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# Outdoor fine and ultrafine particle measurements at six bus stops with smoking on two California arterial highways—Results of a pilot study

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*As indoor smoking bans have become widely adopted, some U.S. communities are considering restricting smoking outdoors, creating a need for measurements of air pollution near smokers outdoors. Personal exposure experiments were conducted with four to five participants at six sidewalk bus stops located 1.5–3.3 m from the curb of two heavily traveled California arterial highways with 3300–5100 vehicles per hour. At each bus stop, a smoker in the group smoked a cigarette. Gravimetrically calibrated continuous monitors were used to measure fine particle concentrations (aerodynamic diameter  $\leq 2.5 \mu\text{m}$ ;  $\text{PM}_{2.5}$ ) in the breathing zones (within 0.2 m from the nose and mouth) of each participant. At each bus stop, ultrafine particles (UFP), wind speed, temperature, relative humidity, and traffic counts were also measured. For 13 cigarette experiments, the mean  $\text{PM}_{2.5}$  personal exposure of the nonsmoker seated 0.5 m from the smoker during a 5-min cigarette ranged from 15 to 153  $\mu\text{g}/\text{m}^3$ . Of four persons seated on the bench, the smoker received the highest  $\text{PM}_{2.5}$  breathing-zone exposure of 192  $\mu\text{g}/\text{m}^3$ . There was a strong proximity effect: nonsmokers at distances 0.5, 1.0, and 1.5 m from the smoker received mean  $\text{PM}_{2.5}$  personal exposures of 59, 40, and 28  $\mu\text{g}/\text{m}^3$ , respectively, compared with a background level of 1.7  $\mu\text{g}/\text{m}^3$ . Like the  $\text{PM}_{2.5}$  concentrations, UFP concentrations measured 0.5 m from the smoker increased abruptly when a cigarette started and decreased when the cigarette ended, averaging 44,500 particles/ $\text{cm}^3$  compared with the background level of 7200 particles/ $\text{cm}^3$ . During nonsmoking periods, the UFP background concentrations showed occasional peaks due to traffic, whereas  $\text{PM}_{2.5}$  background concentrations were extremely low. The results indicate that a single cigarette smoked outdoors at a bus stop can cause  $\text{PM}_{2.5}$  and UFP concentrations near the smoker that are 16–35 and 6.2 times, respectively, higher than the background concentrations due to cars and trucks on an adjacent arterial highway.*

*Implications:* Rules banning smoking indoors have been widely adopted in the United States and in many countries. Some communities are considering smoking bans that would apply to outdoor locations. Although many measurements are available of pollutant concentrations from secondhand smoke at indoor locations, few measurements are available of exposure to secondhand smoke outdoors. This study provides new data on exposure to fine and ultrafine particles from secondhand smoke near a smoker outdoors. The levels are compared with the exposure measured next to a highway. The findings are important for policies that might be developed for reducing exposure to secondhand smoke outdoors.

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## Introduction

With widespread adoption of indoor smoking bans, there has been considerable interest by local communities in restricting smoking outdoors. However, few data exist on air pollutant concentrations near a smoker outdoors to support these policies, and there are no published data comparing secondhand smoke exposure at sidewalk bus stops with the levels at these locations due to traffic.

Several studies of exposure to secondhand smoke outdoors show that high concentrations can occur close to a smoker. Repace (2005) conducted experiments outdoors in a circle of up to 10 smokers on the University of Maryland campus and estimated that particle concentrations were inversely related to distance from the smokers over distances from 1.5 to 5 m.

Klepeis et al. (2007) measured concentrations of fine particles or  $\text{PM}_{2.5}$ —particles smaller than 2.5  $\mu\text{m}$ —on 15 visits to 10 outdoor locations, including parks, sandwich shops, and the patios of restaurants and pubs in California. At an outdoor restaurant patio, the mean  $\text{PM}_{2.5}$  concentrations measured 0.5 m from an active smoker sometimes exceeded 200  $\mu\text{g}/\text{m}^3$  during the cigarette smoking period, and transient concentrations occasionally exceeded 1000  $\mu\text{g}/\text{m}^3$ . Their experiments indicated that outdoor concentrations decreased with distance from an active cigarette over distances from 0.25 to 2 m. Stafford et al. (2010) measured  $\text{PM}_{2.5}$  concentrations in alfresco (outdoor sitting) areas of pubs, restaurants, and public places and found that outdoor concentrations significantly increased when at least one smoker was present. St. Helen et al. (2011) measured exposure to  $\text{PM}_{2.5}$  and carbon monoxide (CO) from secondhand

smoke outdoors at waiting areas and patios of 5 restaurants and bars in downtown Athens, Georgia, and they found that  $PM_{2.5}$  concentrations were associated with the number of smokers present and were a better marker of outdoor secondhand smoke than CO. Klepeis et al. (2009) conducted 100 controlled experiments releasing a CO tracer gas and using up to 36 CO monitors outdoors to measure the outdoor “proximity effect”—the decrease in concentration with distance from a source, including the effect of winds. Cameron et al. (2010) measured  $PM_{2.5}$  concentrations outdoors at restaurant tables in Melbourne, Australia, and found that levels increased by an average of  $27.3 \mu\text{g}/\text{m}^3$  during the smoking period 1 m from a smoker, with a maximum peak of  $484 \mu\text{g}/\text{m}^3$ . López et al. (2012) measured  $PM_{2.5}$  and nicotine in indoor and outdoor hospitality sites in eight European countries, and Licht et al. (2013) reviewed the research literature on studies measuring  $PM_{2.5}$  concentrations from secondhand smoke outdoors. These studies of outdoor settings, although few in number, show that relatively high air pollutant concentrations can occur outdoors close to a smoker and that outdoor exposure can be significant. However, more research is needed to determine how smoking affects outdoor exposures in real settings and the role that other nearby sources play.

Emerging health research indicates that short-term, elevated exposures to secondhand smoke can have adverse physiological effects on humans. Pope et al. (2009) reported that  $PM_{2.5}$  in the outdoor air and  $PM_{2.5}$  from secondhand smoke appear to have similar toxicity. Pope et al. (2001) found that a mean exposure to  $53 \mu\text{g}/\text{m}^3$  from secondhand smoke for 1.75 hr caused a decrement in heart rate variability of 2.3% for each  $10 \mu\text{g}/\text{m}^3$  increase in  $PM_{2.5}$ .

Barnoya and Glantz (2005) state, “In many cases, the effects of even brief (minutes or hours) passive smoking are nearly as large as those from chronic active smoking.” Their review of the cardiovascular effects of secondhand smoke indicates that increases in aortic stiffness were observed after just 4 min of exposure to secondhand smoke, and exposure for 20 min was associated with activation of blood platelets, which could damage the lining of the arteries and facilitate atherosclerosis. Several researchers have reported adverse physiological effects associated with short-term exposure to secondhand smoke, such as damage to the endothelium, the first inner layer of the arteries that is in contact with the blood (Heiss et al., 2008; Argacha et al., 2008; Frey et al., 2012; Surgeon General, 2010).

One of many sources of exposure to ultrafine particles (UFP) is secondhand tobacco smoke (Wallace and Ott, 2011). Because of their very small size, UFP have a higher predicted deposition rate in the human respiratory tract than larger particles (Daigle et al., 2003; Obersdörster and Utell, 2002). These extremely small particles can penetrate deeper into lung tissue than fine or coarse particles, and they have the largest surface area per unit mass (Nel, 2005), with the serious potential for causing adverse cardiovascular effects. UFP also evades clearance in the lung by alveolar macrophages (Frampton, 2007).

The most recent report of the Surgeon General (2010) concluded that exposure to secondhand tobacco smoke leads to a rapid and steep increase in endothelial dysfunction and inflammation, which is implicated in acute cardiovascular events and

thrombosis. Although there is no direct evidence that short-term exposure to secondhand smoke for periods less than 1 hr could cause an acute heart attack, the National Academy of Sciences stated in 2010 that the circumstantial evidence of the health effects of short-term exposure is both complete and biologically plausible (National Academy of Sciences, 2010).

In California, the motor vehicle is the dominant mode of transportation, but most communities also have a bus service, and some towns and cities have a rail service as well. San Mateo County, with a population of 730,000, operates 296 buses that serve customers throughout the county and parts of San Francisco and Palo Alto in adjacent counties. The largest bus travel corridor passing through the county consists of 37 km (23 miles) of El Camino Real (State Highway No. 82), a six-lane arterial highway with three lanes in each direction that carries 22,800 bus passengers per weekday. Smoking is not restricted at these outdoor bus stops.

