Environmental Science & Technology

Ozone, Electrostatic Precipitators, and Particle Number Concentrations: Correlations Observed in a Real Office during Working Hours

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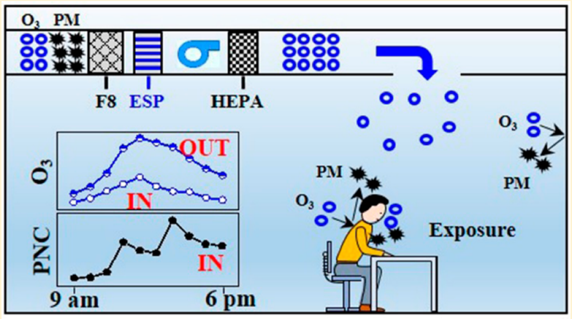
Supporting Information

ABSTRACT: This study investigates the impacts of outdoor and indoor ozone concentrations, ESP operation and occupancy on particle number concentrations within a modern office in Changsha, China. The office's one-pass air handling system contains a mini-bag filter (MERV 12) followed by an electrostatic precipitator (ESP) and high efficiency particulate air (HEPA) filter. Over a five-week period the system was operated either without the ESP (Stage 1, first−third week) or with the ESP (Stage 2, fourth and fifth week). Ozone and particle number concentrations were measured on working days. During both stages, indoor ozone and particle number concentrations tracked the outdoor ozone concentration. When operating, the ESP produced approximately 29 mg h^{-1} of ozone, increasing supply air ozone by 15 ppb and steady-state indoor ozone by about 3 ppb. Occupancy tended to decrease indoor ozone and increase particle levels. During occupancy, indoor particle levels were low (~2600 particle/cm^3) when the supply air ozone level was less than 18 ppb. Above this threshold, the supply air ozone concentration and indoor particle number concentration were linearly related, and ESP operation increased the average indoor particle level by about 22 000 particles/cm^3. The implications for worker exposure to both ozone and particles are discussed.



1. INTRODUCTION

In present day China residents pay considerable attention to $PM_{2.5}$ mass concentration, reflecting the extremely high levels that occur on many days each year. Particle number concentration has received less attention, although it is readily measured in real time using commercially available condensation particle counters (CPCs). The particle number concentration is dominated by particles with aerodynamic diameters less than 100 nm, so-called ultrafine particles (UFPs); such particles contribute more than 70% of the total count measured with CPCs.[1−3] On a per unit mass basis, UFPs have higher specific surface area than larger particles, resulting in extensive contact with the surfaces on which they deposit. Inhaled UFPs can be deposited deep in the human lung, and, in rat studies, have been shown to enter the bloodstream from the lung.[4,5] UFPs have been associated with adverse human health effects including pulmonary and cardiovascular diseases[6−8] and even heritable mutations in an animal model.[9]

It is well-known that ozone reacts with certain indoor pollutants (primarily those containing carbon−carbon double bonds, i.e., alkenes) to generate secondary organic aerosols (SOA).[10−16] The size range of the initially formed SOA are such that they can be classified as UFPs.[17,18] It is also known that electrostatic precipitators (ESPs) are often used in ventilation systems, since they remove particles from the airstream without significantly increasing the pressure drop across the filtration elements. A disadvantage to ESPs is that they generate ozone,[19,20] which in turn can react contributing to indoor UFPs. However, it has proven difficult to directly observe the impact of ESPs on UFPs in an occupied office under typical working conditions. This is because high and varying number concentrations of UFPs in typical office settings, coupled with moderate to high and varying outdoor ozone concentrations, making it difficult to tease out the suspected contribution from ESP-derived ozone reacting with

Received: June 19, 2016
Revised: August 20, 2016
Accepted: August 29, 2016
Published: August 29, 2016

DOI: 10.1021/acs.est.6b03069
Environ. Sci. Technol. 2016, 50, 10236−10244

the indoor organic pollutants that serve as precursors for particle formation.

The present study has been conducted in an office that used a HEPA filter downstream of an ESP in a one-pass-filtration system. Hence, within the office the contribution of particles of outdoor origin to total particle number concentration is small. This makes it easier to detect particles resulting from ozone-alkene chemistry within the office. The present study has also been conducted during December and January when outdoor ozone concentrations are relatively small, resulting in small indoor ozone levels and making it easier to detect the contribution of ESP operation to total indoor ozone levels.

This study has monitored indoor PM number concentrations with a CPC during working hours in a mechanically ventilated office (80 occupants) over a five-week period. During this time the air-handling system operated for 20 days without an ESP and for 17 days with an ESP. Throughout the study other parameters (outdoor and indoor ozone, air exchange rate, occupancy, temperature and relative humidity) that might influence PM number concentrations were simultaneously monitored. In contrast to chamber studies or briefly manipulated indoor environments, our observations have been in a real-world setting over a several week period. The aim of the study has been to specifically examine the impacts of outdoor and indoor ozone levels, ESP operation, and occupancy on indoor particle number concentrations. Such information contributes to a better understanding of the factors that influence such levels and enables more accurate estimates of particle exposures in other indoor settings.

