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# Ultrafine particles generated from coloring with scented markers in the presence of ozone

**Abstract** High concentrations of ultrafine particles (UFPs) have been previously reported during school art activities. This is possibly due to secondary organic aerosols (SOAs) formed from reactions between ozone and volatile organic compounds emitted from art products. Four brands of markers, three scented and one unscented, were tested inside a stainless steel chamber at eight different ozone concentrations between 0 and 300 ppb. Out of the 32 tested markers, only the lemon- and orange-scented markers from one brand reacted with ozone to form UFPs. Limonene, pinene, and several other terpenes were identified as ingredients of ink in SOA-forming markers. Coloring with one lemon-scented marker for 1 min without ozone generated on average approximately 26 ± 4 ppb of limonene inside the chamber. At 150 ppb ozone, using one lemon marker for 1 min formed on average $7.7 \times 10^{10}$ particles. The particle size distribution indicated an initial mode of 15 nm which grew to 40 nm. At 50 ppb ozone and below, no significant SOA formation occurred. The number of particles formed is moderately correlated with the mass of ink used ($R^2 = 0.68$). Based on these data, scented markers are not likely a strong source of SOA under normal indoor ozone levels.

**C.-C. D. Fung, S. Shu, Y. Zhu**

Department of Environmental Health Sciences, Jonathan and Karin Fielding School of Public Health, University of California Los Angeles, Los Angeles, CA, USA

Key words: Ultrafine particles; Secondary organic aerosols; Ozone; Terpene; Exposure assessment.

Y. Zhu
Department of Environmental Health Sciences
Jonathan and Karin Fielding School of Public Health
University of California Los Angeles
Los Angeles
CA 90095-1772
USA
Tel.: +1-310-825-4324
Fax: +1-310-794-2106
e-mail: yifang@ucla.edu

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**Practical Implications**

Children might be exposed to increased concentrations of ultrafine particles during school art activities when using art products containing ozone-reactive terpenes. Testing these products and understanding the important parameters involved will allow teachers and parents to make informed decisions on which art products to use and how to reduce children's exposure to ultrafine particles.

## Introduction

Exposures to ultrafine particles (UFPs) can cause adverse respiratory and cardiovascular effects (Alessandrini et al., 2009; Frampton, 2001). Children, with immature respiratory systems and higher breathing rates per body weight, are particularly susceptible. Indoor sources are a strong contributor to a person's total daily UFP exposures (Wallace and Ott, 2011). For children, who spend approximately 16% of their time in school (Hofferth and Sandberg, 2001), the classroom is an important microenvironment with unique sources of UFPs. Zhang and Zhu (2011) measured a factor of 2.0 increase in particle number concentrations (PNCs) inside classrooms during art activities. Morawska et al. (2009) observed a peak PNC of $5.0 \times 10^4/\text{cm}^3$ inside a classroom (compared with average outdoor PNC of $5.0 \times 10^3/\text{cm}^3$) during children painting and paint drying. They suspected secondary organic aerosols (SOAs) as the source of increased PNCs.

The ozone-initiated reactions with terpenes, both in gas-phase (Atkinson and Arey, 2003) and on indoor surfaces (Shu and Morrison, 2011, 2012), have been studied extensively. These reactions are not only a sink for ozone and terpenes, but more importantly a source of SOAs (Waring and Siegel, 2013; Weschler and Shields, 1999). The reaction mechanism between ozone and terpenes has been detailed elsewhere (Atkinson and Arey, 1998, 2003). Briefly, ozone reacts with the C=C double bonds in the terpene molecules forming criegee intermediates

(CI), which then transform into a series of gas-phase products and can nucleate to form particles. In the indoor environments, each step of this process is affected by many factors, such as the temperature and relative humidity (Fick et al., 2003), air exchange rate (AER), filtration (Fadeyi, 2009), and even the presence of occupants (Fadeyi et al., 2013). The reaction rates of ozone and many different terpenes have also been studied and found to cover three orders of magnitudes (Atkinson et al., 1990; Ham et al., 2006; Harrison and Wells, 2009; Harrison et al., 2007). Among these studied terpenes, *d*-limonene and α-pinene are two ozone-reactive terpenes that are commonly found in household and cleaning products (Coleman et al., 2008). However, similar studies on art products have been limited.

