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Sources of personal PM_{2.5} exposure during pregnancy in the MADRES cohort

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BACKGROUND: Personal exposure to fine particulate matter (PM_{2.5}) is impacted by different sources each with different chemical composition. Determining these sources is important for reducing personal exposure and its health risks especially during pregnancy.

OBJECTIVE: Identify main sources and their contributions to the personal PM_{2.5} exposure in 213 women in the 3rd trimester of pregnancy in Los Angeles, CA.

METHODS: We measured 48-hr integrated personal $PM_{2.5}$ exposure and analyzed filters for $PM_{2.5}$ mass, elemental composition, and optical carbon fractions. We used the EPA Positive Matrix Factorization (PMF) model to resolve and quantify the major sources of personal $PM_{2.5}$ exposure. We then investigated bivariate relationships between sources, time-activity patterns, and environmental exposures in activity spaces and residential neighborhoods to further understand sources.

RESULTS: Mean personal PM_{2.5} mass concentration was 22.3 (SD = 16.6) μ g/m³. Twenty-five species and PM_{2.5} mass were used in PMF with a final R^2 of 0.48. We identified six sources (with major species in profiles and % contribution to PM_{2.5} mass) as follows: secondhand smoking (SHS) (brown carbon, environmental tobacco smoke; 65.3%), fuel oil (nickel, vanadium; 11.7%), crustal (aluminum, calcium, silicon; 11.5%), fresh sea salt (sodium, chlorine; 4.7%), aged sea salt (sodium, magnesium, sulfur; 4.3%), and traffic (black carbon, zinc; 2.6%). SHS was significantly greater in apartments compared to houses. Crustal source was correlated with more occupants in the household. Aged sea salt increased with temperature and outdoor ozone, while fresh sea salt was highest on days with westerly winds from the Pacific Ocean. Traffic was positively correlated with ambient NO₂ and traffic-related NO_x at residence. Overall, 76.8% of personal PM_{2.5} mass came from indoor or personal compared to outdoor sources. **IMPACT:** We conducted source apportionment of personal PM_{2.5} samples in pregnancy in Los Angeles, CA. Among identified sources, secondhand smoking contributed the most to the personal exposure. In addition, traffic, crustal, fuel oil, fresh and aged sea salt sources were also identified as main sources. Traffic sources contained markers of combustion and non-exhaust wear emissions. Crustal source was correlated with more occupants in the household. Aged sea salt source increased with temperature and outdoor ozone and fresh sea salt source was highest on days with westerly winds from the postpole.

Keywords: Personal exposure; PM2.5; Source apportionment; pregnancy; Secondhand smoking source; Traffic source

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INTRODUCTION

Personal exposure to particulate matter with aerodynamic diameter $<2.5 \,\mu$ m (PM_{2.5}) is impacted by indoor, outdoor, and personal activity-related (i.e., behaviors) sources in various microenvironments, spaces, and neighborhoods that individuals typically encounter [1, 2]. Prenatal exposure to PM_{2.5} specifically is associated with adverse maternal and fetal health outcomes [3–5]. Exposure in the 3rd trimester of pregnancy, when most fetal weight gain occurs is thought to be associated with low birth weight and impaired growth [6–8]. The toxicity of PM_{2.5} and its

subsequent impact on health is driven by its chemical composition and main sources contributing to it [8–10]. Identifying and quantifying the main sources of personal $PM_{2.5}$ exposure can shed light on particular mixtures that might pose a greater health risk and might otherwise be missed by solely investigating total $PM_{2.5}$ mass concentration as a whole. This is particularly important in environmental health disparities contexts and for specific vulnerable populations such as pregnant women for whom meaningful recommendations to reduce exposures and health risks are needed [11, 12].