Although smoking is allowed on the sidewalk and on benches while waiting for a bus, no measurement data are available on the exposure a nonsmoker receives while sitting or standing near a smoker at a roadway bus stop. We evaluated four main hypotheses in this study: (1) a proximity effect occurs, with persons closer to the smoker receiving higher mean exposure to  $PM_{2.5}$  than persons farther away; (2) personal exposures to  $PM_{2.5}$  and UFP during the smoking periods are greater than the background concentrations caused by nearby traffic during the nonsmoking periods; (3) the time-series plots of personal exposures show that during smoking persons are exposed to multiple *microplumes*, or many extremely intense concentration peaks lasting a few seconds; (4) the frequency distributions of the exposures, which consist largely of *microplumes*, are approximately lognormal.

## Methods

We conducted exposure measurement experiments at six bus stops along a 5.6-km (3.5-mile) stretch of El Camino Real and also on a second four-lane arterial highway, Woodside Road (State Highway No. 84), which intersects El Camino Real. There was approximately one bus stop per 0.5 km (0.3 mile) on each side of the segment of the El Camino Real highway that we studied. We did not measure the variability between smokers or between brands of cigarettes, but instead we used one smoker and a Marlboro Gold (formerly Marlboro Light) brand of cigarette, the most popular brand in the United States, with a 40% market share, and we used three to four nonsmokers close-by equipped with breathing-zone personal monitors. This methodology expanded upon the approach of Klepeis et al. (2007) in which a cooperating smoker smokes a cigarette in the outdoor location under study. Their study used a single real-time AM510 SidePak monitor (TSI, Inc., Shoreview, MN, USA) to measure  $PM_{2.5}$  concentrations at fixed points at various distances from the smoker at outdoor patio cafes. Our approach was similar to the approach subsequently developed by Acevedo-Bolton et al. (2013) that included multiple nonsmoking subjects along with the smoker, each wearing a real-time SidePak personal monitor with the intake probe located less than 0.2 m of their breathing zone to measure their personal exposure to  $PM_{2.5}$  in indoor and outdoor locations. That study consisted of locations considerably different from the ones in the

present investigation, such as sitting at a card table with a smoker, sitting on a sofa in a home next to a smoker, and sitting at a park bench or picnic table with a smoker. In the current study next to highways, we also included a portable real-time condensation particle counter to measure ultrafine particles. Stieb et al. (2008) reported using a similar activity-based approach to measure the exposures of persons engaging in scripted activities in downtown Toronto.

## Instruments and measurements

The TSI AM510 SidePak monitor is a real-time laser photometer designed to measure particle mass concentrations. It arrives from TSI, Inc., factory-calibrated for the reference aerosol, “ISO 12103-1, A Test Dust” (“Arizona Road Dust”), and the factory default setting of the monitor’s default custom calibration factor (CF) for the road dust sample is  $CF = 1.0$ . The SidePak’s User Guide (TSI, 2006, 2013a) recommends that investigators calibrate the monitor for the particular aerosol under study. In a previous laboratory quality control study, Jiang et al. (2011) conducted quality assurance experiments with up to 17 SidePak monitors side-by-side using a real smoker as the source, comparing the results in a chamber with a filter-based gravimetric sampler. The present study used the same SidePak monitors that were used in that study. In practice, we set the internal custom calibration factor of each SidePak monitor to  $CF = 1.0$ ; then we applied the proper calibration factor based on the quality assurance study by Jiang et al. (2011) by rescaling the data for each monitor in the data analysis phase. A TSI 2.5- $\mu\text{m}$ -size impactor was installed at the intake of each SidePak monitor, which can measure particles down to a diameter of 0.1  $\mu\text{m}$  (TSI, 2013a). Although the size range of secondhand smoke particles is 0.02–2  $\mu\text{m}$ , about 90% of the particle mass is between 0.1 and 0.5  $\mu\text{m}$  (Klepeis et al., 2003). Jiang et al. (2011) found a consistent relationship between the SidePak readings and known mass concentrations of tobacco smoke measured using a gravimetric, pump-driven sampler with filters weighted on a precision laboratory scale.

The calibration study by Jiang et al. (2011) compared the SidePak monitor with gravimetric sampling for several types of aerosols: cigarette smoke, incense sticks, burning wood chips, toasted bread, and ambient air. The strong linear correlation between the gravimetric filter-based measurements and the SidePak monitor measurements gave  $R^2$  values all above 0.995. For secondhand smoke from a real smoker, the SidePak custom calibration factors for 17 different monitors ranged from  $CF = 0.25$  to  $CF = 0.31$ , with a mean of 0.29 ( $SD = 0.02$ ). Comparing the results with similar measurements 1 yr later, they found the calibration factors of the monitors were stable over the year. Their results for tobacco smoke were consistent with a gravimetric comparison reported by Lee et al. (2008), who reported a calibration factor of 0.295 for a SidePak monitor with secondhand smoke as the source. We used the same SidePak monitors described in our published quality assurance study (Jiang et al., 2011), and we applied the same calibration factors: SP-4 ( $CF = 0.25$ ), SP-6 ( $CF = 0.305$ ), SP-7 ( $CF = 0.295$ ), SP-8 ( $CF = 0.29$ ), SP-16 ( $CF = 0.26$ ), and SP-17 ( $CF = 0.26$ ). A published SidePak calibration factor for highway emissions was not

directly available, so we used the same secondhand smoke calibration factors for each monitor during both the smoking and nonsmoking periods. Zhang and Zhu (2010) reported a calibration factor of  $CF = 0.42$  for concentrations measured inside a school bus using a TSI DustTrak monitor, which is similar in design to the TSI SidePak monitor, and Chung et al. (2001) reported  $CF = 0.33$  for the DustTrak in ambient air. We found that applying the much higher factory default calibration factor of  $CF = 1.0$  still gave relatively low mean background  $\text{PM}_{2.5}$  concentrations measured on the sidewalks in our study,  $<10 \mu\text{g}/\text{m}^3$ , and applying the calibration factor of Zhang and Zhu (2010) gave mean sidewalk concentrations  $<4 \mu\text{g}/\text{m}^3$ .

Each participant, including the smoker, wore a SidePak continuous  $\text{PM}_{2.5}$  monitor to measure personal exposure, with the monitor’s sampling intake probe located within 0.2 m of the person’s breathing zone. An acrylic plastic holder was designed with a plastic mount, and a necklace chain worn around the neck held the monitor’s intake tube in place, as described in Acevedo-Bolton et al. (2013). For each participant, the intake in the breathing zone was connected by Tygon tubing (outside diameter = 0.95 cm) to the SidePak monitor’s inlet, and the SidePak monitor was attached by a bracket to the person’s belt. For the smoker, the measured self-exposure reflected only the  $\text{PM}_{2.5}$  concentrations in the smoker’s breathing zone, not the smoker’s directly inhaled mainstream smoke from the cigarette. Prior to beginning each bus stop experiment, the SidePak monitors were synchronized in time, each monitor was zeroed with a zero filter from the manufacturer, and the data logging interval was set to 1 sec.

To measure ultrafine particles (UFP), we used a portable, battery-powered model 3007 condensation particle counter (TSI). Because this instrument was larger than the SidePak monitor, it was not set up as a personal monitor. Instead, it was held on the lap of the person sitting closest to the smoker, approximately 0.5 m from the smoker. This monitor uses isopropyl alcohol to form a supersaturated vapor that condenses around ultrafine particles, causing them to grow large enough to be detected by a laser, and it detects particles between 10 nm and 1  $\mu\text{m}$  in diameter (Hämeri et al., 2002; TSI, 2013b). The manufacturer states that the accuracy of the TSI 3007 monitor for particle counts up to 100,000 particles/ $\text{cm}^3$  is about  $\pm 20\%$ . The battery can power the monitor for 5–10 hr, and the isopropyl alcohol lasts for a comparable time period. The data logger can store about 50,000 data points. We set the sampling intervals of the TSI 3007 monitor to 1 sec, and we closely synchronized all the SidePak AM510 monitor times with the TSI 3007 monitor time. We used a precision clock and handwritten notes on a log sheet to record the starting and ending times of each cigarette and other details as accurately as possible at each bus stop.

The TSI 3007 UFP monitor arrives factory-calibrated, and its flow rate was within 5% of 0.71 m/sec. Wallace and Ott (2011) report a quality control comparison between two TSI-3007 monitors run side-by-side for 100 hr to determine their precision and relative bias. They found that the relative bias was 1–3%, and the mean of their relative precision, calculated as the sum of the absolute value of the differences divided by the sum of each measured pair, was 2.3%. In their experiment with 22,973 1-sec



readings from collocated monitors, they reported the coefficient of determination between the two monitors of  $R^2 = 0.998$ .