Scented markers possibly contain ozone-reactive terpenes. Manufacturers, under the Federal Hazardous Substances Act (FHSA) and the U.S. Consumer Product Safety Commission (CPSC), are required to evaluate and label their products for hazardous substances (CFR 16 § 1500). The formulation of these scented markers is considered proprietary, and manufacturers can be exempt from listing the chemical composition in their material safety data sheet (MSDS). Furthermore, art products conforming to the American Society for Testing and Materials (ASTM) standard D-4236 can be certified as 'non-toxic' by the Art & Creative Materials Institute. Although many terpenes have little to no toxicity, the oxidation products can include formaldehyde (Hatakeyama et al., 1989; Huang et al., 2011) and SOAs with high intensities of reactive oxygen species (ROS) (Chen and Hopke, 2009, 2010). SOAs can cause upper airway irritation and inflammatory responses in cells, which could exacerbate asthma (Delfino, 2002). However, current standards and regulations do not include SOAs. It is possible that coloring activities using scented markers are contributing to SOAs in the indoor environment. The goal of this paper was to test and characterize different scented markers for their SOA-forming potential.

## Methods

### Experimental

A stainless steel reaction chamber with a volume of 0.32 $m^3$ (11 $ft.^3$) was used to measure SOAs formed when using scented markers in the presence of ozone. The chamber was used as a completely mixed flow reactor (CMFR) and AER was set at 0.66/h, which was determined by the $CO_2$ decay method (Gong et al., 2009). The inlet air to the chamber is climate-controlled laboratory air (20 ± 2°C and 40 ± 10% RH) filtered by a high-efficiency particle air (HEPA) and a mesh charcoal filter. The chamber exhaust was led into the laboratory exhaust system. An ozone-resistant neoprene glove is attached to the chamber to allow the use of the markers in the presence of ozone, to simulate coloring activities inside the chamber. A water-based condensation particle counter (WCPC; TSI 3786, TSI Inc., Shoreview, MN, USA) was used to measure total PNC at 1-s resolution. A scanning mobility particle sizer (SMPS; TSI 3936, TSI Inc.), using the same WCPC, was used to measure particle size distribution from 8 to 290 nm (100 s up scan, 20 s down scan). Ozone was introduced into the chamber from an UV lamp ozone generator (Model 600; Jelight Company Inc., Irvine, CA, USA). A dual beam ozone monitor (OM; Model 205; 2B Technologies Inc., Boulder, CO, USA) was used to measure ozone concentration inside the chamber.

Three different brands of scented markers and one unscented brand were tested for their SOA-forming potential. The three scented brands have been coded as 'A' (12 scents), 'B' (8 scents), and 'C' (11 scents), the names of the scents are given in Table S1. In the study by Zhang and Zhu (2011), art activities were found to usually last for approximately 20 min, during which drawing would occur sporadically. In this study, a 20-min 'coloring activity' was simulated using a marker to color an area of 423 $cm^2$ (18.0 × 23.5 cm) on one sheet of paper. Coloring the area on the paper (called 'color phase') took approximately 1 min (the first minute) of the 20-min coloring activity. A set of three initial screening tests for each marker were conducted at 150 ppb ozone. If a marker scent did not produce SOA, no further experiments were conducted. For the markers that produced SOA in the screening test, experiments were conducted at ozone levels of 0, 35, 50, 75, 100, 150, 200, 250, and 300 ppb. Prior to each experiment, the ozone concentration was allowed to stabilize for at least 30 min. The background PNC inside the chamber was approximately 100/$cm^3$. Markers were weighed before and after each coloring activity to determine the amount of ink used.