2. MATERIALS AND METHODS

2.1. Experimental Design. Ozone and particle number concentrations were monitored within a representative Chinese office (40 m × 10 m × 2.4 m, 960 m^3). Located on the third floor of a four-story building in suburban Changsha. The office had concrete flooring, gypsum ceiling tiles, standard office furniture, and well-sealed nonopening windows (0.6 m × 0.5 m) facing south. It was ventilated with a single-pass (no recirculation) air handling unit (AHU) that under standard operating conditions included a mini-bag filter (F8, MERV 12), a two-stage electrostatic precipitator (ESP), and a high efficiency particulate air (HEPA) filter capable of removing more than 99% of the particles in the airstream.[21] The ESP had 0.50 m × 0.25 m × 0.15 m collection plates and operated at a charging voltage of +6.0 kV. The design intent of the ESP was to extend the lifespan of the HEPA filter. This study was performed from December 25, 2014 to January 30, 2015 and consisted of two stages. In Stage 1 (1st−third week), the ESP unit was removed from the AHU, while in Stage 2 (4th−5th week) it was present and operating. Measurements excluded holidays and weekends, and were made on 12 days in Stage 1 and 14 days in Stage 2 (a timeline for the investigation is shown in Figure S1, Supporting Information (SI)). On weekdays, 80 white-collar workers worked from 08:00 to 12:00 and 14:00 to 18:00, taking a break outside the office from 12:00 to 14:00. Smoking, incense burning, air fresheners, or the use of cleaning agents was not allowed within the office.

Indoor particle number concentrations were monitored with a condensation particle counter (CPC 3007, TSI Inc., Shoreview, MN), which measured the total number concentrations of particles larger than 10 nm diameter with an accuracy of ±20%. The particle counter was calibrated in the TSI factory before it was delivered. Moreover, we did zero point calibrations using a dedicated "HEPA zero filter" prior to these studies. Number concentrations measured by the CPC 3007 are considered to be accurate to 100 000 particles/cm^3, but above this limit measurements are not reliable.[22] The present study maintained conditions such that number concentrations resulting from ozone-initiated chemistry, coupled with other indoor sources, were in most cases less than 100 000 particles/cm^3. The number concentration obtained from the instrument is an approximate proxy for UFP concentrations (defined as being smaller than 100 nm), since particles larger than 100 nm typically contribute less than 30% to the total particle number concentration.[1−3] Additionally, secondary organic aerosols from ozone-initiated reactions tend to be smaller than 100 nm in diameter, especially in the initial stages of formation.[17,18]

Indoor ozone was monitored with a photometric ozone analyzer (model 205, 2B Tech., Boulder, CO) while outdoor ozone was measured by an official ambient air quality monitoring station about 4.5 km from the study site. The air exchange rate (AER) was measured using an air velocity sensor in the AHU. The air handling unit (AHU) system can measure the fresh air flow rate using an integrated air velocity sensor and displays the value on a dedicated LED screen. Additionally, the system is self-regulating for fresh air flow rate with automatic feedback control. Thus, the AER of the office was relatively constant. During all the tests, indoor temperature was 22.7 ± 0.8 °C; the relative humidity was 45.6 ± 3.7%; and the AER was 1 ± 0.02 h^{-1}. The ozone monitor was calibrated using an ozone calibration source (model 306, 2B Tech., Boulder, CO). We also made a zero point calibration using an ozone adsorption tube every day before the measurements. Further details regarding the calibration of the ozone monitor are presented in the SI. The PM and ozone monitors were most frequently placed on a table in the human breathing zone, point I3 in Figure 1. In addition to the long-term monitoring at I3, the PM number concentrations and ozone concentrations at I1, I2, I4, I5, S1−S3 and O were measured in turn by the same PM and ozone monitors during a period of less than 20 min on two representative days (see Section 3.1, SI Figure S1 and Table S1 for details). The two 20 min measurements were designed to get a general sense how PM and ozone changed during outdoor-AHU-indoor transport and verify the uniformity of PM and ozone concentrations at different locations within the office at heights close to the human breathing zone.

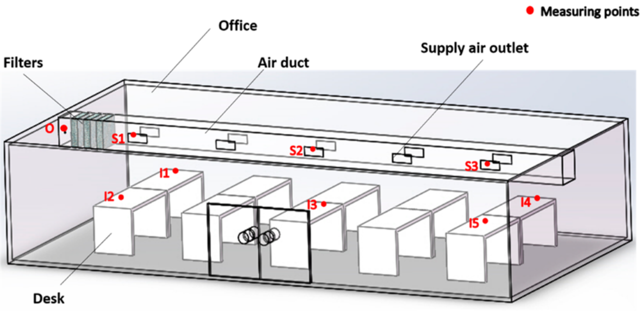


Figure 1. Schematic of the office and measuring points: the human breathing zone (I1−I5), supply air outlets (S1−S3) and the intake for the supply air (O), which was regarded as outdoor air.

2.2. Indoor Ozone Mass Balance Model. Within the office, the change in ozone level with time is given by

$$\frac{dC_{in}(t)}{dt} = \left(1 - \frac{\gamma_{filt}}{2}\right)\cdot\frac{\varepsilon}{V_{room}}\cdot R + (1 - \gamma_{filt})\cdot\lambda_v C_{out}(t) - \lambda_v C_{in}(t) - k_d C_{in}(t) - k_h C_{in}(t) \qquad (1)$$

where $C_{in}(t)$ and $C_{out}(t)$ are indoor and outdoor ozone concentration at time t, respectively [ppb], ε is the emission rate of ozone from the ESP [mg h^{-1}], V_{room} is the volume of the office [m^3], R is a conversion factor 500 [ppb mg^{-1} m^3], γ_{filt} is the combined ozone removal efficiency of the mini-bag filter and the HEPA filter, λ_v is the air exchange rate [h^{-1}], and k_d and k_h are the first order rate constant for ozone's removal by indoor surfaces and human surfaces, respectively, [h^{-1}]. The second and third terms describe sources of indoor ozone—the ESP unit and outdoor-to-indoor transport. We have assumed that the mini-bag filter and HEPA filter share equally in ozone removal, hence "$\gamma_{filt}/2$" in the second term because the ESP was positioned between these two filters. The fourth to sixth terms describe the removal of ozone by ventilation, room surfaces and human surfaces, respectively.