Wallace and Ott (2011) reported that the bulk of the particle size distribution of tobacco smoke by number count lies between 0.01 and 0.45  $\mu\text{m}$ , which is within the 0.01–1.0  $\mu\text{m}$  size range that the TSI-3007 measures (Hämeri et al., 2002; TSI, 2013b). Other investigators using the TSI-3007 condensation particle counter in and near traffic have referred to their measurements as “ultrafine particles” (Westerdahl et al., 2005; Jarjour et al., 2013; Quiros et al., 2013); although this instrument includes counts above the UFP range ( $>0.1 \mu\text{m}$ ), we will use the term UFP in this paper to refer to these measurements.

We used a model 8386 VelociCalc-Plus (TSI) to measure and log the wind speed, temperature, and relative humidity. Although this thermal (“hot wire”) anemometer can measure wind speeds as low as 0.01 m/sec, it does not measure wind direction; our acoustic anemometer that measures both wind speed and direction was too large to use in the confined space of a highway bus stop. We measured wind speeds at a seated person’s breathing height (1 m). We visually observed the smoke plumes from the burning cigarettes to estimate whether a fifth person, who was standing during the experiments, was upwind or downwind of the smoker.

## Study protocol and sampling locations

In the present study, four to five subjects including one smoker participated in exposure experiments at six bus stops on the sidewalks of two major California arterial highways with heavy traffic in the towns of Redwood City, Menlo Park, and Palo Alto. All participants in these studies signed consent forms approved by Stanford University’s Institutional Review Board for research involving human subjects. At each bus stop, the group measured  $\text{PM}_{2.5}$  personal exposures, ultrafine particle (UFP) concentrations, wind speed, temperature, and relative humidity, and made traffic counts. The weather variables

measured using the VelociCalc-Plus instrument are listed as means over the time period of the visit to each bus stop in Table 1. Traffic volume was estimated by visually counting passing vehicles during 1-min time intervals, with each 1-min interval timed by a stopwatch. This approach resulted in 7–11 individual 1-min traffic counts distributed over the measurement visit of about 1 hr, and the mean of these 1-min counts was then multiplied by 60 to give the estimated traffic volume in vehicles/hr (Table 1). At nearly every bus stop, four persons including the smoker sat at the same positions on the bus waiting bench wearing the  $\text{PM}_{2.5}$  personal monitors, with the smoker sitting at one end. At some bus stops, a fifth standing person stood near the smoker (see configuration in Figure 1). At each bus stop, we selected time periods when no other persons or bus passengers were present, and the morning starting times for our experiments ranged from 10:30 a.m. to noon, whereas the afternoon starting times ranged from 2:00 p.m. to 4:00 p.m. The weather was sunny and clear for all experiments, which were conducted in June.

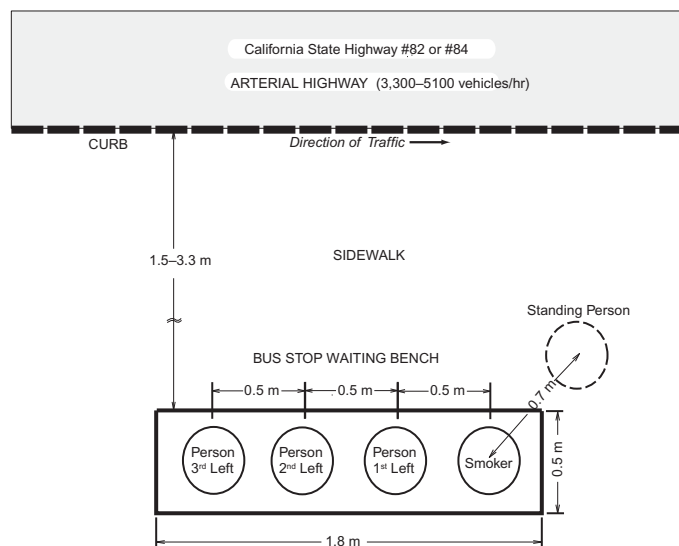
On a 5.6-km (3.5-mile) stretch of El Camino Real, there were a total of 23 bus stops, 11 on one side of the highway and 12 on the other side, or about one bus stop per 0.5 km (0.3 miles) on each side of the road. Of these 23 bus stops, 17 consisted of only a signpost, whereas 6 consisted of a waiting bench with a signpost. On El Camino Real, we selected four bus stops with benches, and on Woodside Road, we selected two additional bus stops with benches. The El Camino Real highway is oriented approximately east–west, and Woodside Road is oriented approximately north–south. Largely residential neighborhoods surround the highways on both sides, and no other major highways are within 0.5 km (0.3 miles) of either of these two arterial highways.

The state of California installs traffic counting machines routinely to measure the *average annual daily traffic* (AADT) volume on state highways. According to the state’s most recent report based on these traffic counts, Woodside Road (State Highway No. 84) had an AADT of 77,000 vehicles, and El

**Table 1.** Locations and characteristics of 6 bus stops in the present study

Bus Stop	Location of Bus Stop	City <sup>a</sup>	Distance from Curb (m)	Traffic Volume <sup>b</sup> (vehicles/hr)	Temp. <sup>c</sup> (°C)	RH <sup>c</sup> (%)	Wind Speed <sup>c</sup> (m/sec)
A	Woodside Rd. near Kentfield Ln.	RC	2.1	3280, <sup>d</sup> 4620	19.4	55	0.74 (SD 0.54)
B	Woodside Rd. near Atherwood Ave.	RC	3.3	3312	21.1	43	1.26 (SD 1.20)
C	El Camino Real near Quarry Rd.	PA	2.7	3480	26.0	49	0.60 (SD 0.52)
D	El Camino Real near Center St.	RC	2.0	4416	20.9	53.8	1.13 (SD 0.60)
E	El Camino Real near Cambridge Ave.	MP	1.5	5100	—	—	—
F	El Camino Real near Encinal Ave.	MP	1.8	3740	17.9	54.7	0.44 (SD 0.37)

Notes: <sup>a</sup>Names of towns: RC = Redwood City; PA = Palo Alto; MP = Menlo Park. <sup>b</sup>Estimated from 7–11 1-min visual vehicle counts during each experiment (see text). <sup>c</sup>Means of TSI VelociCalc-Plus measurements of weather data during each experiment. <sup>d</sup>Nonstandard position for first cigarette: smoker second from end; a separate traffic volume was obtained. The standard position (smoker at end of bench) was used for the 12 remaining cigarettes.



**Figure 1.** Scale drawing showing positions of four to five persons at bus stops along the two California arterial highways.

Camino Real (State Highway No. 82) had an AADT of 61,500 vehicles in 2010 (U.S. Department of Transportation, 2010). Both highways have traffic during most the day for commuting, commerce, and shopping.

Each bus stop we studied included a 1.8 m × 0.5 m public bench for people to sit on while they wait for the bus, and the distances from the bench to the highway curb ranged from 1.5 to 3.3 m (Figure 1). When we measured the horizontal distances between the seated persons, we found their breathing zones (the nose and mouth) were 0.5 m apart while seated on the bench. Except for the first experiment at Bus Stop A (Table 1), the smoker always sat at one end of the bench, and three nonsmokers sat on the same bench next to the smoker. In 5 of the 13 cigarette experiments, a fifth nonsmoker wearing a SidePak personal monitor stood on the sidewalk 0.7 m from the smoker sitting on the bench. In five of the six bus stops, the nonsmokers were positioned left of the smoker facing the roadway, and traffic passed them in the following order: third left, second left, first left, smoker, and then the standing person. At one site (Bus Stop D), we reversed the order to see if traffic direction influenced the exposures. At another site (Bus Stop F), the bench was too short and could not accommodate all four persons, so there were two standing nonsmokers at each end of this bench.

We followed the same protocol at every bus stop: After everyone was seated, we waited a short time to allow measurement of the background concentrations (>5 min). Next, the smoker lit a Marlboro Gold cigarette and smoked it in the normal manner, with the starting and ending times of the cigarette recorded by hand on a data log sheet. After the first cigarette ended, we waited approximately 5 min to allow the background concentrations between the two cigarettes to be measured, and then the smoker lit a second cigarette. The second cigarette was followed by a third waiting period (>5 min) for measuring the background concentration. Thus, there were three background time periods—before smoking, between the two cigarettes, and after the

last cigarette—measured simultaneously using the four to five SidePak personal monitors.

## Results and Discussion

We performed comparisons of the  $PM_{2.5}$  exposures of all persons at the bus stop, comparisons of the individual exposures during smoking with the background concentrations during non-smoking periods, and comparisons of the  $PM_{2.5}$  exposures with the UFP concentrations during smoking and nonsmoking periods.