### Emission of VOCs

The emission of VOCs was measured in the chamber without the presence of ozone, to avoid the need for specialized instrumentation (Shu and Morrison, 2010) or potential loss of samples due to ozone reaction on the surface of sorbent (Calogirou et al., 1996). The transient concentrations of reactive VOCs in the chamber did not allow sufficient time for active sampling to capture the fluctuation of VOC concentrations in the chamber during each 20-min experiment. Therefore, SUMMA canisters were used to take time-averaged air samples. Two experiments were conducted to measure the time-average VOC mixing ratio (ppb) inside the chamber while coloring with the brand A lemon-scented marker. The 6-L SUMMA canisters and chemical analysis were provided by Atmospheric Analysis and Consulting, Inc. (AAC Inc., Ventura, CA, USA). The canisters were equipped with a pressure regulator set to sample for 20 min. Using new markers, the 1-min coloring activity was conducted inside the chamber without the presence of ozone. A duplicate sample was collected on the second experiment to make sure the canister tests were reproducible. The collected canister samples were analyzed using EPA standard TO-15 method with library search. Equations 1 and 2 were used to estimate the emitted mass of VOCs based on the time-averaged concentrations.

$$\int_0^{20} C_i(0)\exp(-\lambda t)\mathrm{d}t = 20 \times C_{i,\mathrm{TA}} \tag{1}$$

$$M_i = C_i(0) \times V \tag{2}$$

where $C_i$ (0) is the mixing ratio of compound *i* at time 0, $\lambda$ is the air exchange rate in the chamber (0.01 per min), and $C_{i,TA}$ is the time-average mixing ratio of compound *i* determined by SUMMA canister analysis, $M_i$ is the mass of emitted compound *i*, and *V* is the volume of chamber. It should be noted that this calculation is a rough estimation because it assumed (i) an impulsive emission of VOCs at time 0, (ii) the VOC deposition to chamber surface is negligible, and (iii) the SUMMA canister has a constant flow rate.

### Identification of VOCs

To further identify the VOCs (either ozone-reactive or none-ozone-reactive) emitted from markers with different scents, a semiquantitative chemical analysis was conducted. Wicks from the markers were placed in glass flasks individually, and samples were collected by impinging the headspace air through 20 ml of hexane (>99.5%; Sigma Aldrich, Saint Louis, MO, USA) at a rate of 1 l/min in a glass impinger for 20 min. Impinging the headspace gas eliminated the need to remove the marker pigments, which may damage the gas chromatography column. The markers tested included the unscented brand, brand B lemon scent, brand C lemon scent, and brand A orange, lemon, and mint scents. Samples were analyzed by gas chromatography–mass spectrometry (GC–MS, Agilent 6890 GC, 5973N MS; Agilent Technologies Inc., Westlake Village, CA, USA). The separation was achieved by a 30-m HP5MS capillary column at a constant pressure of 5.8 psi. The initial oven temperature of 60°C was held for 1 min and then raised to 280°C at a rate of 10°C per min. Each sample run took 23 min, and scan mode was used on the MS detector. The 10 largest peaks in each sample's MS spectrum were integrated and compared with the standard spectrum library. The peaks with less than 60% library search quality were considered as unidentified.

### SOA formation calculation

SOA formation rates were calculated using a mass balance approach as shown by Equation 3.

$$\frac{\mathrm{d}N(t)}{\mathrm{d}t} = \frac{E(t)}{V} - k_p N(t) \tag{3}$$

Where $E(t)$ represents the SOA formation rate (per second) at time *t*, *V* is the volume of the chamber (cm$^3$), $k_p$ is the particle decay parameter (per second), and $N(t)$ is the PNC in the chamber (per second). SOA formation rate can be calculated by solving for $E(t)$ in Equation 4. The values for $N(t)$ are measured using the CPC, and $\Delta t$ is the time resolution.

$$E(t) = V(N(t) - N(t-\Delta t)e^{-k_p \Delta t}) \tag{4}$$

The particle decay parameter $k_p$ is the sum of AER (per second), particle surface deposition rate $\beta_{dep}$ (per second), and coagulation rate $\beta_{coag}$ (per second). The AER was predetermined using the $CO_2$ decay method, and $k_p$ can be calculated by a regression of the decay portion of the particle concentration curve (Gong et al., 2009). Dividing $E(t)$ by the colored area (cm$^2$) will give the flux *EF* (per cm$^2$/s), which allows comparison per unit area of a colored surface. However, this can be affected by the mass of ink used per unit area. In this study, the coloring activity was controlled by covering a single uniform layer, which may not always be the case during real coloring activities.