Under steady-state conditions, $dC_{in}(t)/dt = 0$ and the indoor ozone concentration can be calculated as follows:

$$C_{in}(t) = \frac{\left(1 - \frac{\gamma_{filt}}{2}\right)\cdot\frac{\varepsilon}{V_{room}}\cdot R}{\lambda_v + k_d + k_h} + \frac{(1 - \gamma_{filt})\cdot\lambda_v\cdot C_{out}(t)}{\lambda_v + k_d + k_h} \qquad (2)$$

From 12:00–14:00 h in Stage 1, the ESP was not operating and occupants were not indoors. Under these conditions, ε and k_h were 0, and k_d can be expressed as

$$k_d = \left((1 - \gamma_{filt})\cdot\frac{C_{out}(t)}{C_{in}(t)}\text{-}1\right)\cdot\lambda_v \qquad (3)$$

During working hours in Stage 1, ε was 0, and k_h can be expressed as

$$k_h = \left((1 - \gamma_{filt})\cdot\frac{C_{out}(t)}{C_{in}(t)} - 1\right)\cdot\lambda_v\text{-}k_d \qquad (4)$$

In Stage 2, given that k_d and k_h are known, ε can be calculated as

$$\varepsilon = \frac{((\lambda_v + k_d + k_h)\cdot C_{in}(t) - (1 - \gamma_{filt})\cdot\lambda_v\cdot C_{out}(t))\cdot V_{room}}{R\cdot\left(1 - \frac{\gamma_{filt}}{2}\right)} \qquad (5)$$

Note that the application of eqs 2–5 requires steady-state conditions.

Another method to estimate the ESP ozone emission rate is to use a mass balance model for ozone in the supply air:

$$C_{supply}(t) = (1 - \gamma_{filt})\cdot C_{out}(t) + \left(1 - \frac{\gamma_{filt}}{2}\right)\cdot\frac{\varepsilon\cdot R}{Q_{supply}} \qquad (6)$$

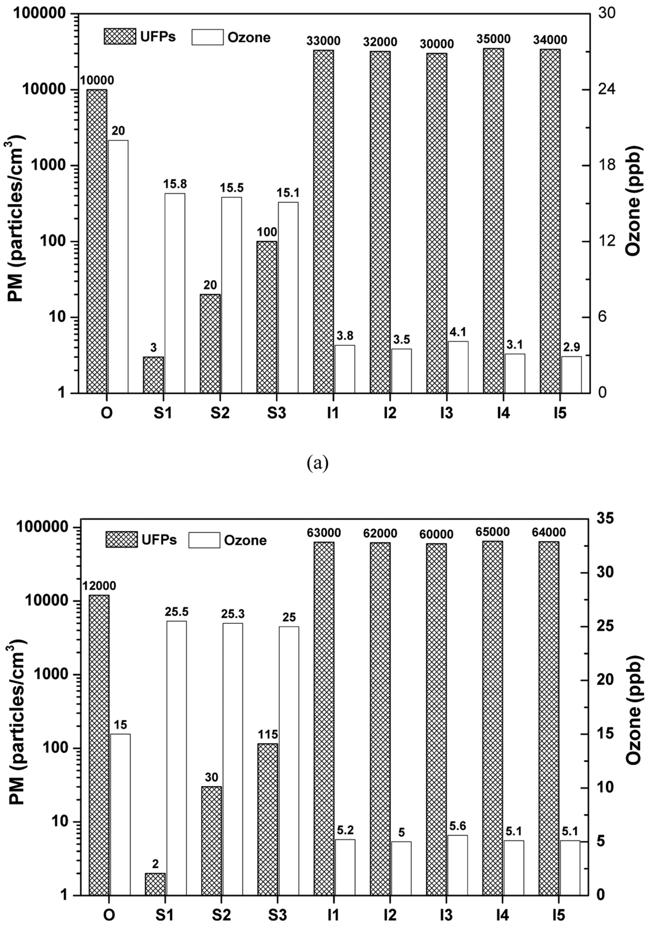
where $C_{supply}(t)$ is supply air ozone concentration at time t, and Q_{supply} is the flow rate for the supply air [m^3 s^{-1}].

Solving eq 6 for ε yields

$$\varepsilon = \frac{(C_{supply}(t)\text{-}(1 - \gamma_{filt})\cdot C_{out}(t))\cdot Q_{supply}}{R\cdot\left(1 - \frac{\gamma_{filt}}{2}\right)} \qquad (7)$$

2.3. Data Analysis. All statistical analyses were conducted using the SPSS statistics software package (version 20.0) with statistical significance defined at the $p < 0.05$ level. The correlation between ozone and particle number concentrations were analyzed using Spearman correlation analysis. The t test was used to verify the significance of linear fitting between hourly indoor particle number concentrations and supply air ozone concentrations.