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Main sources of PM exposure are generally resolved using source- and receptor-oriented modeling approaches [13, 14]. Based on the mass balance principle [15], receptor-oriented approaches utilize speciated measurements at receptors (i.e., receiving) to identify and quantify major sources (or source groups) impacting that location or individual [16, 17]. One of the most commonly used receptor models is the Positive Matrix Factorization Model (PMF) which solves for sources and their chemical profiles without pre-assuming what they are [18–20].

Several studies have conducted source apportionment in outdoor and indoor environments and investigated their health impacts [3, 21, 22]. For example, vehicular emissions, wood smoke, natural gas combustion, ship emissions, secondary aerosols, fresh and aged sea salt, and soil/road dust sources were resolved in outdoor PM (various size ranges) in Los Angeles, CA communities [23, 24]. Hasheminassab et al. [25] found mobile sources were the major contributor to both indoor ($39 \pm 21\%$) and outdoor ($46 \pm 17\%$) PM_{2.5} mass in three retirement homes in Los Angeles, Habre et al. [26] found cooking, cleaning, candle/incense burning, and smoking contributed significantly to indoor PM_{2.5} concentrations in New York City residences.

However, personal exposure to PM_{2.5} and its sources can be more complex to discern for several reasons. First, while personal monitoring (sampling air in the personal breathing zone) is considered the gold standard approach in external individual-level exposure assessment [27, 28], the cost and burden of conducting these studies is still high especially during pregnancy [29, 30]. Second, individuals get exposed to PM2.5 in multiple microenvironments and locations, often in close proximity to indoor sources or while mobile, sometimes generating PM_{2.5} from their activities (e.g., burning candles), and are always impacted by outdoor air pollution that infiltrates indoors or into the personal breathing zone [27, 28, 31, 32]. As such, disentangling sources that contribute to personal exposure can be more challenging compared to outdoor or indoor studies. This challenge is reflected by the relatively small number of studies conducting source apportionment in personal samples, most of which are collected 12 to 48 hour integrated PM samples [2, 33-35].

Of these, Özkaynak et al. [35] found that personal PM_{10} exposure was much higher than outdoor and indoor PM_{10} concentrations in 178 nonsmoking residents in Riverside, CA, and that these explained 16% and 50% of the variation in personal exposures, respectively. Cooking and smoking were important sources of personal exposure. Minguillón et al. [34] found wide variation in personal $PM_{2.5}$ exposures of 54 pregnant women and reported on limitations of questionnaire data (e.g., recall error, accuracy of time and location of travel and activities) in helping to resolve sources.

To the best of our knowledge, no studies to date have conducted source apportionment on personal PM_{2.5} exposure samples in the 3rd trimester of pregnancy. We aimed to chemically characterize the composition of personal PM_{2.5} and resolve its main sources in the MADRES (Maternal And Developmental Risks from Environmental and Social Stressors) cohort in Los Angeles, CA. To accomplish this goal, we analyzed filter-based data from a personal monitoring sub-study in MADRES using the United States Environmental Protection Agency (EPA) PMF model [19]. We leveraged questionnaire data, geolocation monitoring (GPS), and geospatial modeling of environmental exposures (in residential neighborhoods and activity spaces) to confirm predicted source identities and understand how their mass contributions vary in relation to personal behaviors, indoor/ outdoor sources, and time-activity patterns.

METHODS

Study design

A total of 213 women in their 3rd trimester enrolled in the larger MADRES cohort were recruited into this personal monitoring sub-study between

October 2016 and March 2020. MADRES is an ongoing prospective pregnancy cohort focused on predominantly low-income, Hispanic women and their babies residing in Los Angeles, CA. MADRES aims to address critical gaps in understanding environmental health disparities and the impacts of air pollution and social stressors on maternal and child health. The details of eligibility, enrollment, and follow-up of participants are described elsewhere [36]. Briefly, eligible participants for this sub-study were in the 3rd trimester at the time of recruitment, \geq 18 years of age, and could speak either English or Spanish fluently. In the initial design, people living in a household with an active smoker were excluded to reduce the impact from smoking on personal PM_{2.5} exposures. However, in order to encourage all participants to contribute to sub studies, the non-smoking household criterion was not applied consistently throughout the study and was eliminated by the end of 2018. Informed consent was obtained for each participant. The University of Southern California's Institutional Review Board (IRB) approved the study protocol.