## Results and discussion

### SOA formation

$E(t)$ was integrated over the 20-min period to estimate the total number of SOA particles formed for each coloring activity. Figure 1 presents the average number of SOA particles generated from the scented marker brands A, B, C, and the unscented brand. In Figure 1, the results from the non-reactive scents from each brand were combined. Mann–Whitney rank sum test was performed between reactive and non-reactive scents. Test results show a statistical difference in the amount of particles formed by the lemon- and orange-scented marker over the other markers ($P \leq 0.001$). The lemon-scented marker from brand A generated SOA particles $1.2 \times 10^4$ times higher than the non-SOA-forming markers, which were similar to background levels. The orange-scented marker from brand A generated SOA particles 24 times higher. Results suggest not all marker scents contain VOCs that react with ozone to yield SOAs. Furthermore, markers of the same scent (e.g., lemon) from different brands may contain different VOCs (i.e., SOA-forming and

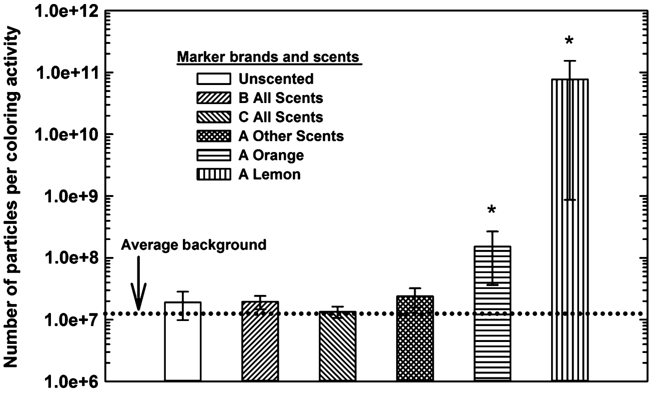

**Fig. 1** Average number of particles formed from the four different brands during the 20-min coloring activity at 150 ppb of ozone is shown. For scented marker brands B and C, all the scents including orange and lemon were combined. The asterisks indicate that particles generated by lemon and orange scents are significantly higher than the other scents from different brands ($P \leq 0.001$)

**Table 1** A summary of the experimental conditions and results for brand A lemon-scented marker: ozone concentrations, number of tests $n$, average particle concentrations $N_{AVG}$, averages of the peak particle concentrations $\bar{N}_{MAX}$, averages of the total number of SOA particles formed $\bar{E}_{TOT}$, and the average fluxes $EF_{AVG}$. Data are presented as average ± s.d.

| Ozone (ppb) | $n$ | $N_{AVG}$ ($\times10^3$/cm$^3$) | $\bar{N}_{MAX}$ ($\times10^3$/cm$^3$) | $\bar{E}_{TOT}$ ($\times10^9$) | $EF_{AVG}$ ($\times10^5$/s/cm$^2$) |
|---|---|---|---|---|---|
| 300 | 3 | 68 ± 25 | 100 ± 30 | 170 ± 60 | 35 ± 12 |
| 250 | 5 | 77 ± 28 | 120 ± 40 | 180 ± 70 | 38 ± 14 |
| 200 | 5 | 63 ± 64 | 95 ± 94 | 160 ± 110 | 31 ± 32 |
| 150 | 14 | 33 ± 33 | 52 ± 51 | 77 ± 80 | 15 ± 16 |
| 100 | 3 | 16 ± 8 | 26 ± 13 | 38 ± 20 | 8 ± 4 |
| 75 | 5 | 0.76 ± 1.10 | 1.30 ± 1.90 | 1.8 ± 2.5 | 0 |
| 50 | 5 | 0.03 ± 0.008 | 0.04 ± 0.02 | 0 | 0 |
| 35 | 5 | 0.06 ± 0.03 | 0.07 ± 0.04 | 0 | 0 |
| 0 | 5 | 0.04 ± 0.005 | 0.10 ± 0.15 | 0 | 0 |

non-SOA-forming). Table 1 summarizes experimental conditions and results for brand A lemon-scented marker on which subsequent discussion will be focused.