3. RESULTS AND DISCUSSION



(a)

(b)

Figure 2. Ozone and PM number concentrations at different locations during the two 20 min measurements on (a) Dec. 30, Stage 1—no ESP and (b) Jan. 20, Stage 2—with an ESP (see Figure 1 for the locations of the measuring points).

3.1. Comparison of Ozone and Particle Number Concentrations at Different Indoor Locations. Figure 2a and b show the concentrations of ozone and PM (particles/cm^3) measured on two representative days at different sampling locations during Stage 1 (without ESP) and Stage 2 (with ESP), respectively. The sampling points are indicated in Figure 1 and included outdoor air, three different supply air outlets and five different working locations within the office. Details are provided in SI Table S1. On a given day the entire measurement process lasted less than 20 min and occurred during a period when outdoor ozone was relatively constant; hence, the measurements can be regarded as occurring at nearly the same time. The measurements indicate that on both occasions the ozone concentrations were similar among the five indoor sampling points (I1–I5), with a spread in values of less than 15% on either day; the same was true for PM number concentrations. Based on these measurements, point I3 was judged reasonably representative of the human breathing zone at this location, and was used as the long-term measuring location.

During Stage 1 (no ESP) the ozone concentrations at the supply air outlets were lower than that outdoors, indicating that reactive pollutants on/in the in-duct filters consumed ozone. By contrast, during Stage 2 (ESP present) the ozone concentrations at supply air outlets were higher than the outdoor concentration, indicating ozone production by the ESP. During both stages, the indoor ozone concentrations were lower than the supply air concentrations, indicating the reaction of ozone with indoor surfaces and human occupants.[16]

The PM concentrations at supply air outlets S1−S3 downstream of the HEPA filter were below 100 particles/cm^3, much lower than the outdoor PM concentrations (10 000−12 000 particles/cm^3). Thus, the HEPA removal efficiency, based on particle number concentrations, was nearly 100%. Importantly, these results indicate that the outdoor PM number concentration had almost no impact on the indoor PM number concentration, since the supply air had such low PM levels compared to the indoor air.

3.2. Daily Variations in Indoor PM Number Concentrations.

During the five-week study, we observed that the daily variation in indoor PM number concentrations (as measured at point I3) often displayed a similar pattern. Figures 3a and 3b show the daily variation in indoor PM concentrations on all measuring days during Stage 1 and 2, respectively. They indicate that on many weekdays, especially during Stage 2 with the ESP operating, PM levels started to increase midmorning, peaking in late morning/early afternoon. Even more frequently, PM levels started to increase shortly after the workers reentered the office following their 12:00−14:00 lunchtime break, peaking a little before 15:00. There were also a few days with low PM concentrations and no obvious peaks.

The pattern displayed in Figure 3 differs from that reported in other studies. For example, Sajani et al. measured UFP concentrations in residences close to traffic and found that indoor UFP peaks occurred during heavy traffic hours.[23] An important difference between the present study and this example is the use of a HEPA filter in the air handling unit servicing the building where the present measurements were made. The HEPA effectively removed outdoor PM from the ventilation air. More will be said about the pattern displayed in Figure 3 in the following subsection.

3.3. Correlations among Outdoor Ozone, Indoor Ozone, And Indoor PM Number Concentrations.

Figure 4a and b illustrate hourly variations of outdoor and indoor ozone and indoor PM number concentrations on several typical days during Stages 1 and 2, respectively. Results for the entire set of measuring days are shown in SI Figure S3. These plots illustrate that variations in indoor ozone concentrations matched variations in outdoor ozone concentrations during both stages. Spearman correlation analysis indicates that the indoor ozone concentration was significantly correlated with the outdoor ozone concentration in both stages ($\rho > 0.7$, $p < 0.0005$), consistent with expectations based on other studies[24−26] that outdoor ozone is an important source of indoor ozone. These figures also show that the indoor PM number concentration varied with outdoor and indoor ozone concentrations. Spearman correlation analysis indicates that indoor PM number concentrations were significantly correlated with outdoor and indoor ozone concentrations (SI Table S2).