Data collection

The 48-hr integrated personal PM_{2.5} measurements were collected to characterize the composition of personal PM_{2.5} and identify its main sources in the MADRES cohort through source apportionment analysis. Several other data sources were used from MADRES questionnaires and measurements and from external data sources in a second follow-on bivariate analysis to confirm source identities and understand the personal drivers that affect the mass contribution of each source. These included the following: questionnaires (collected at trimester 1, 2, and 3, respectively), GPS-derived time-activity patterns and environmental exposures within activity spaces from the personal monitoring study, modeled residential environmental exposures, and outdoor EPA $\rm PM_{2.5}$ chemical speciation data from a central site.

Personal PM_{2.5} measurements. The personal PM_{2.5} sampling design and protocol in the 3rd trimester is described in detail in O'Sharkey et al. [37] and Xu et al. [38]. Briefly, personal, 48-h integrated PM_{2.5} measurements were collected using a Gilian Plus Datalogging Pump (Sensidyne, Inc.) operating on a 50% cycle at 1.8 lpm flow rate with the sampling inlet located in the breathing zone. The pump is connected to a PM_{2.5} Harvard Personal Environmental Monitor (PEM) size-selective impactor with a 37 mm Teflon filter (2 µm pore size; Pall, Inc.). Participants were asked to wear the sampling device for the entire data collection period with a few exceptions. These included when it is unsafe to do so (e.g., driving), showering, or sleeping, in which case they were instructed to place the device near them in an unobstructed location.

Filters were analyzed gravimetrically to determine PM_{2.5} mass using an MT5 microbalance (Mettler Toledo, Columbus, OH, USA) in a dedicated humidity- and temperature-controlled chamber at the USC Exposure Analytics Laboratory. Filters were then sent to RTI International (Research Triangle Park, NC) to determine elemental composition of the following 33 species using Energy Dispersive X-Ray Fluorescence (EDXRF): barium (Ba), calcium (Ca), chlorine (Cl), copper (Cu), iron (Fe), potassium (K), magnesium (Mg), manganese (Mn), sodium (Na), nickel (Ni), sulfur (S), silicon (Si), titanium (Ti), zinc (Zn), aluminum (Al), bromine (Br), cobalt (Co), phosphorus (P), lead (Pb), selenium (Se), strontium (Sr), vanadium (V), cesium (Cs), zirconium (Zr), chromium (Cr), rubidium (Rb), arsenic (As), indium (In), silver (Ag), antimony (Sb), tin (Sn), cerium (Ce), and cadmium (Cd). Filters were also analyzed for concentrations of black carbon (BC), brown carbon (BrC), and environmental tobacco smoke (ETS) using a seven-wavelength optical transmittance integrating sphere method [39, 40].

Questionnaires. Participants completed interviewer-administered questionnaires in trimester-specific visits as part of the larger MADRES cohort and an exit survey after completing the 48-hr monitoring period as part of the personal monitoring sub-study (Table S1 and S2). Data obtained from the MADRES questionnaires include the following: demographics (e.g., age, race, education, employment, income), housing characteristics (e.g., type of dwelling, building age). In addition, data on the following were available from the exit survey: time-activity patterns (e.g., time spent indoors and outdoors, commuting), home ventilation (e.g., window open, air conditioner use), current tobacco smoke exposure (primary and secondhand), and presence of any significant indoor sources of PM_{2.5} such as cooking or candle burning [36]. Participants' home addresses at the 3rd trimester study timepoint were geocoded for residential exposure assessment.

