between concentrations of CO and the other neutral gas. The CO concentrations were computed by taking the ratio of the instantaneous values of both gases and then compensating the ratio for sample temperature and pressure. The measurement range was from 0 to 100 ppm with precision getting 0.5% reading. The counting interval was set as 5 minutes during the sampling period. The comparison between instruments for indoor and outdoor O_3 and CO measurements was conducted by synchronously measuring outdoor samplings before the campaign. The correlations between indoor and outdoor O_3 and CO instruments were both satisfying, with correlation coefficient $r = 0.99$ and 0.98 , root mean square error (RMSE) < 3.0 ppb and < 45 ppb, respectively.

Outdoor $PM_{2.5}$ mass concentration was monitored by a SHARP 5030i (Thermo Scientific) based on light scattering of aerosols and the principle of β attenuation. The sensitivity

of the instrument was $0.1 \mu g m^{-3}$ with the range allowed from 0 to $10,000 \mu g m^{-3}$, and the counting interval was 5 minutes. For the indoor measurement of $PM_{2.5}$ mass concentration, a SidePak Personal Aerosol Monitor AM510 (TSI Inc.) was used, which relied on 90° light scattering and laser diode, whose minimum resolution was $1 \mu g m^{-3}$ and range was from 1 to $20,000 \mu g m^{-3}$. The counting interval was set to be 1 minute. Due to the different operation principles between the SHARP 5030i and SidePak AM510, it is necessary to convert the data acquired by the SidePak AM510 towards the standard of the SHARP 5030i, which has been viewed as the reference instrument with good stability and high accuracy. The comparison between the two instruments was carried out before the indoor-outdoor campaign. Linear fitting was used and shown satisfying result:

$$PM_{2.5} (SHARP 5030i) = 0.43 \times PM_{2.5} (SidePak AM510); \\ r = 0.92, p < 0.001 \quad (1)$$

During the later indoor-outdoor measurement, indoor data gained by the SidePak AM510 was converted to the standard of the SHARP 5030i using the linear fitting equation above.

RESULTS AND DISCUSSION

The Time Series and Statistical Results

As presented in Table 1, for O_3 , the statistical results fluctuated between 0.9 and 26.6 ppb indoors (average: 4.5 ppb; median: 3.2 ppb), and 0.0 and 105.7 ppb outdoors (average: 32.5 ppb; median: 30.5 ppb). Except the minimum value, the statistical values of outdoor O_3 concentrations mentioned above were all higher than those from indoor measurement.

As shown in Fig. 2(a), the outdoor concentrations of O₃ were much higher during most of the measurement period, especially in daytime. This resulted from the active photoreactions outdoors generating large amount of outdoor O₃, while indoor concentrations relied predominantly on indoor-outdoor transport with continuous indoor deposit but almost no indoor source. Another research conducted in Finland reached a similar conclusion that indoor ozone levels climbed only during a few hours in 1 day (Koponen *et al.*, 2001). Nevertheless, the indoor minimum value was higher than that of outdoor results, which was ascribed to

extremely low outdoor level that appeared usually before dawn (Fig. 2(a)). This was caused by intense all-night outdoor O₃ consumption, the process of which was weak indoors. I/O ratio (the ratio of indoor concentration to outdoor concentration) of O₃ was 0.14 attributed to significant gap between indoor and outdoor concentrations during daytime. Other O₃ measurement campaigns found I/O ratios ranging from 0.2 to 0.7 (Romieu *et al.*, 1998; Bernard *et al.*, 1999; Weschler, 2001), while the values in this research were less probably owing to tightly sealed building structure and absence of indoor sources.

Table 1. The statistical results of indoor/outdoor O₃, CO and PM_{2.5} concentrations and I/O ratios.

	Ozone (ppb)	Carbon Monoxide (ppb)	PM _{2.5} (μg m ⁻³)
Indoor			
Mean	4.5	447.3	23.7
Median	3.2	395.8	17.3
SD	3.9	204.9	16.8
Range	0.9–26.6	154.6–1451.2	5.3–133.4
Outdoor			
Mean	32.5	450.9	56.1
Median	30.5	386.1	37.2
SD	23.5	246.9	51.2
Range	0.0–105.7	156.9–2177.2	2.7–242.5
I/O ratio (median)	0.14	1.01	0.46

Fig. 2. The time series of indoor (red lines) and outdoor (blue lines): (a) O₃ and (b) CO concentrations from September 20 to November 3, 2018, and (c) PM_{2.5} concentrations from October 13 to December 6, 2018.