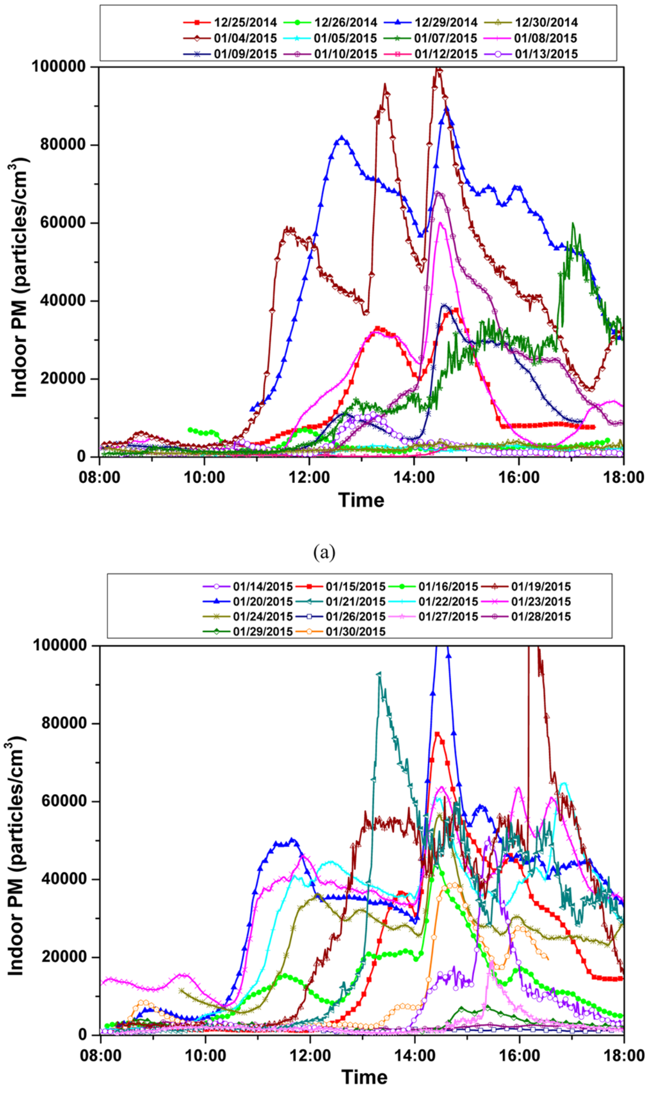


Figure 3. Daily variations of indoor PM number concentrations sampled at location I3: (a) Stage 1—no ESP; (b) Stage 2—with an ESP.

As discussed previously, ozone can react with unsaturated organic compounds (e.g., terpenes from building materials and cleaning products; squalene and unsaturated fatty acids from occupants' skin oil), resulting in SOA formation. The limited number of other indoor PM sources in this office (i.e., the prohibition on smoking, cooking and incense use) likely improved the correlation between indoor PM concentrations and ozone concentrations (both outdoors and indoors).

During both Stages 1 and 2, the Spearman correlation coefficient between indoor PM number and outdoor ozone concentrations is somewhat larger than that between indoor PM number and indoor ozone concentrations (SI Table S3), indicating that outdoor ozone concentration is a better predictor of indoor PM number concentrations than indoor ozone concentration. This may reflect the fact that measured indoor ozone levels are "residual concentrations"—the ozone which remains after a fraction of supply air ozone has been consumed by reactive indoor organics—and therefore less directly linked to the ozone-initiated chemistry that contributes to indoor PM.

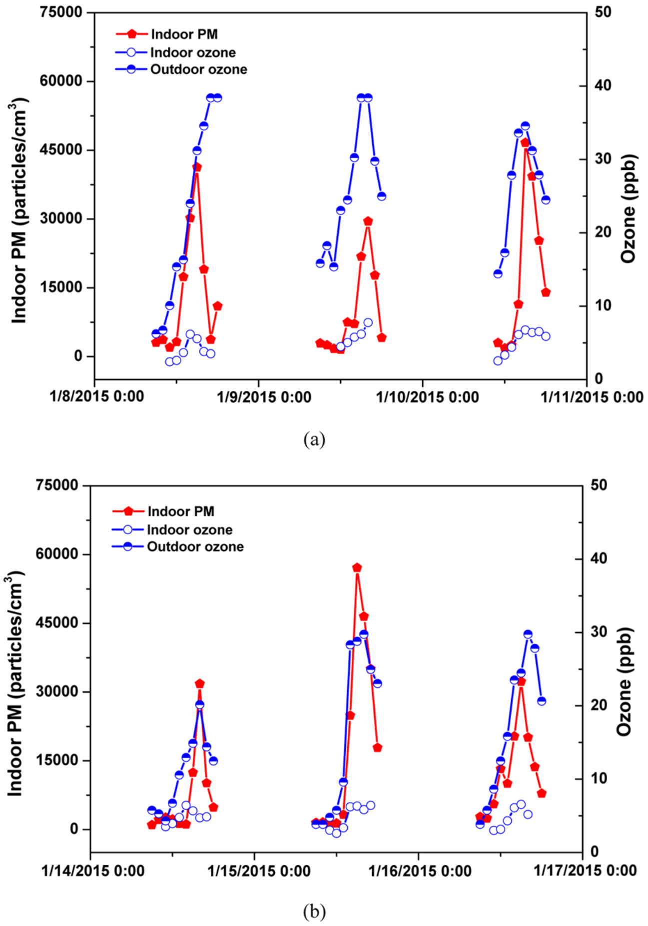


Figure 4. Hourly temporal variation of indoor and outdoor ozone and PM number concentrations for typical days: (a) Stage 1—no ESP; (b) Stage 2—with an ESP.

The strong correlation between indoor particle number concentration and outdoor ozone helps to explain the pattern that is observed in Figure 3. The late morning/early afternoon peak occurred on days when the outdoor ozone increased gradually throughout the morning. The more distinct peak that occurred shortly before 15:00 corresponded with the daily peak in outdoor ozone; the six instances shown in Figure 4 illustrate this point. We suspect that this peak is somewhat more pronounced than it might otherwise be as a consequence of the workers returning to the office at 14:00. We are not suggesting that this is due to occupant activities or resuspension caused by occupants. Abt et al. showed that the movement of people results in the resuspension of larger particles (aerodynamic diameters between 0.7 and 10 μm)—not submicron particles.[27] The latter are tightly held to surfaces by van der Waals forces and are difficult to resuspend. More likely, given the lack of other sources, the increase in PM levels between 14:00 and 15:00 partially reflects reactions between ozone and reactive organics present in human skin oils and personal care products.[16,28] If all else remains unchanged, a sudden increase in human occupants (at 14:00) would reduce indoor ozone levels[16] but could either decrease[29] or increase indoor SOA levels.[28,30] The net effect depends on the relative levels of reactive organic compounds from nonhuman sources (typically scented products containing terpenes) versus reactive organics from human sources (typically squalene, unsaturated fatty acids and terpenes). If the concentrations of reactive organic compounds from nonhuman sources are relatively low compared with those from human sources, SOA levels are anticipated to be larger with humans than absent humans. On the other hand, if the concentrations of reactive organic compounds from nonhuman sources are high compared to human sources, then SOA levels are anticipated to be smaller with humans than absent humans.[29] In the case of this office environment, it appears that the concentrations of reactive organic compounds from nonhuman sources was low compared with those from human sources.