### Personal exposure to $PM_{2.5}$

A total of 13 cigarettes were smoked on seven visits to six bus stops (Tables 1 and 2). The  $PM_{2.5}$  exposures measured during the smoking periods showed considerable variation from cigarette to cigarette due to the random effects of wind speed, wind direction, turbulence, and other factors, which were expected based on results from an earlier outdoor experimental proximity study using a controlled tracer gas as the emission source (Klepeis et al., 2009). Despite this variability, the arithmetic mean exposure of the nonsmokers in the 13 smoking experiments decreased monotonically with distance from the smoker, showing a clear proximity effect (Table 2).

Each person's mean exposure to  $PM_{2.5}$  during the smoking period was much higher than the overall mean background concentration of  $1.7 \mu\text{g}/\text{m}^3$  (Table 2). The mean concentration measured in the breathing zone of the smoker while smoking ( $192 \mu\text{g}/\text{m}^3$ ; not mainstream smoke) was 113 times the background concentration, whereas the mean personal exposure of the nonsmoker sitting 0.5 m from the smoker ( $59 \mu\text{g}/\text{m}^3$ ) was 31% of the smoker's exposure and 35 times the background concentration. The mean exposure of the person sitting 1.0 m from the smoker ( $40 \mu\text{g}/\text{m}^3$ ) was 21% of the smoker's exposure and 24 times the background concentration. Finally, the mean exposure of the nonsmoker sitting farthest from the smoker (1.5 m) was  $28 \mu\text{g}/\text{m}^3$ , or 15% of the smoker's exposure and 16 times the background concentration. Cigarette smoking times ranged from 3:29 to 6:03 min, with a mean of 4:38 min. As a result, each person's exposure during the smoking periods consisted of between 209 and 363 1-sec  $PM_{2.5}$  data points.

Figure 2 shows an example of the  $PM_{2.5}$  time series of the personal exposures of the four persons sitting on the bench, including the smoker, at Bus Stop E on El Camino Real next to Cambridge Avenue in Menlo Park. In this figure, note that the horizontal axes of the four graphs all have the same time scales, but the vertical exposure scale of the smoker is 0–6000  $\mu\text{g}/\text{m}^3$  (top graph), whereas the vertical scales of the three nonsmokers close to the smoker are reduced to 0–1000  $\mu\text{g}/\text{m}^3$  (the three bottom graphs). The vertical dotted lines indicate the two cigarette smoking periods (from 11:57:00 a.m. to 12:00:45 p.m. and from 12:06:56 to 12:10:25 p.m., with smoking durations of 3:45 and 3:29 min).

Numerous sharp  $PM_{2.5}$  concentration peaks, or *microplumes*, can be seen during each of the two smoking periods. Microplumes are brief packets of intensely high concentrations that occur close to a source during the emission period. They

**Table 2.** PM<sub>2.5</sub> personal exposure of one smoker and three nonsmokers sitting on bus stop benches during smoking period ( $\mu\text{g}/\text{m}^3$ )

Cigarette No.	Bus Stop	Cigarette Time (min)	Background <sup>a</sup>	Smoker's Exposure <sup>b</sup>	Exposure of Nonsmokers		
					0.5 m	1.0 m	1.5 m
1	A <sup>c</sup>	4:33	1.9	422	102 <sup>d</sup>	1.9	—
2	A	5:05	1.2	417	153	173	83
3	A	4:45	1.2	118	24	12	11
4	A	6:03	1.9	89	17	17	16
5	B	4:30	1.9	138	51	29	13
6	B	4:57	1.6	42	15	5	3
7	C	3:55	2.5	76	46	24	28
8	C	4:15	1.3	211	59	28	32
9	D <sup>e</sup>	5:16	1.3	26	70	44	24
10	D <sup>e</sup>	5:00	1.5	88	138	49	33
11	E	3:45	1.7	192	37	39	21
12	E	3:29	1.7	424	96	78	48
13	F	4:40	2.7	283	59	16	—
Mean	—	4:38	1.7	192.0	59.0	40.0	28.0
SD	—	0:41	0.5	145.7	46.0	45.0	22.1

Notes: <sup>a</sup>Mean of background before cigarette 1, between cigarettes, and after cigarette 2. <sup>b</sup>Mean of 1-sec readings in the smoker's breathing zone during smoking.

<sup>c</sup>Nonstandard position: Smoker was second from end. <sup>d</sup>Measurements on either side of the smoker (1.8 and 202  $\mu\text{g}/\text{m}^3$ ) were averaged. <sup>e</sup>Positions reversed with respect to direction of traffic.

were first reported by Furtaw et al. (1996) using a sulfur hexafluoride tracer gas from a point source in a chamber and subsequently measured indoors for both gases and particulate matter by McBride et al. (1999) in experiments with controlled emissions from a point source. The frequency distributions of the microplumes have been studied in indoor proximity experiments in two residences by Acevedo-Bolton et al. (2012) using multiple carbon monoxide (CO) sensors with a continuous CO tracer gas point source. Microplumes of PM<sub>2.5</sub> have been measured close to a smoker indoors and outdoors in a companion study (Acevedo-Bolton et al., 2013). Due to the microplumes, the 1-sec PM<sub>2.5</sub> exposures for the first cigarette at Bus Stop E ranged from 1.1 to 4840  $\mu\text{g}/\text{m}^3$  for the smoker and from 0.9 to 388  $\mu\text{g}/\text{m}^3$  for the person sitting next to the smoker.

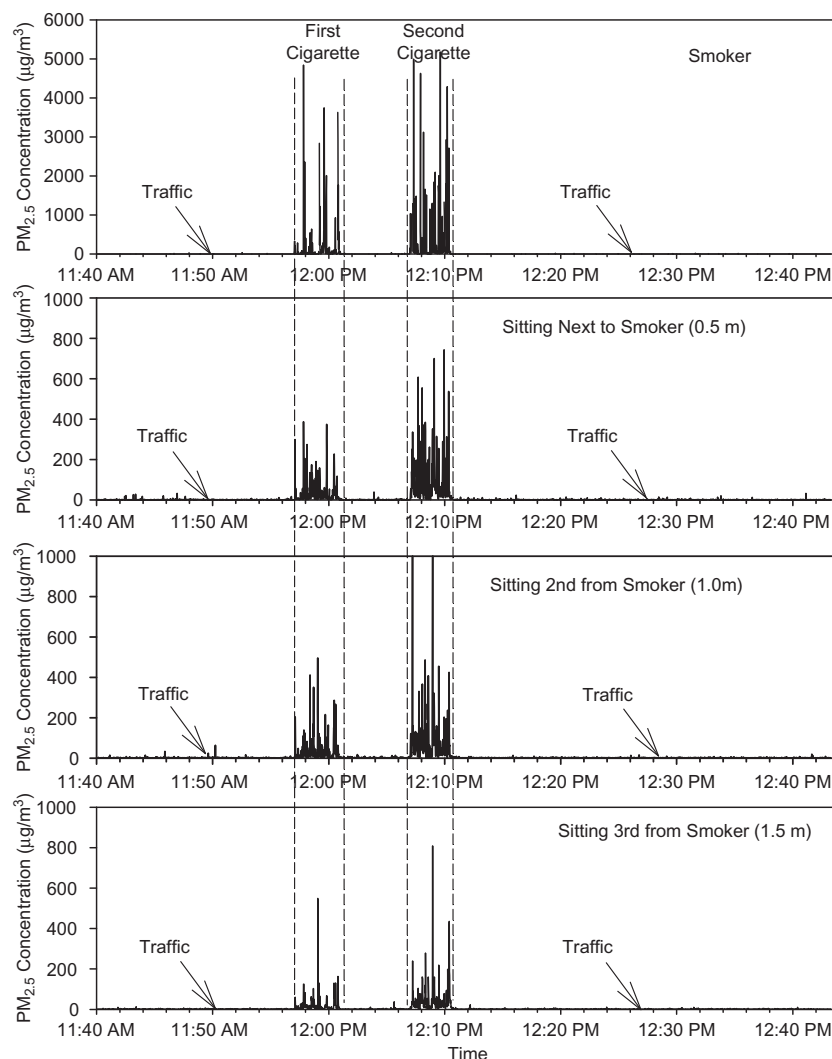
At all bus stops, the PM<sub>2.5</sub> concentrations during the three background time periods (before smoking, between the two cigarettes, and after smoking) were very low and did not vary appreciably. At Bus Stop E, for example, the arithmetic mean background concentration of the four monitors before smoking started was 1.5  $\mu\text{g}/\text{m}^3$  (SD = 1.7  $\mu\text{g}/\text{m}^3$ ;  $n = 1016$ ); the mean PM<sub>2.5</sub> during the background period between the two cigarettes was 2.2  $\mu\text{g}/\text{m}^3$  (SD = 9.3  $\mu\text{g}/\text{m}^3$ ;  $n = 364$ ); the mean for the background period after the second cigarette ended was 1.7  $\mu\text{g}/\text{m}^3$  (SD = 2.7  $\mu\text{g}/\text{m}^3$ ;  $n = 1972$ ). The weighted mean of these three background exposures was 1.7  $\mu\text{g}/\text{m}^3$  (Table 2). In contrast with these relatively low PM<sub>2.5</sub> background concentrations, the mean exposure of the closest nonsmoker (0.5 m) during the two smoking periods at Bus Stop E ranged from 37 to 96  $\mu\text{g}/\text{m}^3$ .