Residential environmental exposure assessment. Daily ambient concentrations of PM_{2.5}, PM₁₀, nitrogen dioxide (NO₂), and ozone (O₃) were interpolated at the residence using inverse distance squared weighted interpolation from US EPA Air Quality System data [36]. Daily local trafficrelated nitrogen oxides (NO_x) concentrations at the residence were estimated using the CALINE4 line source dispersion model by roadway class [41]. Daily meteorology (temperature, precipitation, specific humidity, relative humidity, downward shortwave radiance, wind direction and wind speed) was assigned based on a 4 km × 4 km gridded model developed by Abatzoglou [42]. Forty-eight-hour integrated averages were calculated from daily measurements to correspond to the personal monitoring dates. For wind direction, four categories were created based on the 48-hr mean (arithmetic mean of two vector averages) as follows: 0-90° as wind blowing from northeast (NE), 91-180° as southeast (SE), 181-270° as southwest (SW), and 271-360° as northwest (NW), where a direction of 0° is due North on a compass.

GPS-derived time-activity patterns and environmental exposures within activity spaces. Smartphones with the study-developed madresGPS Android app pre-installed and programmed were used to log participants' geolocation (GPS and metadata) and motion sensor data continuously at 10-s intervals for the 48-hr monitoring period. Data were then analyzed to derive time activity patterns as minutes spent staying at home or non-home locations (assumed to be indoors) and minutes spent on the road (or in transit, travel mode unknown) and then converted to percentages out of the 48-hr period for use in the analysis. The methods used to derive time-activity patterns were based on [43, 44] and described in more detail in Xu et al. [38].

GPS data was also used to construct 48-hr activity spaces and calculate environmental exposures encountered within them [38]. Briefly, activity spaces are defined as the local areas that individuals interact with when they move around during their daily activities [45, 46]. We constructed activity spaces using Kernel Density Estimation (KDE) method for each individual and then calculated the following environmental exposures within them: walkability index score, Normalized Difference Vegetation Index (NDVI, greenness), parks and open spaces, traffic volume on primary roads, and road lengths (for primary and secondary roads combined and for minor streets). Data sources for these measures are listed in Table S3. Briefly, KDE integrates time and space to account for durations of time spent at certain locations and incorporates a distance decay kernel function to assign higher weight to environmental features closer to the locations where participants spent the most time in (compared to locations they passed through) using pre-defined bin (e.g., 25 m) and neighborhood sizes (e.g., 250 m) [45, 47]. Activity space calculations were conducted in ArcGIS Pro 2.5 (Esri, Redlands, CA).

*Outdoor PM*_{2.5} chemical speciation in study region. PM_{2.5} metals and carbonaceous components concentrations were available every third day from the Chemical Speciation Network (CSN) [48] at the Downtown Los Angeles monitoring station—the most proximal and central site in the study area—and were downloaded from the EPA Air Quality System. Then, the data was linked to personal monitoring 48-hr time periods based on the overlapping dates.

Data analysis

Descriptive statistics were first conducted to plot the distributions of all the variables (e.g., individual/residential environment data) for the participants. Then, using USEPA PMF5.0 model, source apportionment analysis was performed on personal PM_{2.5} measurements to identify main sources, along with their contributions to PM_{2.5} mass. Once sources were resolved, bivariate analysis was conducted between each PM_{2.5} source and several variables describing personal behaviors, indoor/outdoor sources, and time-activity patterns to confirm source identities and understand the personal drivers that affect PM_{2.5} exposures.

Descriptive analysis. Descriptive statistics were calculated in SAS 9.4 (SAS Institute Inc) to describe the distribution of personal PM_{2.5} mass concentration, elemental components and carbon fractions (used in final PMF analysis), questionnaire variables (home ventilation, time-activity patterns, indoor sources, etc.), and environmental exposures (residential or GPS-derived).