3.4. Ozone Removal by Filters and Surfaces; Ozone Generation by the ESP.

Using Equations 3−7 and measurements made during periods close to steady-state for indoor ozone, we calculated the combined ozone removal efficiency of the mini-bag filter and the HEPA filter (γ_{filt}), the first order rate constant for ozone removal by indoor surfaces (k_d) and by human surfaces (k_h), and, finally, the ozone generation rate for the ESP (ε). The results are summarized in SI Table S3. (Due to the changing solar flux coupled with varying meteorological conditions, there were a limited number of periods when the outdoor ozone concentration was relatively constant from hour-to-hour. It was only during such periods that indoor ozone levels were close enough to steady-state that we could make the calculations just described. Our criteria: if outdoor ozone varied by less than 3.5 ppb during the previous 2 h, the period was deemed appropriate for making the calculations just described.)

The combined ozone removal efficiency of the mini-bag filter and HEPA filter (γ_{filt}) was determined by measurements immediately upstream and downstream of the filter-bank and was approximately 20%. The mini-bag filter had been in service for ~3 months and the HEPA filter had been in service for one year. A number of studies have measured ozone removal efficiencies for particle filters similar to the mini-bag filter (F8, MERV 12) used in this office; reported values cluster in the range of 5−20%.[31−34] Tamas et al. measured an ozone removal of ~5% by a HEPA filter that had been in service on a commercial aircraft for more than a year.[35] Since the face velocity in their study was relatively high (2.3−3.1 m/s), the ozone removal for the loaded HEPA filter in the present study, with a much smaller face velocity, is assumed to be larger. Our combined removal efficiency of 20% is in reasonable agreement with these prior measurements.

The first order rate constants for ozone removal by indoor surfaces (k_d) was 2.8 ± 0.2 h^{-1} (, SI Table S4). The office has a nominal area-to-volume (A/V) ratio of 1.1 m^{-1}, and we estimate its "effective" A/V, including furnishings, to be approximately 2 m^{-1}. Using the latter, we calculate an average value of 0.04 ± 0.01 cm s^{-1} (i.e., 2.8 h^{-1}/2 m^{-1} = 1.4 m h^{-1}) for the deposition velocity (v_d) of ozone to the exposed surfaces in the office. The office surfaces were dominated by marble flooring, low-VOC-painted walls, ceiling tiles and wooden desks. Measured ozone deposition velocities to such surfaces span a range of values: marble flooring, 0.00010−0.027 cm s^{-1};[36] low-VOC-painted walls, 0.01 cm s^{-1};[37] ceiling tiles, 0.13−0.18 cm s^{-1};[38] wooden desks, 0.02−0.38 cm s^{-1}.[39] The average deposition velocity to room surfaces in this study is consistent with these reported values.

The calculated first order rate constants for ozone removal by human surfaces (k_h) was 0.6 ± 0.5 h^{-1} (SI Table S5). The ratio of net occupant surface area to office volume was 0.14 m^{-1}; and hence the calculated deposition velocity (v_h) was 0.12 cm s^{-1} (4.3 m h^{-1}). Ozone deposition velocities to human

DOI: 10.1021/acs.est.6b03069
Environ. Sci. Technol. 2016, 50, 10236−10244

surfaces reported in the literature range from 0.1 to 0.6 cm s^{-1};[16,29,40,41] the present study's value is in this range, but on the lower end. Given variable amounts of skin oil on individuals,[42] different types of clothing, and anticipated differences in near surface airflows, we judge that the value of v_h measured in this study is in reasonable agreement with literature values.

The rate at which the ESP generated ozone (ε, mass/time) was calculated with three different sets of measurements. Using an indoor ozone mass balance model, ε was first calculated when the building was unoccupied, and k_h could be neglected (12:00−14:00) and, second, during working hours when k_h had to be included in the calculation. The calculated value for ε was (25 ± 18) mg h^{-1} using measurements from the unoccupied period (SI Table S6) and (23 ± 10) mg h^{-1} using measurements from working hours (SI Table S7). The third set of measurements used to calculate ε were ozone concentrations in the supply air upstream and downstream of the filter bank when it included the ESP. Using a simple supply air mass balance model and the measurements shown in Figure 2, the calculated value for ε was 29 mg h^{-1}. There is considerable variability in the ozone generation rate obtained applying the indoor ozone mass balance model to either the first or second set of data. This may partially be due to varying relative humidity for the air passing through the ESP, but it also reflects difficulties accurately measuring the indoor ozone concentration, C_{in}, at low concentrations. Nonetheless, it is reassuring that the average results obtained with the mass balance model during unoccupied and occupied conditions are similar to those obtained from ozone measurements in the supply air duct upstream and downstream of the ESP-containing filter bank. Considering that the latter is a more direct way to measure ε, we consider the ozone generation rate of 29 mg h^{-1} the more reliable value.