These results are based on the SidePak PM<sub>2.5</sub> monitor calibration factors for secondhand smoke described earlier, and although we do not have a custom calibration factor for the aerosol emitted by traffic sources, the calibration factor of 0.42

that Zhang and Zhu (2010) reported for school buses was not very different from the calibration factors we used. The experiments by Jiang et al. (2011) compared the SidePak monitors with gravimetric measurements of other combustion sources: large and small incense sticks (CF = 0.33–0.35), wood chips (CF = 0.77), and toasting bread (CF = 0.79). They also measured ambient outdoor aerosols in six experiments at two locations in Northern California, yielding a range of calibration factors from CF = 0.66 to CF = 0.93. Applying the higher factory default calibration factor CF = 1.0 to our SidePak data in the nonsmoking periods still would give relatively low background PM<sub>2.5</sub> concentrations for the sidewalks (<10  $\mu\text{g}/\text{m}^3$ ), which would include the contribution from nearby traffic.

We obtained the hourly average PM<sub>2.5</sub> concentrations measured at the closest ambient air monitoring station—the Bay Area Air Quality Management District air monitoring station at 897 Barron Avenue in Redwood City—for the same times as our experiments, and we found that these concurrent ambient readings all were low (<10  $\mu\text{g}/\text{m}^3$ ). This air monitoring station was located 1 km from Woodside Road, and its distance to the bus stops ranged from 1.6 km for Bus Stop D to 6 km for Bus Stop C.

At 3 bus stops, we included a fifth person who stood approximately 0.7 m from the smoker (Figure 1) wearing a PM<sub>2.5</sub> personal monitor with the same breathing zone necklace apparatus as the others. At Bus Stop E, the standing person's mean exposure during the first and second cigarettes was 4.7 and 2.3  $\mu\text{g}/\text{m}^3$ , respectively, or only slightly above the background concentration. In this experiment, the wind direction was observed to be away from the standing person and toward the smoker, so the fifth person was standing upwind of the smoker. The breathing zone of this person, who was 1.8 m (5 feet 10 inch) tall, was approximately 1.6 m above the sidewalk, whereas the sitting



**Figure 2.** Time series of breathing-zone  $\text{PM}_{2.5}$  exposures of one smoker and three nonsmokers measured during the 1-hr visit to Bus Stop E on El Camino Real and Cambridge Avenue.

smoker's breathing zone was approximately 1 m above the sidewalk, and the greater height may explain the lower exposure of the standing person compared with the exposure of the nonsmokers on the bench. An experimental indoor air quality proximity study with an array of up to 37 CO monitors reported that the highest concentrations occurred at the same height as the source (Acevedo-Bolton et al., 2012), and a similar phenomenon could apply to smoking outdoors.

At one site, Bus Stop D, we reversed the order of the participants with respect to the direction of traffic. Here, the smoker's breathing-zone exposure for both cigarettes (26 and  $88 \mu\text{g}/\text{m}^3$ ) turned out to be lower than at the other sites, so turbulence caused by the moving traffic may have reduced the smoker's exposure. Nevertheless, the mean exposures of the persons sitting next to the smoker— $104 \mu\text{g}/\text{m}^3$  at 0.5 m,  $47 \mu\text{g}/\text{m}^3$  at 1.0 m, and  $29 \mu\text{g}/\text{m}^3$  at 1.5 m—still showed a monotonically decreasing proximity effect for the two cigarettes. At this bus stop, the exposure to  $\text{PM}_{2.5}$  of the standing person was  $4.7 \mu\text{g}/\text{m}^3$  during the first cigarette and  $16.7 \mu\text{g}/\text{m}^3$  during the second cigarette,

compared with a mean background concentration for the two cigarettes of  $1.4 \mu\text{g}/\text{m}^3$ .

At Bus Stop F, where only one cigarette was smoked, there were two standing nonsmokers because of the short bench that could seat only three persons. The standing person near the smoker received an exposure of  $79.5 \mu\text{g}/\text{m}^3$  during the smoking period, and the farther standing person at the end of the bench (approximately 2 m from the smoker) received an exposure of  $9.1 \mu\text{g}/\text{m}^3$ , compared with a mean background concentration of  $2.7 \mu\text{g}/\text{m}^3$ .

### Frequency distributions of $\text{PM}_{2.5}$ microplumes

For each person's exposure, we sorted the  $\text{PM}_{2.5}$  concentrations from lowest to highest in one column of a worksheet using SigmaPlot, version 11 (Systat, San Jose, CA, USA). In another column, we computed the plotting position as  $100i/(n + 1)$ , where  $i$  is the order of the observation and  $n$  is the number of observations. We then created a logarithmic-probability graph by



choosing a logarithmic scale to the base 10 for the  $PM_{2.5}$  concentration on the vertical axis and a probability scale showing the plotting position on the horizontal axis. Because of its probability scale, the horizontal axis is the integral of the normal (Gaussian) probability density function multiplied by 100, which can be expressed as cumulative percentages, or *percentiles*. The straightness of the data points when plotted a graph of this type indicates the tendency of the frequency distribution to be approximately lognormal (Ott, 1995).

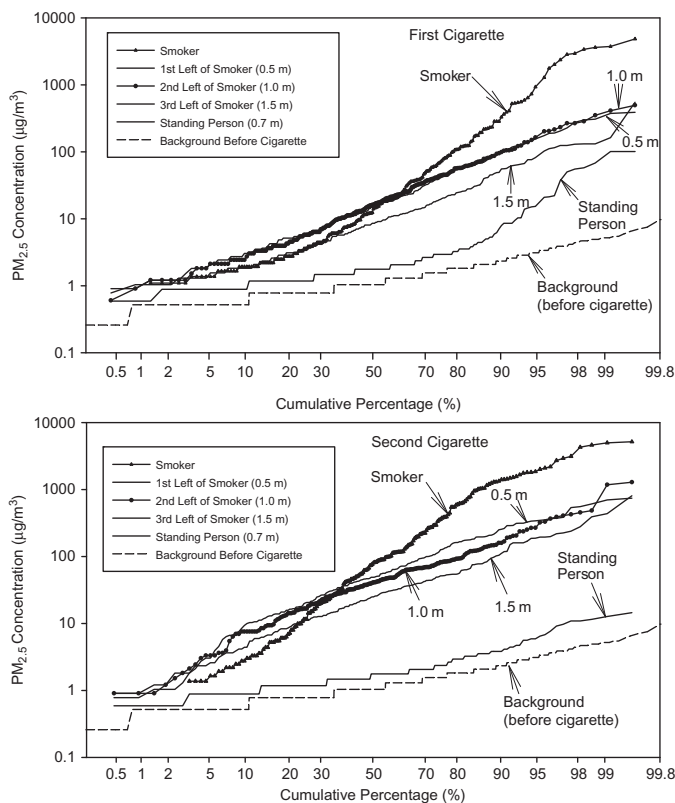
For both cigarettes, the resulting graph lines for Bus Stop E show that the smoker's breathing-zone exposure frequency distribution was higher than the nonsmokers' frequency breathing-zone distributions and had a greater slope (Figure 3). This is the same bus stop in which the  $PM_{2.5}$  exposure time series for the sitting persons were plotted in Figure 2. The two frequency distributions of the nonsmokers sitting 0.5 and 1.0 m from the smoker were relatively close together during both cigarettes, and both frequency distributions were higher than the frequency distribution of the nonsmoker 1.5 m from the smoker. The logarithmic-probability graphs for the three nonsmokers sitting on the bench were reasonably straight lines, although the lower frequency distributions of the standing person's exposure and the background concentration exhibited a gradual concave-upward curvature. The frequency distribution of the standing person's exposure was higher during the first cigarette than the second cigarette, and all the personal exposure frequency distributions were higher than those of the background concentrations.

All the bus stops showed microplumes during the smoking periods, and the resulting 95th-percentile values were relatively high (Table 3). For example, at Bus Stop B, located on Woodside Road near Atherwood Avenue, the smoker's cumulative frequency distribution for the first cigarette intersected the 95th percentile at  $868 \mu\text{g}/\text{m}^3$ , whereas the cumulative distribution of the nonsmoker sitting 0.5 m from the smoker intersected the 95th percentile at  $204 \mu\text{g}/\text{m}^3$ . Thus, 5% of the smoker's 1-sec readings were above  $868 \mu\text{g}/\text{m}^3$ , and 5% of the 1-sec exposures of the nonsmoker next to the smoker were above  $204 \mu\text{g}/\text{m}^3$ . The 95th-percentile background concentrations were computed as the mean of the individual 95th percentiles of the four to five monitors, which were low both before and after smoking ( $\leq 4.8 \mu\text{g}/\text{m}^3$ ).