Figure 2 shows an example of PNC time series of the lemon marker from brand A and B in 150 ppb ozone. Ozone was not consumed during the experiment using brand B lemon marker (Figure 2b), and no SOA particles were detected. This indicates that brand A lemon marker contained a sufficient amount of ozone-reactive- and SOA-forming compounds, while brand B lemon marker did not. The shape of the *EF* curve (solid gray line) indicates a single-burst formation of SOAs. As the color phase was completed within the first minute, the ozone reaction rapidly consumed the reactive VOCs which resulted in the single-burst profile. Continuous coloring will likely produce a continuous particle formation, but the initial burst will likely be much stronger than the steady state formation.

The size-dependent particle decay parameter $k_p$ was calculated by conducting regression analysis. Particle deposition rate $\beta_{dep}$ and coagulation rate $\beta_{coag}$ were estimated using models developed by Lai and Nazaroff (2000) and Fuchs (1964), respectively. The results of $k_p$, $\beta_{dep}$, and $\beta_{coag}$ for particles from 8 to 290 nm are presented in Figure S1. Based on model results, surface deposition was not a strong contributor to particle decay except for particles smaller than 14 nm. This is due to the low air velocity inside the chamber. Coagulation is a strong contributor for particles from 14 to 35 nm. The model results matched well with the calculated $k_p$ for particles larger than 20 nm. The discrepancies for smaller particles are possibly due to uncertainty in the size distribution measurements. In the particle formation processes, nucleation and growth stages (mode diameters from 15 to 35 nm) are most affected by coagulation. As particle growth stabilizes (35–40 nm), AER becomes the strongest decay mechanism. Using the size-adjusted $k_p$ to calculate particle emission accounts for the effects of coagulation and surface deposition.

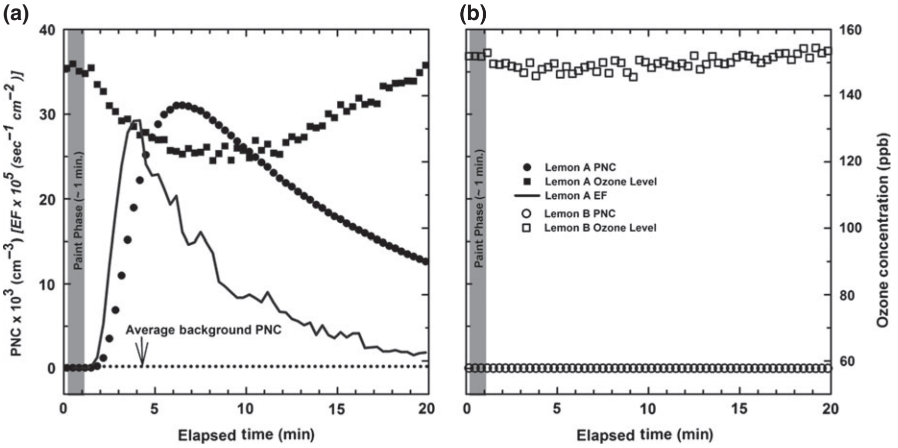

**Fig. 2** Example of particle number concentration and flux profiles of particles formed by lemon-scented marker from brands A and B at ozone levels of 150 ppb during a 20-min coloring activity. In (a), the single-burst concentration profile for the lemon-scented marker from brand A is shown. The flux is represented by the solid gray line. The average background particle concentration is approximately 50/cm$^3$ and is represented by the dotted line. In (b), no ozone was consumed by the lemon-scented marker from brand 'B' and no particles were detected above the background concentration

Surface reactions on the chamber wall are not expected to contribute significantly to SOA formation even with a relatively large surface-area-to-volume ratio (S/V = 5.77/m). The marker ink was applied directly onto a piece of paper. It would require volatilization and then deposition for the limonene to be adsorbed to the chamber walls. The gas-phase reactions are likely to occur before the terpenes can deposit and react with ozone on the stainless steel chamber walls. Limonene directly applied to the paper during coloring activities might form SOAs due to surface ozone reactions. However, this is expected to be less important compared with gas-phase reactions, because (i) the limonene in the marker ink mixture will not saturate the colored surface based on our protocol, and (ii) the high deposition velocity of ozone to paper limits its availability for other reactions (Kleno et al., 2001).