For CO, the range of indoor concentrations was from 154.6 to 1451.2 ppb (average: 447.3 ppb; median: 395.8 ppb), and from 156.9 to 2177.2 ppb (average: 450.9 ppb; median: 386.1 ppb) outdoors (Table 1). The outdoor minimum and maximum CO concentrations were both greater than those indoors, which were in line with the measurement in Shanghai (Zhong *et al.*, 2013). Different from indoor and outdoor O₃ concentrations, as shown in Fig. 2(b), the indoor and outdoor CO concentrations were quite close. It could be because CO have longer lifetime than O₃. The outdoor standard variation was 246.9 ppb compared to 204.9 ppb indoors. Fig. 2(b) indicates that when outdoor CO concentrations encountered short-term spikes, indoor concentrations responded weakly, which explains higher standard variation of outdoor CO concentrations. I/O ratio of CO was close to 1.0, which resulted from, as mentioned above, quite similar indoor and outdoor concentrations except short-term fluctuations. This value was comparable with other measurements (WHO, 2010; Zhong *et al.*, 2013).

For PM_{2.5}, it shows that indoor PM_{2.5} mass concentrations varied from 5.3 to 133.4 $\mu\text{g m}^{-3}$ (average: 23.7 $\mu\text{g m}^{-3}$; median: 17.3 $\mu\text{g m}^{-3}$), and the range of outdoor concentrations was from 2.7 to 242.5 $\mu\text{g m}^{-3}$ (average: 56.1 $\mu\text{g m}^{-3}$; median: 37.2 $\mu\text{g m}^{-3}$) (Table 1). Maximum, average and median values of indoor concentrations were significantly lower than values outdoors, which indicates low indoor-outdoor air exchange rate and dependence of indoor concentration on transport from outdoor sources. However, the indoor minimum was greater than the outdoor minimum, and this occurred when clean air mass passed outdoors but indoor PM_{2.5} was not transported outdoors timely due to low air exchange rate. The indoor standard variation (16.8 $\mu\text{g m}^{-3}$) was considerably less than the outdoor one (51.2 $\mu\text{g m}^{-3}$), revealing sharper fluctuations of PM_{2.5} mass concentrations outdoors. The median I/O ratio of PM_{2.5} was

0.46 in this study. The value was comparable to other measurements ranging from 0.35 to 0.59 (Monn *et al.*, 1995; Landis *et al.*, 2001; Ho *et al.*, 2004; Polidori *et al.*, 2007), but lower than some other measurements (Adgate *et al.*, 2003; Ramachandran *et al.*, 2003; Brunekreef *et al.*, 2005). Therefore, it should be noted that I/O ratio of PM_{2.5} varies widely depending on locations, seasons, ventilation modes and a series of other factors.

Characteristics of Diurnal Cycles

Diurnal cycles of indoor/outdoor O₃, CO and PM_{2.5} concentrations as well as I/O ratios are presented in Fig. 3. The indoor and outdoor O₃ concentrations were generally higher from 10:00 LT to 17:00 LT when radiation intensified the formation of outdoor O₃ through photochemical reactions. The indoor O₃ concentration tracked outdoor concentration well, as other experiments pointed out (Sabersky *et al.*, 1973; Hales *et al.*, 1974), but its variations were much slighter with median values less than 10 ppb all the day. The highest value of indoor O₃ appeared at 16:00 LT (7.4 ppb), 1 hour after the zenith of outdoor level. The I/O ratio of O₃ peaked at 05:00 LT, when the outdoor concentration was low and indoor concentration remained relatively constant during nighttime. Interestingly, when human activities appeared indoors in general (09:00–12:00 LT, 03:00–06:00 LT), indoor concentration continued to increase or maintains high, and even peaked at 16:00 LT, which was inconsistent with some research revealing occupancy of human indoors resulted in reduction of O₃ concentrations due to surface reaction with human skin (Rim *et al.*, 2009; Wisthaler and Weschler, 2010; Fischer *et al.*, 2013). It was likely that indoor human appearance, in this study, broke the relatively good airtightness by frequent opening of the door and consequently promoted the transport of outdoor O₃ to the room.