During each cigarette, there was a 1-sec  $PM_{2.5}$  maximum exposure, and thus each person on the bench experienced 13 maxima, one for each cigarette. The smoker's 1-sec maximum exposures ranged between 1020 and  $5180 \mu\text{g}/\text{m}^3$  for the 13 cigarettes, and the range for the nonsmoker sitting next to the smoker was between 346 and  $5110 \mu\text{g}/\text{m}^3$ . The 1-sec maximum exposures of the nonsmoker 1.0 m from the exposure ranged from 12 to  $1850 \mu\text{g}/\text{m}^3$ , and the maximum exposures of the nonsmoker at 1.5 m ranged from 11 to  $850 \mu\text{g}/\text{m}^3$ . By comparison, the standing nonsmokers were exposed to six cigarettes, and their six maximum 1-sec exposures ranged from 14 to  $2773 \mu\text{g}/\text{m}^3$ . The high concentrations and wide ranges can be attributed to the multiple microplumes reaching each person.

Acevedo-Bolton et al. (2012, 2013) reported a proximity study in which adjusting the exposure by subtracting a small quantity from each measurement improved the linearity of the distributional fit. The quantity they subtracted—obtained by using a trial-and-error approach maximizing  $R^2$ —was very small. Following a similar approach, we found a best fit by subtracting  $1.3 \mu\text{g}/\text{m}^3$  from all the  $PM_{2.5}$  exposure measurements at Bus Stop B before plotting them (Figure 4). We found that the resulting frequency distributions were more linear after the adjustment than before the adjustment, especially for the lower graphs. This small adjustment moved the frequency distributions of the standing person and the background concentrations further downward at the smaller percentiles than at the larger percentiles, causing the lines on the graph to straighten. A physical interpretation of this adjustment may be that it helps separate the two concentration components that make up the background concentration measured on the sidewalk: the outdoor ambient concentration from more distant sources and the concentration caused by pollutant emissions from traffic on the adjacent roadway, which combine to make up the background concentration on the sidewalk.

The result of introducing this small incremental concentration that is subtracted causes a third parameter to be added to the two-parameter lognormal model, making it a three-parameter lognormal model, and the incremental parameter is called the *shift parameter* (Acevedo-Bolton et al., 2012). The adjusted concentrations fit to the lognormal model with geometric mean  $\mu_g$  and geometric standard deviation  $\sigma_g$  all had  $R^2$  values of 96.7% or higher (Table 4). Here,  $\sigma_g$  showed a striking decrease with increased distance from the smoker. These results reflect the high intensity of the many microplumes reaching the

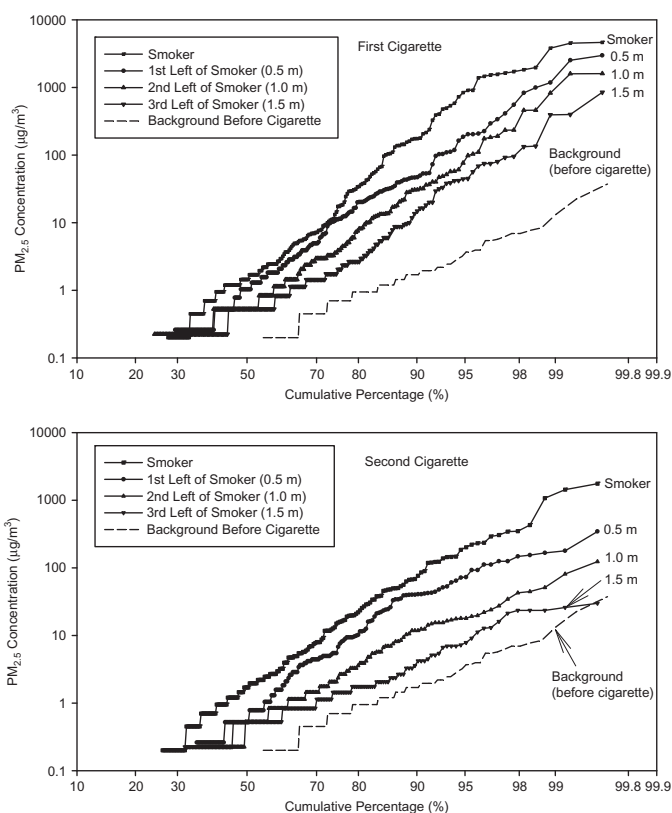


**Figure 3.** Frequency distributions of  $PM_{2.5}$  breathing-zone exposures of four persons sitting on the bench at Bus Stop E along with a standing person showing the first cigarette (top) and second cigarette (bottom).

**Table 3.** Ninety-fifth percentile  $PM_{2.5}$  exposures in  $\mu g/m^3$  for 13 cigarettes smoked at six bus stops

Cigarette No.	Bus Stop	Smoker	Nonsmokers Sitting on Bench			Nonsmoker Standing	Background (95th percentile)
			0.5 m	1.0 m	1.5 m		
1	A	2032	1064	3.5	—	—	4.0 <sup>a</sup>
2	A	2737	697	953.	428	—	3.2 <sup>b</sup>
3	A	510	112	68	47	—	2.0 <sup>a</sup>
4	A	470	83	74	12	—	2.8 <sup>b</sup>
5	B	868	204	87	46	—	4.8 <sup>a</sup>
6	B	198	74	19	9	—	0.9 <sup>b</sup>
7	C	340	98	181	84	—	3.8 <sup>a</sup>
8	C	1038	117	244	101	—	4.0 <sup>b</sup>
9	D	53	252	156	115	16.2	2.8 <sup>a</sup>
10	D	445	640	194	112	56.4	3.4 <sup>b</sup>
11	E	1032	157	170	157	16.1	3.4 <sup>a</sup>
12	E	1905	343	283	192	3.8	3.5 <sup>b</sup>
13	F	1885	302	55	—	79.5 <sup>c</sup> , 23.7 <sup>d</sup>	4.0 <sup>a</sup> , 4.9 <sup>b</sup>

Notes: <sup>a</sup>Background before smoking. <sup>b</sup>Background after smoking. <sup>c</sup>Standing person in normal position. <sup>d</sup>Standing person at end of bench from smoker.



**Figure 4.** Frequency distributions of  $PM_{2.5}$  breathing-zone exposures of four persons on the bench at Bus Stop B for the first cigarette (top) and second cigarette (bottom) showing the relative straightness of distributions after adjustment by subtracting a small shift parameter from all exposures ( $1.3 \mu g/m^3$ ).

nonsmokers during the cigarette's source emission period, and the decrease in  $\sigma_g$  indicates that their variability tends to decrease with distance from the smoker. The frequency

distributions at Bus Stops B and E (Figures 3 and 4), which show 4 of the 13 cigarettes smoked, were similar to those at the other four bus stops.

Statistical hypothesis tests for the normality of a distribution assume that the observations are independent random variables. The 1-sec  $PM_{2.5}$  time-series data at the bus stops exhibit serial correlation, or autocorrelation, and successive observations are not independent. As an alternative to probability tests, the coefficient of determination  $R^2$  is a useful indicator of the degree to which a frequency distribution is approximately lognormal, because it provides a quantitative measure of the linearity of the logarithmic-probability graph.

### Ultrafine particle (UFP) concentrations

The overall UFP concentrations measured during smoking periods also differed from cigarette to cigarette (Table 5), and the means during the smoking periods ranged from 8300 to 120,000 particles/ $cm^3$ , which were generally higher than the UFP background means during the nonsmoking periods. The overall mean of the three background UFP concentrations for all cigarettes was 7200 particles/ $cm^3$ . For the first cigarette, the mean UFP concentration for seven smoking periods at the bus stops was 33,900 particles/ $cm^3$ , or about 5 times the mean of the background concentrations. For the second cigarette, the mean UFP concentration for six cigarettes was 55,100 particles/ $cm^3$ , or 8 times the mean of the background concentrations. The overall UFP mean for the 13 smoking periods was 44,500 particles/ $cm^3$ , or 6.2 times the mean background concentration.