Source apportionment analysis. The USEPA PMF 5.0 model was used to resolve and identify major sources of $PM_{2.5}$ and quantify their mass

contributions using measured concentrations and sample-specific uncertainties as inputs. Briefly, the PMF model uses factor analysis to identify source contributions and profiles for a given number of sources through solving the following equation: [19, 20, 49]

$$X_{ij} = \sum_{k=1}^{n} g_{ik} f_{kj} + e_{ij} \tag{1}$$

where X_{ij} represents the concentration of chemical component *j* in sample *i*, g_{ik} represents the mass contribution of factor *k* in sample *i*, f_{kj} represents the loading of chemical component *j* on factor *k*, and e_{ij} is the residual error for sample *i* and species *j*.

The PMF model solves Eq. (1) by minimizing the sum of squares object function Q for a given number of factors k: [20, 49]

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{e_{ij}}{|u_{ij}|} \right]^2$$
(2)

where u_{ij} is the uncertainty of species *j* in sample *i*. The model decomposes the concentrations matrix into a contributions *g* matrix and profiles *f* matrix and constrains results to be positive (or not significantly negative) [20, 50]. Each observation is individually weighted by its uncertainty in Eq. (2); therefore, samples with higher analytical uncertainties will have less influence on the solution.

Based on the PMF-calculated signal-to-noise ratio (S/N) indicating the degree of analytical noise relative to the concentration of each species [19], we categorized species as "Bad" (S/N \leq 0.2, excluded from analysis), "Weak" (0.2 \leq S/N \leq 1, downweighted in the analysis), and "Strong" (S/N \geq 1, retained). Although Pb and V had S/N < 0.2, they were included in the analysis as potentially important tracers of traffic and fuel oil, respectively, and set to "Weak."

Of the 36 elemental and carbon species, the following 16 were finally included as "Strong" in the PMF analysis: BC, BrC, Ba, Ca, Cl, Cu, Fe, K, Mg, Mn, Na, Ni, S, Si, Ti, and Zn. We also included 9 "Weak" species as follows: Al, Br, Co, ETS, P, Pb, Se, Sr, and V. PM_{2.5} mass was designated as the total variable. An extra 10% modeling uncertainty was added to account for sampling or modeling errors not captured in the sample-specific analytical uncertainties [19]. Missing concentrations were replaced by species' medians. One out of 213 (0.5%) samples were excluded as outliers based on multiple species' concentrations.

Solutions with five to seven factors were scanned first to decide upon a reasonable factor number in a final model with 100 runs. Q values were checked for no undue influence from outliers and no local minimum solutions. Based on chemical loading profiles and prior knowledge, the optimal number of sources was selected which provided the most physically interpretable solution [50]. The convergent solution with the lowest Q_{robust} value (goodness-of-fit parameter excluding points with uncertainty-scaled residuals greater than 4) was selected [19]. Fpeak rotations, where positive F peak values sharpen the F matrix and negative values sharpen the G matrix were performed next to refine the solution. The optimal Fpeak value for solution rotation was chosen based on the smallest change in Q [19]. Residuals and $R^{2'}$ s for each species were checked for normality and model fit, respectively. Finally, diagnostics analysis of Displacement (DISP), Bootstrap (BS) (100 bootstraps, 0.6 minimum correlation), and Bootstrap-Displacement (BS-DISP) were performed to estimate the variability in the PMF solution under different scenarios. DISP focuses on effects of rotational ambiguity in the profiles or loadings; BS identifies whether there are a small set of observations that can disproportionately influence the solution; and BS-DISP includes effects of random errors and rotational ambiguity [19].

Bivariate confirmatory analysis of source identities and drivers. Bivariate analyses were conducted to further confirm source identities and examine trends in their mass contributions. Scatterplots, boxplots, and nonparametric statistics (Spearman correlations for continuous variables and Kruskal-Wallis test for categorical ones) were used to describe and test the relationships of select factors (selected from the literature and prior knowledge) with each source's predicted mass contributions. These included demographics, time-activity patterns, home characteristics, indoor air pollution sources, and outdoor residential and activity space based environmental exposures as described earlier. Categorical variables with unbalanced values (≥85% of the records have one value) or with high missingness (≥80%) were dropped from further analysis.