The pattern of indoor-outdoor CO diurnal variations was

Fig. 3. Diurnal cycles of indoor concentrations (red lines, bottom panels), outdoor concentrations (blue lines, bottom panels) and I/O ratios (black lines with points, top panels) of (a) O₃, (b) CO and (c) PM_{2.5}. Note: shadow areas represent the ranges between 25th and 75th percentiles of indoor and outdoor concentrations.

considerably different from that of O_3 , although indoor concentrations also tracked the variations of outdoor concentrations (Fig. 3(b)). The I/O ratio of CO fluctuated around the value of 1.0. The peaks of indoor/outdoor CO concentrations were found in the morning and late afternoon, which could be caused by incomplete combustion of fuels from vehicles during rush hours (Ye *et al.*, 2000). The indoor concentration peaks were lower than outdoor ones due to the incomplete response of indoor concentrations towards sharp outdoor concentration variations. During the noon, concentrations were slightly lower compared with other periods, which rose from the dilution by boundary layer evolution. The time delay of indoor CO variations compared to outdoor CO was observed. Based on the lag correlation calculation, the time lag is about 65 minutes during the campaign. The obvious time lag is regarded as a typical behavior characterizing the response of indoor CO concentrations to the fluctuations of outdoor CO when air exchange rate is low (Hayes, 1991; Chaloulakou and Mavroidis, 2002; Chaloulakou *et al.*, 2003). No evidential influence of indoor human activities on indoor CO concentrations was recorded.

Outdoor concentrations of $PM_{2.5}$ were higher than indoor concentrations all day long with the I/O ratio fluctuating from 0.38 to 0.67 (Fig. 3(c)). During nighttime, outdoor values were averagely higher, mainly due to lower boundary layer height and accumulation of particles while the indoor concentrations were lower at night. Indoor $PM_{2.5}$ concentration as well as I/O ratio peaked at 10:00 LT ($22.6 \mu g m^{-3}$) and 17:00 LT ($23.8 \mu g m^{-3}$), which corresponded to the schedule of indoor human activity. Considering some behavior, like smoking, was strictly prohibited in the room, the walking of researchers, which triggered re-suspension of particles, was deemed to be the main contributor of the concentration increase. Previous studies have also found the impact of human walking on indoor particle concentrations (Fromme *et al.*, 2007). Additionally, as discussed before, the potential increase of indoor-outdoor air exchange attributed to behavior like opening the door, which changed the “openness” condition of the room, can be another factor that gave rise to indoor concentration increase (Hayes, 1989, 1991).

Indoor-outdoor Correlations

The correlation between indoor and outdoor O_3 concentrations was moderately positive ($r = 0.68$; Fig. 4(a)), which was consistent with other measurements (Blondeau *et al.*, 2004; Loupa *et al.*, 2006; Walker and Sherman, 2013). The results also show that when outdoor concentrations were higher than 40 ppb, the linear discrete degree turned out to be more scattering. Therefore, it should be taken into consideration that when outdoor level rises to be relatively high, corresponding indoor level can vary relatively widely. In specific cases, to assess indoor concentration based on outdoor concentration and linear function may lead to potential underestimation of indoor concentration. The primary coefficient acquired by linear regression (0.11) was quite close to the I/O ratio 0.14.

The correlation with respect to indoor-outdoor CO was positive with high correlation coefficient ($r = 0.82$; Fig. 4(b)). In comparison to the results of O_3 , the phenomenon of increasingly scattering linear discrete degree was not so marked. Hence, to evaluate indoor CO concentration using both outdoor concentration and a linear function tends to be accessible and reasonable, at least when CO level is not so high. However, the primary coefficient from linear regression (0.73) was obviously less than the median I/O ratio (1.01).