At Bus Stop E, which had the highest observed traffic volume (5100 vehicles/hr) of all the bus stops, the UFP concentrations showed a great increase when each of the two cigarettes was smoked (Figure 5). This was the same bus stop for which the  $PM_{2.5}$  time series of four persons were shown in Figure 2. The background  $PM_{2.5}$  exposures were very low during the

**Table 4.** Adjusted PM<sub>2.5</sub> exposures for two smoking periods at Bus Stop B, with arithmetic and geometric parameters of lognormal model estimated by linear regression

		Arithmetic Statistics <sup>a,b</sup>		Geometric Parameters <sup>b</sup>		
Monitor ID	Location	Mean (μg/m <sup>3</sup> )	SD (μg/m <sup>3</sup> )	μ <sub>g</sub> (μg/m <sup>3</sup> )	σ <sub>g</sub>	R <sup>2</sup> (%)
Cigarette 1						
SP-4	Smoker	136	539	1.52	36.8	98.9
SP-16	Sitting first left of smoker	50	264	1.00	24.0	99.3
SP-8	Sitting second left of smoker	28	153	0.70	17.0	98.2
SP-6	Sitting third left of smoker	12	64	2.17	13.5	97.6
SP-4	Background before cigarette 1	1.3	1.7	0.15	7.0	97.2
Cigarette 2						
SP-4	Smoker	41	169	0.64	19.8	99.0
SP-16	Sitting first left of smoker	14	36	1.28	17.7	96.8
SP-8	Sitting second left of smoker	4	0.7	0.43	10.8	98.0
SP-6	Sitting third left of smoker	1.6	4.3	0.41	6.0	97.7
SP-4	Background between cigs	1.8	1.6	0.12	8.1	96.7
SP-4	Background after cigarette 2	2.4	5.5	0.12	6.9	97.0

Notes: <sup>a</sup>Statistics of 1-sec measurements; increment of 1.3 μg/m<sup>3</sup> subtracted. SD = arithmetic standard deviation of model. <sup>b</sup>Parameters of lognormal model fit by linear regression.

**Table 5.** Ultrafine particle (UFP) concentrations measured by the person sitting next to the smoker for 13 cigarettes during seven bus stop smoking experiments<sup>a</sup>

Bus Stop	Before First Cigarette	During First Cigarette	Between Both Cigarettes	During Second Cigarette	After All Cigarettes
A1	9600	8300	11,200	35,000	10,500
A2	2300	56,000	5400	96,600	10,500
B	2500	13,600	6500	24,000	5400
C	7500	21,400	4200	20,200	2900
D	5100	31,100	2200	55,500	2200
E	9000	120,000	15,200	99,300	9900
F	11,500	14,900	9800	—	—
Mean	6800	33,900	7800	55,100	6900
SD	3600	41,700	4500	35,400	3900

Note: <sup>a</sup>Raw data: Background levels not subtracted.

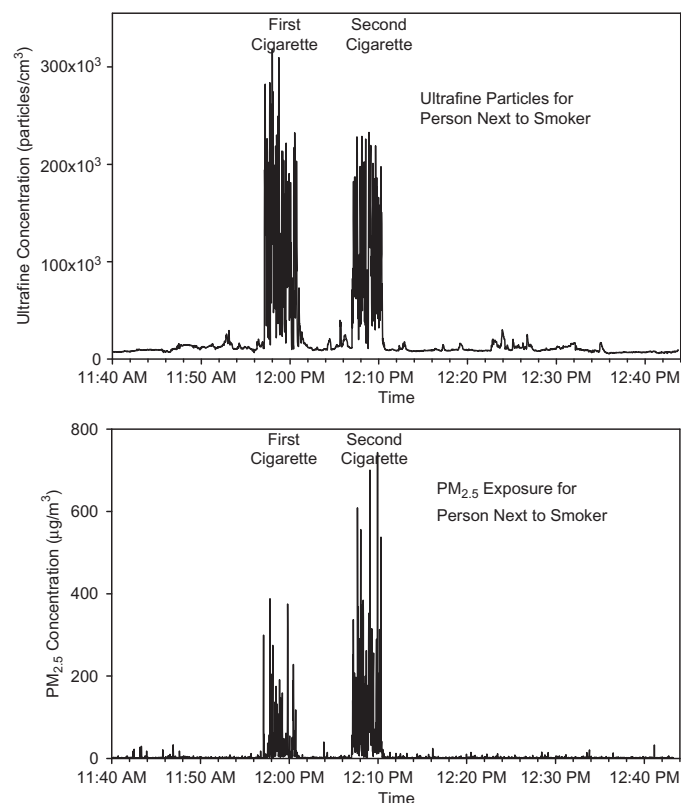
nonsmoking periods (1.7 μg/m<sup>3</sup>), but the exposures of the non-smoker sitting 0.5 m from the smoker increased to 37 and 96 μg/m<sup>3</sup> during the first and second cigarettes, respectively (Table 2), which was not exceptional compared with the other bus stops. The mean UFP background concentration, on the other hand, was 11,400 particles/cm<sup>3</sup> for the three background periods at this bus stop, and the means increased to 120,000 and 99,300 particles/cm<sup>3</sup> during the first and second cigarettes, respectively (Table 5), with an overall mean of 110,000 particles/cm<sup>3</sup> for the two cigarettes. It was not clear why the increase in the UFP concentrations during smoking at this bus stop was greater than at the other bus stops, although it may be due to variations in temperature, wind, and other factors. The UFP monitor was held on the lap of the person sitting next to the smoker (height ~0.5 m), whereas PM<sub>2.5</sub> was measured by personal monitors with intakes in each person's breathing zone (height ~1 m), so differences in the

movement of the smoke plumes at different heights above the sidewalk next to this busy highway could explain differences in the PM<sub>2.5</sub> exposures relative to the UFP concentrations.

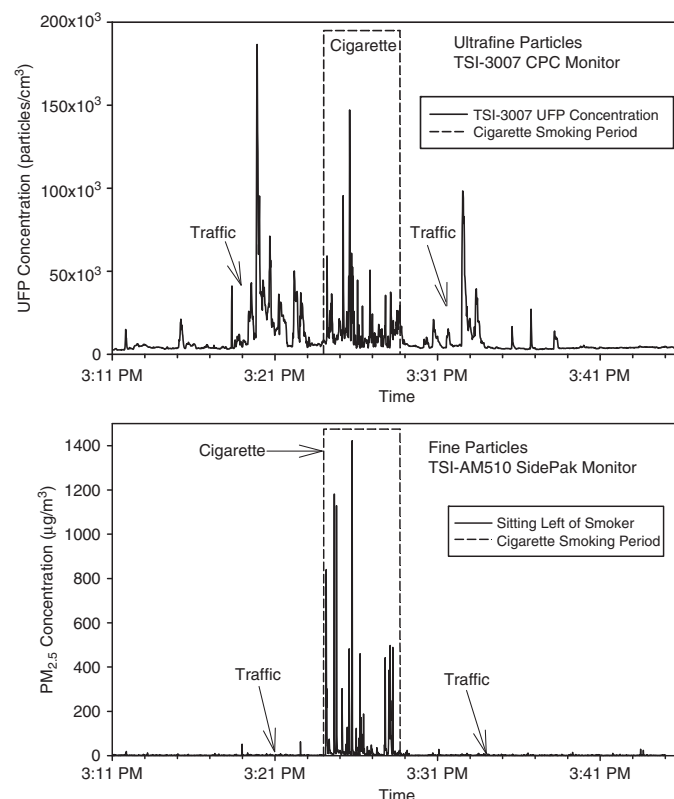
Unlike the low PM<sub>2.5</sub> concentrations measured during the nonsmoking periods, elevated peaks in the UFP background levels sometimes occurred during the nonsmoking periods at some bus stops, as illustrated by the UFP concentration time-series plot for Bus Stop F (Figure 6). By looking at oncoming traffic visually and viewing the monitor's real-time display, we observed that these elevated background UFP peaks appeared to be associated with large older trucks passing the bus stop.

### Uncertainties and implications

Based on results from a quality assurance study of 17 SidePak monitors (Jiang et al., 2011), the TSI SidePak monitor,



**Figure 5.** Time series of ultrafine particle concentration (top) and  $PM_{2.5}$  exposure (bottom) for the person sitting closest (0.5 m) to the smoker during the 1-hr visit to Bus Stop E.



**Figure 6.** Time series of ultrafine particle concentration (top) and  $PM_{2.5}$  exposure (bottom) for the person sitting closest (0.5 m) to the smoker during the half-hour visit to Bus Stop F.

when compared with gravimetric sampling, had a 95% confidence interval of  $\pm 4\%$ . Using individual gravimetric-based calibration factors for secondhand smoke for each monitor, as in the present study, would reduce the measurement error further. As discussed in Methods, the calibration factor for traffic emissions is less well established than for tobacco smoke, and the calibration factor for the TSI DustTrak reported by Zhang and Zhu (2010) for  $PM_{2.5}$  in school buses (0.42) was closer to the mean of the calibration factors for our seven SidePak monitors (0.28) than to the SidePak factory default value of 1.0. Even using the factory default calibration factor, however, the mean sidewalk background  $PM_{2.5}$  concentrations in our study all were  $<10 \mu\text{g}/\text{m}^3$ . The hourly average  $PM_{2.5}$  concentrations measured concurrently at the local air monitoring station also were  $<10 \mu\text{g}/\text{m}^3$ .