Previous studies have measured ozone emission rates for ESPs while they were in service. Viner et al. measured emission rates ranging from 20 to 30 mg h^{-1} for three ESPs designed to be used in a building's ductwork.[43] Bowser and Fugler measured emission rates ranging from 13 to 62 mg h^{-1} for unspecified ESPs located in the air distribution ducts in 15 homes.[44] Morrison et al. measured emission rates for a Honeywell F300 ESP and a Trane Clean Effects ESP; the in-field emission rates for these two devices ranged from 3 to 48 mg h^{-1}.[45] Most recently Poppendieck et al. measured emission rates for two ESP units operating in the NIST test house; these units had overall dimensions and charging voltages similar to the unit used in the present study. The reported emission rates were 51 ± 14 and 60 ± 15 mg h^{-1} for Brand 1 with different collection plates, and 22 ± 4 mg h^{-1} for Brand 2.[19]

ESP ozone emission rates depend on multiple factors including the ionization voltage, electrode spacing, plate dimensions, relative humidity and amount of soiling on the collection plates.[19,45] Given the variety of influencing factors, it is somewhat surprising that the emission rates reported in the literature are as clustered as they are. This comparison at least indicates that the emission rate measured in the present study (29 mg h^{-1}) is not unusual, and that it falls within the cluster of values reported for field measurements of similar ESP units.

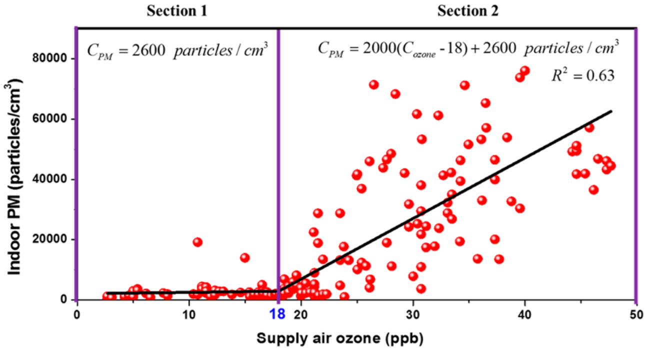


Figure 5. Variation of hourly indoor PM number concentrations with supply air ozone concentrations during occupied hours (excluding hours 12:00 to 14:00).

3.5. Relationship between Supply Air Ozone Concentrations and Indoor PM Number Concentrations. The ozone concentration at the supply air outlet consists of ozone from outdoors as well as that from the ESP when it is present. Figure 5 shows the variation of hourly indoor particle number concentrations with supply air ozone concentrations for all sampling days during working hours (excluding hours 12:00 to 14:00). When the supply air ozone concentration was lower than approximately 18 ppb, indoor PM number concentrations were low and there was no significant correlation between indoor PM number concentrations and supply air ozone concentrations (Spearman correlation: $\rho = 0.12$, $p = 0.38$). When the supply air ozone concentration was greater than 18 ppb, indoor PM number concentrations was significantly correlated with supply air ozone concentrations ($\rho = 0.80$, $p < 0.0005$); the slope of linear fitting between indoor PM and supply air ozone concentration was 2000 (particles/cm^3)/(ppb O_3) with an $R^2 = 0.63$. In other words, it appears that in this particular office environment the ozone level in the supply air must exceed a certain threshold (18 ppb) for the ozone-initiated generation of particles. The fact that a linear relationship was observed in the second part of Figure 5 indicates that the levels of reactive organic compounds in the office were relatively constant and that particle generation was ozone limited rather than organic limited. It also suggests that the indoor mass concentration of airborne particles was not varying substantially, since the SOA yield will vary with the mass concentration of airborne particles.[30] In addition to reactive organics and airborne particle mass, variability in factors such as the air exchange rate and the coagulation and deposition of PM will affect the linear fit. Hence, the scatter seen in Section 2 of Figure 5 is not surprising.

We are aware of two previous studies that have reported an ozone threshold for UFP production. In studies at a primary school in Brisbane, Morawska et al. found that indoor UFP number concentrations did not increase significantly until the indoor ozone concentration was higher than 5 ppb.[46] Rai et al. examined the generation of ozone-initiated particles from reactions with human-worn clothing in an environmental chamber, and found that the UFP number concentrations were negligible when ozone was lower than 5 ppb, but was substantial when the ozone concentration was 22 ppb.[47] The underlying reason(s) for PM generation occurring after ozone concentration exceeded some critical value requires further investigation; enhanced understanding may help with setting up control strategies to reduce indoor PM, especially UFPs from ozone-initiated chemistry.

3.6. Exposure to Ozone and PM Resulting from ESP Operation. The contribution of the ESP to indoor ozone levels can be estimated by mass balance modeling. Let α and β represent the increase, due to ESP operation, in the supply and indoor air ozone concentrations [ppb]. Then, based on eqs 2 and 6, α and β can be expressed as follows:

$$\alpha = \frac{\varepsilon \cdot R}{Q_{\text{supply}}} \cdot \left(1 - \frac{\gamma_{\text{filt}}}{2}\right) \tag{8}$$

$$\beta = \frac{\varepsilon \cdot R}{(\lambda_{\text{v}} + k_{\text{d}} + k_{\text{h}}) \cdot V_{\text{room}}} \cdot \left(1 - \frac{\gamma_{\text{filt}}}{2}\right) \tag{9}$$

Based on eqs 8 and 9, we estimate that ESP operation increased ozone levels in the supply air by 13.5 ppb and ozone levels in indoor air by 3 ppb.