Fig. 4(c) shows that the correlation between indoor and outdoor $PM_{2.5}$ mass concentrations was positive with high correlation coefficient ($r = 0.82$). This was consistent with other measurements, e.g., in Guangzhou and Beijing (Huang *et al.*, 2007; Zhao *et al.*, 2015). The primary coefficient resulting from linear regression (0.27) indicated that nearly 27% of outdoor $PM_{2.5}$ can infiltrate into indoor environment under the measurement condition. However, the primary coefficient was not comparable with the I/O ratio (0.27 vs. 0.46 for primary coefficient and I/O ratio, respectively).

Impact of Meteorological Factors

As shown in Fig. 5(a), greater wind speed may enhance the transport into indoor microenvironment, and thus caused larger I/O ratio of O_3 . The slope for indoor O_3 concentration above 25 ppb was greater than that for O_3 concentration below 10 ppb (0.083 vs. 0.027), indicating that wind speed

Fig. 4. The correlations of indoor and outdoor (a) O_3 , (b) CO and (c) $PM_{2.5}$ concentrations with boxes showing 10th, 25th, 50th, 75th and 90th percentiles in each section. Note: the red lines are the linear fitting results.

Fig. 5. Scattering plot of (a) wind speed vs. O_3 I/O ratios (color-coded with indoor O_3 concentration; lines represent linear fittings for all data points [black], points with indoor concentration below 10 ppb [red] and points with indoor concentration above 25 ppb [green]), (b) wind speed vs. $PM_{2.5}$ I/O ratios (color-coded with outdoor relative humidity; the line represents linear fitting for all data points), and (c) daily averaged wind speed vs. daily time lag of CO (color-coded with daily averaged I/O ratios of CO; the line represents linear fitting for all daily data points). Note: the points only with outdoor concentration above 40 ppb and $50 \mu g m^{-3}$ were included in Figs. 5(a) and Fig. 5(b), respectively.

played its part more significantly under high-indoor-concentration condition. This can result from human activities that increased indoor-outdoor exchange by altering “openness” condition of the room (e.g., open the door) and consequently caused higher I/O ratio (Hayes, 1991), which also allowed sharper I/O ratio increase when wind speed rose.

Different with O_3 , Fig. 5(b) indicates that wind speed had little impact on I/O ratio of $PM_{2.5}$ ($r < 0.1$), but the relationship between outdoor relative humidity and $PM_{2.5}$ I/O ratio can be seen here. Statistical analysis shows that when outdoor relative humidity ranged from 20% to 50%, the corresponding median values of outdoor and indoor concentrations and I/O ratio were $60.5 \mu g m^{-3}$, $24.4 \mu g m^{-3}$ and 0.41, respectively (only the data with outdoor $PM_{2.5}$ concentration higher than $50 \mu g m^{-3}$ was considered in the analysis). As a comparison, when RH range increased to 50–80%, the median values became $118.1 \mu g m^{-3}$, $36.5 \mu g m^{-3}$ and 0.30, respectively. Therefore, the aqueous formation of secondary aerosols and hygroscopic growth of PM when RH climbed (Seinfeld *et al.*, 2001; Hu *et al.*, 2010) can increase the outdoor $PM_{2.5}$ concentration and lead to lower I/O ratio. However, when RH increased to 80–100%, the outdoor $PM_{2.5}$ concentration decreased while the indoor $PM_{2.5}$ concentration and I/O ratio increased (the median values of outdoor and indoor concentrations and I/O ratio were $93 \mu g m^{-3}$, $41.6 \mu g m^{-3}$ and 0.38, respectively). It is mainly caused by the strong outdoor wet deposition during drizzling rain or fog, which was common in the season of the measurements. Some studies also claimed that high RH and associated strong hygroscopic growth enhance particle deposition (Hänel, 1982; Quinn and Ondov, 1998; Wolkoff and Kjærsgaard, 2007). The negative relationship between PM I/O and outdoor relative humidity is consistent with the findings by Chan (2002).

Fig. 5(c) shows the impact of wind speed on the time lag between indoor and outdoor CO variations. The daily time lag of CO, which was estimated based on the lag correlation calculation, tended to decrease following the increase of daily averaged outdoor wind speed. It is because high

outdoor wind speed enhanced indoor-outdoor exchange and thereby decreased the time lag between indoor and outdoor variations. The existence of time lag can explain the CO I/O ratios exceeding 1.0 (Table 1 and Fig. 5(c)). When outdoor concentration started declining especially after obvious peaks, indoor concentration was rising due to the time lag and caused the indoor concentration higher than the outdoor concentration.