### VOCs identification and quantification results

The chemical analysis conducted on the SUMMA canister samples revealed that one coloring activity can generate a 20-min average mixing ratio of approximately 26 ± 4 ppb of limonene. Other studies have observed SOA formation at 50 ppb ozone at similar limonene concentrations (Chen and Hopke, 2010; Waring et al., 2011). In this study, SOA formation was not observed when the ozone concentration was lower than 50 ppb. It is possible that the response factor used by the AAC laboratory overestimated the limonene concentration because it was not among the list of target VOCs of TO-15 method. Other VOCs detected in the sample include methanol, ethanol, acetone, 2-propanol, and toluene. The concentrations of these compounds are summarized in Table 2 ranging from approximately 1–39 ppb. The second chemical analysis method allowed the identification of other compounds that did not have enough mass to be quantified using the SUMMA canisters. These included the reactive terpenes: 3-carene, camphor, camphene, myrcene, and linalool, as well as none-ozone-reactive fragrance compounds: ethyl butyrate, 4-methyloctane, isoamyl acetate, isoamyl butyrate, isoamyl 2-methyl butyrate, isopulegol, trans-menthone, cis-menthone, cis-1,3 trans-1,4 menthyl, and menthyl acetate.

**Table 2** Summary of the results from the VOC chemical analysis

| | Test 1 | | Test 2a | | Test 2b | |
|---|---|---|---|---|---|---|
| Compounds identified | Mixing ratio (ppb) | Mass emitted ($\mu$g) | Mixing ratio (ppb) | Mass emitted ($\mu$g) | Mixing ratio (ppb) | Mass emitted ($\mu$g) |
| Limonene | 21.7 | 73 | 29.0 | 98 | 28.1 | 95 |
| α-Pinene | 1.6 | 5.0 | 2.1 | 7.0 | 2.1 | 7.0 |
| Methanol | 30.5 | 24 | 37.2 | 30 | 39.3 | 31 |
| Ethanol | 31.7 | 36 | 29.7 | 34 | 30.7 | 35 |
| Acetone | 16.1 | 24 | 18.9 | 27 | 19.2 | 28 |
| 2-Propanol | 7.7 | 11 | 6.0 | 9.0 | 6.3 | 9.0 |
| Toluene | 1.1 | 3.0 | 1.3 | 3.0 | 1.2 | 3.0 |

### Particle size distribution

Figure 3 shows an example of the size distribution of the SOA particles generated from the lemon-scented marker. The contour plot indicates a unimodal size distribution with an initial mode of approximately 15 nm. The particle concentration peaks within a minute after coloring begins and proceeds to decay, during which the particle mode grows to and stabilizes at approximately 40 nm. Coleman et al. (2008) characterized the formation and growth of SOA particles in four stages. Stage 1 is when a nucleation burst occurs as a result of ozone added to a steady concentration of reacting vapor. Stage 2 is characterized by the growth of the particles formed in stage 1. In stage 3, nucleation resumes while the particles from the initial event remain. In stage 4, a constant or oscillating particle concentration occurs. In this study, only stages 1 and 2 were observed which was possibly because the amount of reactive VOCs from the markers was finite. Stages 3 and 4 would only occur when a continuous supply of reactive VOC and ozone is present.

### Effects of ozone concentration and marker usage

Results on the effects of ozone concentration on SOA formation from the scented markers are shown in Figure 4. The number of SOA particles increases with the ozone concentration. However, ozone levels above 200 ppb did not significantly increase the number of SOA particles. This is due to the amount of reactive VOCs being limited by the amount of marker ink used. The slight decrease in particles emitted at 300 ppb ozone is likely due to the effect of marker age (as