4

Descriptive statistics

RESULTS

Most of the participants (>98%) resided in central and east Los Angeles, CA. The majority were Hispanic (78%), working (48%) during the 3rd trimester, and completed grade 12 or less (55%) (Table S1). Mean age was 28 years at consent (range 18–45 years) and mean parity was 2 (range 1–6). Among participants who reported annual household income, most of them had annual household income <\$30,000 (68%).

Personal PM_{2.5} mass and component concentrations are provided in Table 1. Mean (SD) PM_{2.5} concentration was 22.3 (16.6) μ g/m³. The optical carbon fractions BC, BrC, and ETS combined constituted on average 17% (3.8) μ g/m³ of the total PM_{2.5} mass.

The distributions of home characteristics, indoor PM sources, and selected time activities as reported in questionnaires or derived from GPS are presented in Table S2. Based on the exit survey, 60% of participants spent some time near traffic when outdoors, and 34% spent more than 2 hrs per day commuting during the 48-hr monitoring period. During sampling, 60% of participants opened windows more than half of the time, 26% used air conditioning and 37% used fans at home. In terms of indoor PM sources, 38% were close to cooking smoke, 24% were close to burning candles or incense, and 39% spent time close to people smoking (cigarette, cigar, hookah, or pipe smoke) nearby. In addition, 56% of participants lived in an apartment, 44% were part of a household with >3 persons, and 43% lived in a home

Table 1. $PM_{2.5}$ mass and chemical component concentrations (in units of ng/m³ unless otherwise noted).

	N	Mean	SD
PM _{2.5} mass (µg/m ³)	209	22.3	16.6
Carbon species			
BC (μg/m ³)	209	1.1	1.7
BrC (µg/m ³)	206	1.1	0.8
ETS (µg/m ³)	210	1.6	6.1
Elements			
Al	212	12.4	47.2
Ва	212	14.2	13.5
Br	212	3.0	3.1
Ca	212	85.6	141.2
Cl	212	126.5	253.2
Со	212	0.5	0.8
Cu	212	18.7	12.2
Fe	212	122.2	110.7
К	212	105.6	142.0
Mg	212	39.0	62.6
Mn	212	2.6	2.9
Na	212	306.0	301.4
Ni	212	2.3	2.7
Р	212	5.4	17.8
Pb	212	1.4	2.6
S	212	401.6	293.3
Se	212	1.6	1.8
Si	212	165.7	203.3
Sr	212	1.8	6.7
Ti	212	10.2	12.7
V	212	0.6	1.2
Zn	212	13.1	17.5

built after the 1980s. Participants also spent an estimated mean (SD) of their time at home 78.5% (19.6%) and at non-home locations 15.2% (15.7%).

Source apportionment results

Fourteen observations had missing values and were replaced by species median, including PM mass (3 obs), BC (3), BrC (6), and ETS (2). A five-factor solution combined fuel oil and secondhand smoking, while seven factors resulted in a non-interpretable factor with single high loading of Zn. A six-factor solution was chosen as the optimal, physically interpretable one ($Q_{robust} = 5845.4$ and $Q_{true} = 6143.2$). An Fpeak rotation of 0.1 was then applied with 100 bootstraps, which resulted in no unmapped factors (Table S4). Some species with higher S/N ratio, e.g., BC, Cl, K, S, Ca, and Zn, had non-normal residuals (Table S5).

The six sources together explained 48% of the variability in $PM_{2.5}$ mass concentration and included the following: Traffic, Secondhand Smoking, Aged Sea Salt, Fresh Sea Salt, Fuel Oil, Crustal (Fig. 1). Each of these is presented in more detail below along with supporting relationships from confirmatory bivariate analyses.

