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Estimation of personal ozone exposure using ambient concentrations and influencing factors

Yue Niu[a,1], Jing Cai[a,b,1], Yongjie Xia[a], Haofei Yu[c], Renjie Chen[a,b], Zhijing Lin[a], Cong Liu[a], Chen Chen[a], Weidong Wang[a], Li Peng[b], Xiaoling Xia[d], Qingyan Fu[d], Haidong Kan[a,e,*]

[a] *School of Public Health, Key Lab of Public Health Safety of the Ministry of Education and Key Lab of Health Technology Assessment of the Ministry of Health, Fudan University, Shanghai, China*
[b] *Shanghai Key Laboratory of Meteorology and Health, Shanghai Meteorological Service, Shanghai, China*
[c] *Department of Civil, Environmental, and Construction Engineering, University of Central Florida, Orlando, FL, USA*
[d] *Shanghai Environmental Monitoring Center, Shanghai 200235, China*
[e] *Key Laboratory of Reproduction Regulation of National Population and Family Planning Commission, Shanghai Institute of Planned Parenthood Research, Institute of Reproduction and Development, Fudan University, Shanghai, China*

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ABSTRACT

Evidence is limited regarding whether ambient monitoring can properly represent personal ozone exposure. We conducted a longitudinal panel study to measure personal exposure to ozone using real-time personal ozone monitors. Corresponding ambient ozone concentrations and possible influencing factors (meteorological conditions and activity patterns) were also collected. We used linear mixed-effect models to analyze personal-ambient ozone concentration associations and possible influencing factors. Ambient ozone concentrations were around two to three times higher than personal ozone (43.1 μg/m^3 on average) and their correlations were weak with small slopes (0.35) and marginal R square (R_M^2) values (0.24). Larger R_M^2 values were found under high temperature (> 29.5 °C), low humidity (< 62.1%), good ventilation conditions (> 4 h) and for individuals spent longer time outdoors (> 0.6 h). In final model, personal ozone exposure was positively associated with ambient concentrations and ventilation conditions, but inversely correlated with ambient temperature and humidity. The models explained > 50% of personal ozone concentration variabilities. Our results highlight that ambient ozone concentration alone is not a suitable surrogate for individual exposure assessment. Meteorological conditions (temperature and humidity) and activity patterns (windows opening and outdoor activities) that affecting personal ozone exposure should be taken into account.

1. Introduction

Ground-level ozone is a highly reactive gaseous pollutant mainly produced from photochemical reactions involving its precursors such as nitrogen oxides and volatile organic compounds (U.S.EPA, 2013). In the past decades, ozone air pollution has been increasingly becoming a concern in China. As the largest mega city in China, Shanghai has witnessed a substantial increase in ambient ozone concentration, from annual average value of 32.7 μg/m^3 in 2006 to 55.4 μg/m^3 in 2015 (Gao et al., 2017). In summer 2013, a daily maximum ozone concentration as high as 343.5 μg/m^3 was observed (Pu et al., 2017).

The potential associations between ozone exposure and adverse health outcomes have been investigated in much previous epidemiological studies (Carey et al., 2013; Jerrett et al., 2009; Krewski et al., 2009), though inconsistent or even conflicting findings are still not uncommon. In these studies, the assessment of exposure to ozone pollution are primarily based on fixed-site monitoring, which use measured ambient concentrations as a proxy for exposure assessment. The variation of ozone concentrations in different microenvironments and the effects of individual activity patterns were not accounted. Such approaches are likely to introduce additional uncertainties and could contribute to the inconsistent results found in previous studies.

Personal exposures to ambient air pollution are affected by several factors. As an example, factors influencing personal exposure to $PM_{2.5}$ (particulate matter with an aerodynamic diameter < 2.5 μm) include meteorological variables, the mode of transportation, ventilation status, time spent outdoors and cooking frequency, all of which have been sufficiently documented in previous studies (Kaur and Nieuwenhuijsen,

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* Corresponding author at: Department of Environmental Health, School of Public Health, Fudan University, P.O. Box 249, 130 Dong-An Road, Shanghai 200032, China.
E-mail address: kanh@fudan.edu.cn (H. Kan).

[1] These authors contributed equally to this work.

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2009; Lanki et al., 2007; Sarnat et al., 2005). However, the factors affecting personal ozone exposures have not been fully investigated, partially due to the unavailability of personal monitors that are important for quantifying actual exposure level for individuals. Small and light weight passive monitors were often used in previous studies (Karakatsani et al., 2017; Kerckhoffs et al., 2015). Depending on the environmental concentrations and experimental designs, passive monitors often require extended sampling time to acquire adequate absorption on passive filters for subsequent chemical analysis. Hence, they hardly provide real-time data reflecting abrupt changes in ozone concentrations.

In this study, we performed repeated exposure measurements using real-time personal ozone monitors (POMs) on college students in Shanghai, China. We aimed to: 1) examine whether fixed-site ambient monitoring is able to properly represent personal exposure levels to ambient ozone pollution; and 2) identify important factors that may influence personal ozone exposures. Detailed information on study design, subjects, and sampling methods are provided in the next section, followed by results and discussion.