We visited six bus stops and collected more than 5000 1-sec readings at each bus stop using five to six simultaneous real-time monitors. This approach of using controlled experiments in real-life settings has been applied to other locations by Acevedo-Bolton et al. (2013) and is an alternative to conducting a larger-scale field survey. In this approach, investigators have full control of many variables of interest (for example, smoking times, positions of the subjects, distances between nonsmokers and the smoker, use of sampling probes in the breathing zone). This methodology is similar to the *scripted activity pattern* approach for measuring exposure in other studies (Stieb et al., 2008; Quiros et al., 2013).

The ratios of the standard deviation to the mean  $PM_{2.5}$  for the 13 smoking periods ranged from 0.78 to 1.14, indicating that the exposures at a given distance from the smoker varied considerably (Table 2). A larger sample size might increase the precision of the means, but the large differences between the smoking and nonsmoking periods are likely to persist. Lee and Cho (2012) are conducting a field study of 100 bus stops in Seoul, Korea, using SidePak monitors to compare  $PM_{2.5}$  concentrations before and after adoption of an outdoor smoking ban.

The elevated  $PM_{2.5}$  exposures of the nonsmokers caused by a single cigarette in this study were striking and were much higher than the effect of traffic on a nearby arterial highway on exposures (e.g.,  $<10 \mu\text{g}/\text{m}^3$  at 1.5–3.3 m). The ratios of the mean  $PM_{2.5}$  exposures of the nonsmokers during smoking to the background levels were large for all 13 cigarettes—ranging from 9.4 to 128 at 0.5 m, from 3.1 to 144 at 1.0 m, and from 1.9 to 69 at 1.5 m (Table 2).

Our study noted that elevated UFP concentrations during the nonsmoking periods at some bus stops appeared related to the passing of large, older trucks. Similar UFP concentrations near roadways and in traffic have been reported in other studies (Hagler et al., 2009; Zhang and Zhu, 2010). To reduce emissions from aging diesel trucks and buses in the state, the California Air Resources Board has approved a Truck and Bus rule that began its phase-in on January 1, 2012. This law requires nearly 1 million diesel trucks and buses in California to meet emission



standards and to apply a filter for particulate matter (Natural Resources Defense Council [NRDC], 2013). It is not clear how effectively the California Truck and Bus rule will reduce UFP concentrations on or near roadways, but the methodology of the present study could be used to identify high-emission vehicles, a subject for future research.

Our finding of a large number of microplumes—extremely high concentrations of short duration (2–5 sec) from highly concentrated packets of pollution close to a source—may be relevant to human health effects. Prior health effects studies have exposed human subjects to SHS in smoking lounges (Pope et al., 2001) and in chambers with aged smoke (Frey et al., 2012), but, to our knowledge, health studies have not focused on the intense microplumes that occur within 2 m from an active source. Intense, short-term exposures to particles could have more serious cardiovascular effects than exposures that are more uniform with respect to time, even if the mean values over the exposure time are the same. Future health effects research could address this issue by exposing subjects in controlled laboratory settings to concentrations in close proximity to an actively emitting source, such as a cigarette. It would be helpful to verify in such health effects studies that the statistical parameters of the lognormally distributed microplumes are similar to the statistical parameters measured in the present investigation.

In the United States, ambient  $PM_{2.5}$  concentrations have been decreasing for two decades due to air pollution regulations. However, a single cigarette is a potent source of particulate matter, emitting about 14 mg of  $PM_{2.5}$ , and causing relatively high concentrations near the smoker. An active cigarette also causes relatively high UFP concentrations near the smoker. These data indicate that a single cigarette can cause greater exposure to fine and ultrafine particles to persons close to a smoker than if they sat or stood by themselves on the sidewalk close to a major roadway.

## Conclusion

This study has shown that the outdoor personal exposure to  $PM_{2.5}$  of three nonsmokers located 0.5–1.5 m from a smoker and 1.5–3.3 m from a major arterial highway during 13 active smoking periods ranged from 1.9 to 144 times the background levels. The exposures of a person close to the smoker outdoors increased abruptly when the cigarette started (<6 sec) and returned to the background levels almost immediately after the cigarette ended (<6 sec).

For the 13 cigarette experiments at six bus stops, the mean  $PM_{2.5}$  exposure measured during the smoking period in the breathing zone of the smoker ( $192 \mu\text{g}/\text{m}^3$ ) was 113 times the mean background concentration ( $1.7 \mu\text{g}/\text{m}^3$ ), using each SidePak's individual custom gravimetric calibration factor for secondhand smoke. The mean exposure to  $PM_{2.5}$  of nonsmokers sitting close to the smoker decreased with increased distance from the smoker, supporting the hypothesis of a proximity effect outdoors at distances ranging from 0.5 to 1.5 m and probably beyond. The mean  $PM_{2.5}$  exposure of the nonsmoker sitting 0.5 m from the smoker ( $59 \mu\text{g}/\text{m}^3$ ) was 35 times the mean background concentration, whereas the mean exposure to  $PM_{2.5}$  at

1.0 m distance ( $40 \mu\text{g}/\text{m}^3$ ) was 23 times the background concentration, and the mean exposure of the nonsmoker at 1.5 m ( $28 \mu\text{g}/\text{m}^3$ ) was 16 times the background concentration. These results are for a single cigarette, but multiple smokers close together could cause higher exposures that potentially might last for longer periods.

In five of the six cigarette experiments with standing nonsmokers in our study, the standing person had mean  $PM_{2.5}$  exposures during the smoking periods of  $28 \mu\text{g}/\text{m}^3$  or less, and in one experiment the standing person had an exposure of  $79.5 \mu\text{g}/\text{m}^3$ . The exposures of the standing persons were higher than the background concentrations but usually lower than the exposures of persons sitting on the bench. The generally lower exposures of the standing persons may be due to the greater height of their breathing zone, compared with persons sitting on the bench.

The SidePak's custom calibration factor for secondhand smoke was based on gravimetric measurements, but the calibration factor is less well established for highway emissions. If we apply the high factory default calibration factor of  $CF = 1.0$  to the SidePak monitoring data for the background concentrations measured near the highways, then the mean  $PM_{2.5}$  background concentrations at the bus stops 1.5–3.3 m from the roadways would be less than  $10 \mu\text{g}/\text{m}^3$  at all sites. Thus, these arterial highways, with 3300–5100 vehicles/hr, made a relatively small contribution to  $PM_{2.5}$  exposures measured on the sidewalk close to the road, compared with a single cigarette smoked at a distance of 0.5–1.5 m.

The UFP concentrations measured next to the smoker at each bus stop also greatly increased during the smoking of a single cigarette, compared with the background concentrations measured near the smoker before, in between, or at the end of the cigarettes. The mean UFP exposure for the two cigarettes was  $44,500 \text{ particles}/\text{cm}^3$  for the 13 cigarettes, or 6.2 times the mean background UFP concentration of  $7200 \text{ particles}/\text{cm}^3$ . Unlike the  $PM_{2.5}$  concentrations, which were relatively low during the nonsmoking periods (mean of  $1.7 \mu\text{g}/\text{m}^3$ ), UFP concentrations had occasional high peaks during nonsmoking periods at some bus stops, which appeared associated with older, heavy-duty trucks passing on the roadway.

Both the  $PM_{2.5}$  and UFP concentrations measured during the smoking periods exhibited brief but intensely high concentrations, or microplumes. These peaks were similar to the microplumes measured in controlled proximity experiments conducted close to an emitting point source (McBride et al., 1999; Klepeis et al., 2009; Acevedo-Bolton et al., 2012, 2013). Although there has been increasing interest in the acute health effects of short-term mean exposure to  $PM_{2.5}$  for several minutes up to 1.75 hr, the health effects of  $PM_{2.5}$  personal exposures of high-intensity microplumes for extremely short durations (seconds or minutes) seem important and should be a candidate for future research on health effects. We have suggested how such health effects studies might be carried out.

The present study indicates that the exposure to fine and ultrafine particles that a person receives close to a smoker outdoors at a sidewalk bus stop can greatly exceed the levels measured on the sidewalk caused by a major arterial highway with heavy traffic (3300–5100 vehicles/hr). The results indicate

that close proximity (1.5 m or less) to a person smoking at a bus stop on a sidewalk can cause substantial exposure to both fine and ultrafine particles.

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