Traffic. The first source identified was traffic with high loadings of BC, Zn, and Ba (Fig. 1). It contributed on average 2.6% of personal $PM_{2.5}$ mass (Table 2). Traffic was moderately positively correlated with crustal and inversely correlated with fresh sea salt and fuel oil sources (Table 3). It was strongly correlated with outdoor residential ambient (NO₂ and PM_{2.5}) and traffic-related (total CALINE4 NO_x) air pollutants. It was also negatively correlated with greater length of primary roads in the KDE activity space (Table 4).

Secondhand Smoking. This second source had a high loading of BrC and ETS (Fig. 1). With an average mass contribution of 11.9 μ g/m³, it contributed the majority of personal PM_{2.5} mass (65.3% on average) (Table 2). Participants living in apartments seemed to have slightly higher exposure to this source compared to those living in houses (13.0 vs. 10.5 μ g/m³, respectively, not significant, Fig. 2a). This source was also slightly negatively correlated with greater window opening time. Secondhand smoking (SHS) was also slightly higher when participants were close to 2+ persons smoking nearby as compared to one (among ~33% of participants reporting being close to someone smoking in the 48-hr monitoring period) (13.2 vs. 11.0 μ g/m³ respectively, not significant). To eliminate the possibility that this source is capturing an outdoor biomass burning signal, we checked its correlation with outdoor potassium (elemental, K), potassium ion (K⁺), and elemental and organic carbon concentrations at the Downtown



Fig. 1 PMF-predicted source loading profiles (in % of species).

Sources are color coded as shown: Aged Sea Salt Freeh Sea Salt Free Oil CSN site, all of which showed insignificant weak correlations (Table 4). SHS was also not correlated with any questionnaire variables on indoor cooking (frequency, use, etc.).

Aged sea salt. This third source had high loadings of Na, Mg, and S (Fig. 1). It contributed on average 4.3% of the personal $PM_{2.5}$ mass (Table 2). Aged sea salt was negatively correlated with the SHS source (Table 3). It was strongly positively correlated with outdoor O_3 concentration and temperature and negatively correlated with wind speed (Table 4). Aged sea salt was also significantly positively correlated with window opening time (Fig. 2b).

Fresh sea salt. This fourth source had high loadings of Cl. Na. and Mg (Fig. 1). It contributed on average 4.7% of the personal PM_{25} mass (Table 2). Fresh sea salt was negatively correlated with traffic and SHS sources (Table 3). The mass contribution of this source were significantly different across different wind directions, with the highest contribution on days when average wind direction originated from the west (NW followed by SW, Fig. 2c). It was also positively correlated with outdoor residential wind speed and relative humidity. To eliminate the possibility of this being an indoor source related to aerosolized minerals from domestic water use or salt used in cooking [32, 35, 51], we checked its relationships with humidifier usage and time close to smoke from cooking, respectively. Even though the sample size was unbalanced (30 out of 212 reported using a humidifier), average mass contributions were lower (not significant) when people used a humidifier compared to not (0.5 vs. 0.9 µg/m³, respectively). Similarly, mass contributions were lower when participants reported spending more time close to smoke from cooking. In addition, fresh sea salt was moderately positively correlated with ambient Cl and Cl⁻ as measured at the Downtown Los Angeles site (Table 4).

Table 2.	Predicted source contributions to personal $PM_{2.5}$ mass
concentr	ations.

Sources	Average (SD) mass contribution (µg/m ³)	% contribution to personal PM _{2.5} mass (%)
Traffic	0.5 (0.6)	2.6
Secondhand smoking	11.9 (9.2)	65.3
Aged sea salt	0.8 (0.8)	4.3
Fresh sea salt	0.9 (2.1)	4.7
Fuel oil	2.1 (1.6)	11.7
Crustal	2.1 (3.6)	11.5

Fuel oil. This source had high loadings of Cu, Ni, and V (Fig. 1). It contributed the second largest share of personal $PM_{2.5}$ mass (11.7%) with an average mass contribution of 2.1 µg/m³ (Table 2). Fuel oil was positively correlated with crustal and negatively correlated with traffic sources (Table 3). This source was slightly higher (non-significant) in participants living in homes built before 1980 compared to after (2.4 vs. 1.9 µg/m³). Fuel oil was also positively correlated with outdoor NO₂ and negatively correlated with O₃ (Table 4).