Indoor Air Quality during the O_3 and $PM_{2.5}$ Pollution

To better understand the indoor air quality, especially the features during O_3 and $PM_{2.5}$ polluted days, typical episodes were selected and analyzed.

An ozone pollution episode was observed from October 2 to October 7, 2018 (Fig. 6). During this episode, continuous high O_3 concentrations were observed with highest value reaching about 109 ppb on October 7. The continuous high O_3 concentration was caused by appropriate synoptic weather and high anthropogenic emissions during Chinese National Day. Indoor O_3 concentration was 10.7 ppb when the outdoor O_3 concentration got its maximum on October 7 (109.1 ppb). The highest indoor O_3 concentration during the episode was observed on October 6, with the value around 30 ppb. On October 7, although the outdoor O_3 concentrations was highest at about 14:00 LT, the indoor O_3 concentrations peaked at 10:00 LT. Human activities that may increase air exchange (e.g., open the door) (Hayes, 1991; Koponen *et al.*, 2001) can explain such differences according to the working records of the station. Additionally, the indoor O_3 concentrations on October 4 and 5 were higher than those on October 2 and 3 although the outdoor O_3 concentrations were actually lower. Much higher wind speed was observed on October 4 and 5, supporting that high wind speed promoted the outdoor O_3 transport into the room as described above. Indoor and outdoor CO concentrations during the episode were also presented in Fig. 6. About 60 minutes’ time lag of indoor and outdoor CO concentrations was observed during the episode, which was consistent with the discussion above.

Fig. 6. Time series of (a) temperature, relative humidity; (b) wind speed, wind direction; (c) indoor/outdoor CO concentrations and (d) indoor/outdoor O₃ concentrations from October 2 to October 7, 2018.

From November 27 to December 2, 2018, PM_{2.5} pollution event with peaks exceeding 200 $\mu\text{g m}^{-3}$ was observed at SORPES (Fig. 7). The pollution was mainly caused by local accumulation with obvious near-ground inversion layer and heavy fog. Indoor concentration (42.6 $\mu\text{g m}^{-3}$) during the episode was averagely higher than outdoor level during non-episode days (40.2 $\mu\text{g m}^{-3}$). Indoor PM_{2.5} concentration can get 65.8 $\mu\text{g m}^{-3}$ when outdoor concentration reached its maximum (242.5 $\mu\text{g m}^{-3}$). Some indoor peaks, such as the one on the afternoon of November 29 with the maximum value of 131.6 $\mu\text{g m}^{-3}$, were recorded. According to the working records of the station, it is likely that it was human activities (e.g., opening the door or walking frequently indoors) that contributed to the peaks indoors by increasing indoor-outdoor air exchange and re-suspension of particles.

The increase of outdoor PM_{2.5} concentration was significantly more than the rise of concentration indoors during pollution days (e.g., November 29 and November 30). This means when outdoor concentration climbed to certain level, the transport towards indoor microenvironment through infiltration did not increase proportionally as outdoor concentration rose. A feasible explanation is that a penetration limitation existed. It is likely that the increase of transport slowed down when outdoor concentration climbed to certain level, because of limited space of cracks and leaks for infiltration and some other factors. This can also be substantiated by I/O ratios, which was lower during episode than during non-episode days (episode average: 0.40 vs. non-episode average: 0.61). Thus, averaged I/O ratio may not be an effective indicator of indoor pollutant accumulation under pollution circumstances, at least for PM_{2.5}.

Estimations of Indoor Pollutant Concentrations

A statistical method was used for the simulation of indoor

O₃, CO and PM_{2.5} concentrations. The method applied in this study is based on a mass-balance equation for pollutants that flow into and out of the indoor microenvironment, including sinks and sources (Shair and Heitner, 1974; Esmen, 1978; Ishizu, 1980). The initial mass-balance equation is from previous study (Hayes, 1991; Chen and Zhao, 2011) and simplified based on the research conditions in this study:

$$\frac{dc_i}{dt} = PF_a c_o - F_a c_i - F_s c_i \quad (2)$$

where c_i and c_o are indoor and outdoor concentrations, respectively. F_a represents the indoor-outdoor air exchange rate, mainly through infiltration in the case of this study. F_s represents the indoor sink rate of pollutants and P represents the penetration factor.