2. Methods

2.1. Study design and subjects

Forty-three nonsmoking college students from the medical campus of Fudan University were recruited into this longitudinal panel study. All students were randomly divided into 6 groups, and their personal ozone exposures were repeatedly measured four times during the study period, from 29 May to 12 October of 2016. To expand the coverage on temporal variations of ozone concentrations, we scheduled measurements at different days for different group. Before personal monitoring begins, demographic information was collected through a questionnaire.

The Institutional Review Board of the School of Public Health, Fudan University approved this study protocol (NO. 2014-07-0523), and all participants signed informed consent during enrollment.

2.2. Personal ozone measurement

Personal exposure to ozone was monitored in real time with POMs (2B Technologies, USA). POM is a miniaturized ozone monitor with low weight (340 g) and small size (10.1 × 7.6 × 3.8 cm). It measures ozone concentration based on ultraviolet absorption at the wavelength of 254 nm. Compared with passive samplers, POM has a much quicker response to changes in ozone level. During personal monitoring, all devices were set to acquire 5-min average ozone data at a flow rate of 0.8 L/min.

Personal monitoring for each participant was conducted for three consecutive days during daytime only (from 8:00 a.m. to 6:00 p.m.), since ozone pollution is generally not a concern at nighttime. Our participants were instructed to carry a backpack which houses the POM and a Teflon tube drawing air from the breath zone. They were asked to carry POMs for the entire 10 hour sampling time period except for during certain activities such as bathing, exercising or sleeping, when they can take off the bag and place it near him/her.

Personal ozone data collection strictly followed the quality assurance and quality control (QA/QC) procedures. Before personal monitoring, we calibrated all POMs against a transfer standard (Thermo Electron 49i-PS, Thermo Scientific Co., USA), which was calibrated by Shanghai Environmental Monitoring Center (SEMC) against Standard Reference Photometer. After calibration, the mean of the percent deviation between POMs and reference analyzer ranged from −0.4% to 1.3%, and coefficients of determination between units were larger than 0.99. Furthermore, before each visit, we conducted collocation comparisons among all POM units. Good consistency between POMs was observed, as relative standard deviation (%RSD) were 6.7%, 10.4%, 9.0% and 14.7%, respectively, for 4 collocation comparisons. Data processing also complied with QA procedures. We calculated 8-h (from 10:00 a.m. to 6:00 p.m.) average as daily personal concentrations only when the measurements covered > 75% of this time period. Eventually, 97.5% (459/471) of data were valid. For the measurements below the limit of detection (LOD), 9.6 μg/m^3 (4.5 ppb), we replaced them with LOD. In this study, approximately 3.0% (14/471) of measurements were below LOD.

2.3. Ambient ozone monitoring

Ambient ozone data from 4 state-controlled monitoring stations were retrieved from the database of the SEMC. The geographic locations of the 4 monitoring stations, and the medical campus (where participants were recruited), are shown in Supplement Fig. S1. Distances between monitoring stations and the medical campus range from 3.3 km (Station A) to 23.9 km (Station D). Fixed-site monitoring stations were located away from major emission sources (e.g. emissions of traffic, industry and residential dwellings). All monitoring instruments were placed on top of tall buildings at least 15 m off the ground. Therefore, the measurements represent general urban background levels of ambient ozone.

Ambient ozone levels were measured using a fixed-site ozone analyzer (Model 49i, Thermo Fisher Scientific Inc., MA, USA) based on the method of ultraviolet absorption. The instruments were operated and maintained properly followed the Automated Methods for Ambient Air Quality Monitoring (HJ/T 193-2005). Scheduled QA/QC procedures included automatic zero set, daily span and precision checks, and quarterly multiple-point calibrations. One-min average concentrations of ozone were recorded and 8-h (from 10:00 a.m. to 6:00 p.m.) average data were calculated for analyses.

Before personal monitoring, we performed a side-by-side sampling to examine the agreement between POMs and the fixed-site monitor. The result showed POMs agreed well with the fixed-site monitor (R square = 0.75–0.90, slope = 0.91–1.13, see Supplement Fig. S2).

2.4. Meteorological data

Meteorological data, including hourly mean temperature and relative humidity (RH), were obtained from the Shanghai Meteorological Bureau. All meteorological data were measured at a location 1.4 km from the medical campus (Fig. S1). Given that we used the 8-h values for ozone concentration, we calculated 8-h average for meteorological data in this study.

2.5. Time Activity Diary (TAD)

Information on daily activities of all participants related to ozone exposures were gathered through TADs for the 8-hour time period (from10:00 a.m. to 6:00 p.m.). The TAD coded locations into two categories: "indoors" and "outdoors". The "indoors" category refers to microenvironments such as dormitories and office buildings; and the "outdoors" category consists of two sub-categories: "in transportation", and "outdoors other than in transportation" for locations such as parks. Corresponding times spent in each coded microenvironment were recorded by each participant. To account for indoor-outdoor air exchange, all participants were also asked to record the length of time windows stay opened when they were in indoor environment. Based on the collected TAD data, we calculated the time each participant spent indoors and outdoors, and the time they spent indoors with windows open as a proxy for ventilation.