The mean hourly outdoor ozone concentration during working hours (08:00–18:00, excluding 12:00–14:00) from April 1, 2014 to March 31, 2015 in Changsha was (39 ± 24) ppb (SI Figure S4). Based on eq 9, if there was no ESP unit in the filter bank, the mean hourly indoor ozone level during working hours would have been 7 ppb. In other words, during working hours with the ESP operating, the contribution of the ESP to indoor ozone levels was substantial.

In the previous section we presented a linear relationship between supply air ozone concentration and indoor PM number concentration when the supply air ozone concentration was greater than 18 ppb. Based on the equation displayed in Figure 5, the 13.5 ppb increase in supply air ozone resulting from ESP operation during working hours is estimated to increase PM number concentrations by roughly 22 000 particles/cm^3. The estimated hourly indoor PM concentrations without and with the ESP operating during working hours are shown in SI Figure S4. The detailed calculations of the ESP contribution to indoor PM are also shown in the SI.

Numerous epidemiological studies have established associations between an increase in both short and long-term ozone exposure and an increase in morbidity and mortality (refs 48–52 and references therein).[48−52] Daily indoor ozone exposures are estimated to be between 25% and 60% of total daily ozone exposures,[53] and variations in indoor exposures partially explain city-to-city variations in ozone-mortality coefficients.[54] In contrast, there are only a few epidemiological studies exploring the impact of UFP exposures on morbidity and mortality. Such studies are hampered by a dearth of long-term and large-scale measurements of outdoor UFP number concentrations. However, there are some early stage estimates.[55−58] A panel of European experts judged that an increase of 1000 UFP particles/cm^3 was associated with a median increase of 0.3% in all-cause mortality, although there were "substantial differences in the estimated UFP health effect and its uncertainty".[55] A preliminary study in Erfurt, Germany, for the period 1995–1998, reported that a 12 700 particles/cm^3 increase in the UFP number concentration was associated with an increase in mortality of 4.5%.[56,57] Stölzel et al. extended the study period to include the years 1995–2001 and updated the analysis, reporting that a 10 000 particles/cm^3 increase in UFP number concentration was associated with an increase of 2.9% for all-cause mortality and 3.1% for cardiorespiratory mortality.[58] More recently, Lanzinger et al. reported a positive delayed and prolonged, but not statistically significant, association between exposures to UFP in five European cities and respiratory mortality.[59]

In the present study ESP operation increased indoor ozone by approximately 3 ppb and indoor PM number concentration by approximately 22 000 particles/cm^3. While the ozone increase is relatively small, the increase in PM number concentration is within the range of typical outdoor UFP concentrations (~4000–30 000 particles/cm^3).[2,23,59,60] It is also comparable to the interquartile difference observed in the Erfurt study[56−58] that was associated with a statistically significant increase in mortality. The air handling system in the office that was the focus of the present study consists of a mini-bag filter, followed by an ESP unit, followed by a HEPA filter. Given the ozone generated by the ESP and its subsequent impact on indoor PM number concentrations, operating without the ESP appears to be a better configuration for the workers' health. However, this would decrease the lifetime of the HEPA filter. An alternative would be to include a charcoal filter for ozone removal downstream of the ESP.[61−63]

3.7. Limitations of the Study.

Due to both sampling and budget constraints, we did not measure various indoor gas-phase species that react relatively fast with ozone, including terpenes, terpene alcohols, unsaturated fatty acids and nitric oxide. We also did not measure reactive organic compounds on indoor surfaces or associated with settled dust, including occupant-derived compounds such as squalene and 6-hexadec-6-enoic acid. Hence, we cannot develop a more complete picture of the indoor chemistry influencing indoor ozone concentrations and contributing to particle production.

We did not have instrumentation to determine the size distributions of airborne particles. The lack of such information precludes quantitative evaluations of the effects of coagulation and deposition on indoor particle dynamics.

Outdoor ozone concentrations were based on measurements made at a government monitoring site about 4.5 km from the study location. Hence, any local perturbations on outdoor ozone concentrations have been overlooked.

The threshold concentration (18 ppb) that supply air ozone had to exceed prior to a sharp increase in particle production is specific to the conditions for this office. This includes the air exchange rate, temperature, relative humidity, and concentrations of reactive organics on surfaces. While the actual value of the threshold is expected to vary, we suspect that such a threshold occurs in other buildings. This requires verification. Future studies might also be designed to examine the factors that determine the threshold. Such investigations would benefit from continuous size-resolved particle number measurements to sizes as small as 2 nm, time-resolved concentrations of organic and inorganic gas phase species that react with ozone fast enough to impact indoor chemistry and other influencing factors such as air exchange rates, temperature, and humidity.

The quantitative results obtained in this study are specific to the office where the study was conducted. However, the findings support expectations based on other studies and are generalizable to any mechanically ventilated office environment.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b03069.

Additional information as noted in the text (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The research was supported by the Natural Science Foundation of China (Nos. 51420105010, 51478235, 51136002, and 51521005) and the Special Fund for Scientific Research in the Public Interest, Ministry of Environment Protection of China (No. 201409080). We thank the Board Group (Changsha, Hunan Province, China) for providing the offices for this investigation. However, the company played no role in study design and in the interpretation of the results.

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