To simplify the equations, F_a , F_s and P were considered constant and this treatment is common for a given space under the same window/door “openness” condition (Hayes, 1991; Liu and Zhang, 2019). Although the experimental conditions were not strictly unchanged, the simplification is reasonable considering relatively short time of disruption, e.g., human activities. The penetration factor P should be taken into consideration for PM_{2.5} because only a proportion of particles in the infiltration air succeed in passing through the building shell, while it was set to be one for gases. The rapid surface removal of O₃ by indoor materials was significant, so when conducting simulation on indoor O₃ concentration, two factors F_a and F_s were both included. However, CO had lower reactivity and longer lifetime indoors, so F_s was set to be zero when dealing with the prediction of indoor CO concentration. The equation was solved by iteration with 5 minutes as the length of one step

Fig. 7. Time series of (a) temperature, relative humidity; (b) wind speed, wind direction and (c) indoor/outdoor $\text{PM}_{2.5}$ concentrations from November 27 to December 2, 2018.

and the data from first 30 days was used. The combination of parameters (F_a , F_s and P) that resulted in smallest RMSE between measured and simulated concentrations was calculated for each pollutant.

Based on the CO data, the F_a was calculated to be 0.408 h^{-1} . Considering infiltration as the primary pathway, some other research obtained the value of 0.4 h^{-1} (Persily et al., 2010), $0.02\text{--}0.82 \text{ h}^{-1}$ (Shi et al., 2015) and $0.36\text{--}1.17 \text{ h}^{-1}$ (Bekö et al., 2016) by modeling or measurement. The value obtained in this study was comparable to them. Considering it is the same indoor environment, it is reasonable to apply this value of F_a , which represents indoor-outdoor air exchange of the room, to the simulations of O_3 and $\text{PM}_{2.5}$. Based on O_3 data, the F_s of O_3 was calculated, which resulted in 2.64 h^{-1} . The F_s and P of $\text{PM}_{2.5}$ were also obtained based on $\text{PM}_{2.5}$ data, with value of 0.12 h^{-1} and 0.60 , respectively. The penetration factor P (0.60) was reasonable in comparison with the results of some other research which obtained the values from measurements in both real buildings and laboratories (Vette et al., 2001; Liu and Nazaroff, 2003; Thatcher et al., 2003).

About 5-day indoor data, which was not applied in obtaining the parameters, was used to verify the estimation (Fig. 8). For simulated indoor O_3 concentrations, the simulated result turned out to be fairly congruent during most of the period ($r = 0.54$; $\text{RMSE} = 4.6 \text{ ppb}$). It also showed that the method lacked the ability for simulating some short-term fluctuations. Additionally, from the afternoon of October 31 to the afternoon of November 1, 2018, the simulated concentration showed underestimation (r and RMSE become 0.75 and 1.6 ppb , respectively, if this period is excluded). The incompatibility of this ~ 1 -day period may merely be brought about by disruption, e.g., the door was not completely closed. A five-fold indoor-outdoor air exchange rate was applied for this period (black dashed line in Fig. 8(a)), which resulted in significantly better agreement between observation and simulation ($r = 0.82$; $\text{RMSE} = 3.2 \text{ ppb}$). As for CO, the consistency between measured and simulated concentrations turned out to be markedly good ($r = 0.96$; $\text{RMSE} = 29.2 \text{ ppb}$), although the concentration from

simulation was unable to respond to some steep and short-term oscillations (Fig. 8(b)). Simulation of indoor $\text{PM}_{2.5}$ concentration also got moderate consistency ($r = 0.78$; $\text{RMSE} = 18.1 \mu\text{g m}^{-3}$) (Fig. 8(c)). For some periods (e.g., after December 1), simulated concentration fitted well although short-term fluctuations cannot be embodied. It can be found that from November 28 to December 1, the outdoor $\text{PM}_{2.5}$ peaks frequently exceeded $150 \mu\text{g m}^{-3}$, and the simulation showed obvious overestimation. This means that high outdoor $\text{PM}_{2.5}$ concentration could cause quite high indoor concentration based on the simulated result, while according to the observation it is limited by penetration as described above. Using lower penetration factor ($P = 0.38$) can improve the simulations (Fig. 8(c)), with correlation coefficient reaching 0.82 and RMSE declining to $9.3 \mu\text{g m}^{-3}$.