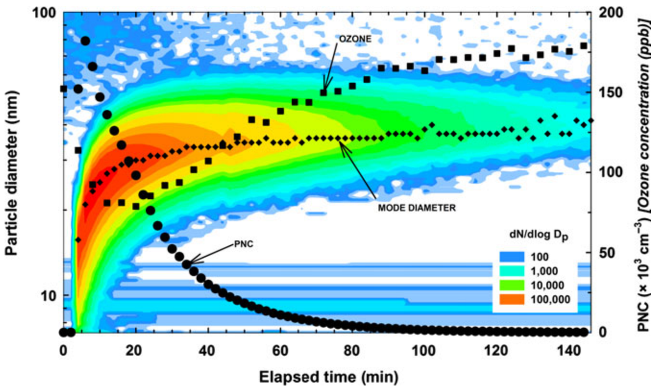

**Fig. 3** Particle size distribution contour plot with particle concentration and ozone depletion profile overlay. Contour plot demonstrates the initial burst of nucleation followed by particle growth and decay in the chamber. The black dots indicate the mode of the particle size distribution. The particle mode at the peak particle concentration is approximately 25 nm and stabilizes to approximately 40 nm

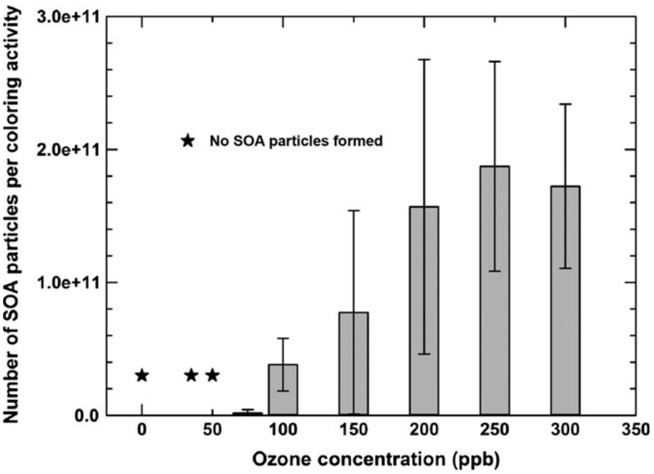

**Fig. 4** Effect of ozone concentration on the average number of particle formed from lemon marker A during coloring activities. Ozone concentrations higher than 200 ppb had little effect on the number of particles formed. Particle formation decreases with ozone concentration; no particles were detected at ozone levels of 50 ppb and lower

discussed further in Figure 5) as the 300 ppb experiments were conducted near the end of the marker's useful life.

No SOA formation was detected at 50 ppb ozone and below. Other studies also observed a decrease in SOA particle number concentration with a decrease in ozone concentration (Coleman et al., 2008; Waring et al., 2011). However, Waring et al. (2011) observed SOA formation from 50 ppb of *d*-limonene and 25 ppb ozone. The average limonene concentration approximately 26 ± 4 ppb was measured when a new marker was used. It is possible that the limonene concentrations during the SOA formation experiments, which were not measured, were much lower.

During experiments, the measured PNC was observed to decrease as the lemon marker was used more times. To examine this effect, the number of SOA particles was tracked with the number of times a marker was used. Because the data included experiments conducted at 150, 200, 250, and 300 ppb ozone levels, the PNC needed to be corrected to the same ozone level. This was accomplished using the ratios of PNCs at 150 ppb ozone to those at 200, 250, and 300 ppb ozone, from the data in Figure 4, as a correction factor. In Figure 5a, mass of ink used is plotted against the corrected PNCs. The linear regression demonstrates a moderate correlation ($R^2 = 0.68$) between mass of ink used and number of SOA particles formed. In Figure 5b, the mass of ink and the corrected PNCs are plotted against the number of times the marker is used. Both ink mass and corrected PNCs display negative slopes that are statistically significant ($P < 0.05$). These results suggest as a marker was used more, the ink was transferred onto the paper less efficiently. Therefore, fewer particles would be formed for older markers.