CONCLUSIONS

This study monitored indoor and outdoor concentrations of O_3 , CO and $\text{PM}_{2.5}$ in real time in suburban Nanjing, located in the Yangtze River Delta, China, from September to December 2018. The indoor measurements were conducted in a room with closed windows and relatively regular daily indoor human activity. The statistical results and diurnal variations were analyzed, and the indoor-outdoor relationships were characterized. A simulation method was used to evaluate parameters for predicting the indoor concentrations. Our findings are summarized as follows:

- 1) The concentration of O_3 was higher outdoors (median: 30.5 ppb) than indoors (median: 3.2 ppb); this difference was especially noticeable during the daytime. The indoor concentrations traced the outdoor variations with a moderately positive correlation ($r = 0.68$) and a median I/O ratio of 0.14 . The outdoor (median: 386.1 ppb) and indoor (median: 395.8 ppb) concentrations of CO remained close during most of the period, except during steep short-term oscillations outdoors, and displayed an overall positive correlation of 0.82 and a median I/O ratio of 1.01 . A delay of approximately 65 minutes in

Fig. 8. Comparisons of observed outdoor (blue dashed lines), observed indoor (red solid lines) and simulated indoor (black solid/dashed line) (a) O_3 , (b) CO and (c) $PM_{2.5}$ concentrations during tested periods. Note: the dashed black lines represent the improved simulated indoor concentrations of O_3 and $PM_{2.5}$ considering indoor human activities and limited penetration, respectively.

the indoor variations compared to the outdoor ones was also observed. The outdoor concentration of $PM_{2.5}$ (median: $37.2 \mu g m^{-3}$) significantly exceeded the indoor one (median: $17.3 \mu g m^{-3}$), and the latter followed the former. The indoor-outdoor correlation was high ($r = 0.82$), and the median I/O ratio was 0.46. Although human activity indoors greatly influenced the indoor concentrations of O_3 and $PM_{2.5}$ by breaking the airtightness of the room or re-suspending particles, it showed no obvious effect on the indoor CO concentration.

- 2) The outdoor wind speed exhibited a positive correlation with the I/O ratio for O_3 , indicating that a higher wind speed may boost transport into indoor microenvironments, but no obvious correlation with the I/O ratio for $PM_{2.5}$ was found. Instead, when the relative humidity (RH) was low, the $PM_{2.5}$ I/O ratio decreased as the RH increased, whereas when the RH was high, this ratio was positively

correlated with it. Furthermore, the wind speed was negatively correlated with the time lag for the indoor CO concentration, suggesting that a higher wind speed increases indoor-outdoor exchange rate and reduces the delay indoors.

- 3) During the O_3 pollution episode of October 2–7, 2018, the indoor concentration was 10.7 ppb at the time when the outdoor concentration reached its maximum of 109.1 ppb; the indoor concentration can reach approximately 30 ppb during the episode. During the $PM_{2.5}$ pollution episode of November 27–December 2, 2018, the average indoor concentration reached $42.6 \mu g m^{-3}$, exceeding even the outdoor concentration on unpolluted days ($40.2 \mu g m^{-3}$). The indoor concentration was $65.8 \mu g m^{-3}$ when the outdoor concentration reached its maximum of $242.5 \mu g m^{-3}$, and the I/O ratio decreased during the episode, reflecting the limitation on penetration.

- 4) The indoor-outdoor air exchange rate, indoor sink rate and penetration factor were evaluated using statistical calculation based on a simplified indoor-outdoor mass-balance equation. These parameters and the equation were then applied to simulate the indoor concentrations of the pollutants, and a comparison with the measurements confirmed that the predictions fitted the observed data during most of the testing period, especially after considering the indoor human activity and limited particle penetration.

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