### Implications for UFP exposures

Two previous studies have found increased UFP concentrations in classrooms during art activities (Morawska et al., 2009; Zhang and Zhu, 2011) and use of scented markers containing *d*-limonene is a possible contributing source. Based on the results from this study, scented markers alone may not be a strong source of secondary UFPs under normal indoor conditions. The 20-min average mixing ratio of limonene emitted from the use of a lemon-scented marker in this study was approximately 26 ± 4 ppb which did not lead to SOA formation when ozone levels were below 50 ppb. In a real classroom setting, this concentration can vary. The classroom in the study by Zhang and Zhu (2011) has an approximate volume of 85 $m^3$ and an AER of 0.45/h. Assuming 20 students all use the brand A lemon marker to color for 1 min, the 20-min average mixing ratio of limonene in the classroom will only be approxi-

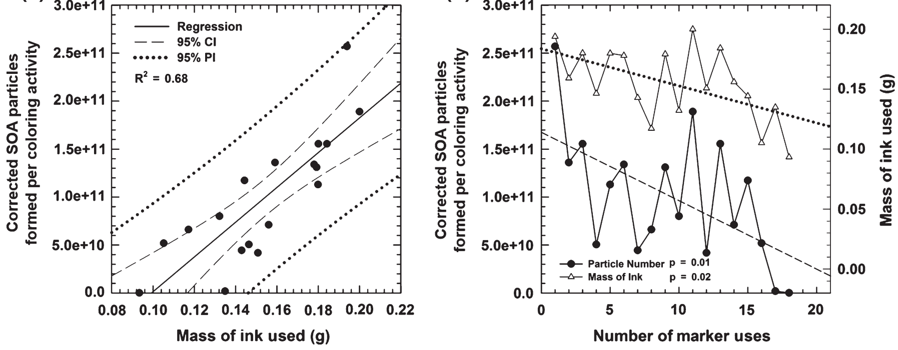

**Fig. 5** Effect of usage on the particle formation from brand A lemon-scented marker. In (a), the mass of ink used is plotted against the corrected number of particles formed. The number of particles formed demonstrates a moderate correlation ($R^2 = 0.68$) with the mass of ink used. In (b), the corrected number of particles formed during a coloring activity and the ink mass used is plotted against the number of marker uses. The closed circles represent the number of particles, and the open triangles represent the mass of ink used for each corresponding experiment. The regression lines show a negative slope for mass of ink used ($P = 0.02$) and number of particles formed ($P = 0.01$)

mately 1.8 ppb. This concentration is 14 times lower than what was measured in the chamber and is unlikely to have major contributions to SOA formation. The study by Morawska et al. (2009) found increased particle concentrations when children were using paint and while the paint was drying. This suggests that other art products such as paints and glues, which were not tested in this study, may contain SOA-forming compounds and need to be tested in future studies.

Another important factor in SOA formation is the indoor ozone level. The biggest contributor to indoor ozone is penetration from the ambient air. Studies have found a wide range of ozone indoor/outdoor ratios (Stephens et al., 2012; Weschler, 2000). In addition, sources such as printers, copiers, ion generators, and air purifiers can increase indoor ozone levels, which may lead to the formation of UFPs and other gaseous pollutants such as formaldehyde (Alshawa et al., 2007; Waring et al., 2008; Weschler, 2000). One animal study has found inflammatory effects in rats when exposed to ozone/limonene reaction products (Sunil et al., 2007). However, a recent animal study found that ozone/limonene reaction products do not promote allergic inflammatory effects in the mice respiratory system (Hansen et al., 2013). Such contradicting findings suggest that more research is needed to better understand the health effects of exposure to indoor SOAs and other ozone/terpene reaction products.

## Conclusions

In this study, three brands of scented markers were tested inside a stainless steel chamber at different ozone levels for their SOA formation potential. At ozone levels above 50 ppb, only the lemon- and orange-scented markers from brand A formed SOA particles. Chemical analysis revealed these two markers contained limonene and other terpenes. Coloring with brand A lemon-scented marker for 1 min resulted in a limonene mixing ratio of approximately 26 ± 4 ppb in the chamber air on a 20-min average. This low limonene level supports the finding that at low ozone concentrations SOA formation were not detectable. This study suggests that scented markers are not a strong source of SOA under normal indoor ozone levels. Other art products such as paints need to be tested to evaluate their SOA-forming potential. Finally, the findings from this study raise concerns about other consumer products that are not previously recognized as forming SOAs.

## Acknowledgements

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## Supporting Information

Additional Supporting Information may be found in the online version of this article:

**Figure S1.** Calculated Size Dependent Particle Decay Parameter $k_p$.
**Table S1.** Names of the Scents for Marker Brands A, B, and C.

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