Crustal. The sixth and final source had high loadings of crustal elements Ca, Si, Ti, and Al (Fig. 1). It contributed 11.5% (2.1 μ g/m³) of personal PM_{2.5} mass on average (Table 2). Crustal was moderately positively correlated with traffic and fuel oil sources (Table 3). Living in a household with 4+ occupants was associated with greater contributions of this source compared to fewer occupants (2.5 vs. 1.2 μ g/m³, *p* value = 0.04, Fig. 2d). It was also positively correlated with relative humidity and precipitation (Table 4).

DISCUSSION

We identified six sources of personal PM_{2.5} exposure along with their mass contributions in the 3rd trimester of pregnancy, in a population of 212 low-income, predominantly Hispanic pregnant women living in Los Angeles, CA. Secondhand smoking (SHS) followed by fuel oil and crustal were the biggest contributors to personal PM_{2.5} exposures. Of the six sources, SHS and crustal appeared to be of indoor origin (or closely related to indoor activities), while traffic, aged and fresh sea salt, and fuel oil were of outdoor origin. Secondhand smoke was the single largest contributor to total personal PM_{2.5} mass concentrations. The combined indoor source contributions $(14 \pm 10 \,\mu\text{g/m}^3)$ were more than triple those of outdoor sources $(4.2 \pm 2.9 \,\mu\text{g/m}^3)$, highlighting the importance of the indoor environment in contributing to personal exposures during this critical window of time.

SHS is also a well-known indoor air contaminant [52, 53] associated with a suite of adverse health effects [54–56]. This source had high loadings of BrC and ETS (the lab-reported marker of SHS), and some loadings of Br and K which were related to tobacco smoke in previous studies [39, 57, 58]. In order to avoid overloading the samplers with particles from primary tobacco smoke which would also overshadow chemical fingerprints from other sources if present, by design, we excluded participants who reported smoking themselves (this did not occur in this population) or those with an active smoker permanently residing in their household (despite this latter criterion not being

Table 3. Spearman correlation matrix of PMF-predicted PM_{2.5} source contributions, colored from low (blue) to high (red).

	Traffic	Secondhand Smoking	Aged Sea Salt	Fresh Sea Salt	Fuel Oil	Crustal
Traffic						
Secondhand Smoking	-0.08					
Aged Sea Salt	-0.01	-0.27				
Fresh Sea Salt	-0.20	-0.24	0.07			
Fuel Oil	-0.20	-0.01	-0.07	-0.01		
Crustal	0.34	0.01	-0.09	-0.09	0.14	

Values in bold font represent significant p values at p < 0.05 level.

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Table 4. Spearman correlations between PMF-predicted $PM_{2.5}$ source contributions and select continuous variables.

Sources	Variables	Spearman correlation
Traffic	Outdoor air pollution at the residence	
	O ₃	-0.35
	NO ₂	0.61
	PM _{2.5}	0.43
	Total NO _x from local traffic on all road classes (CALINE4)	0.15
	Total length of primary roads within KDE area	0.15
Secondhand Smoking	Ambient air pollution at Downtown Los Angeles monitoring site	
	Potassium ion (K ⁺)	0.12
	Potassium (element, K)	-0.04
	Elemental carbon	0.04
	Organic carbon	0.09
Aged Sea Salt	Outdoor air pollution and meteorology at residence	
	O ₃	0.53
	Wind speed	- 0.22
	Temperature	0.55
Fresh Sea Salt	Outdoor meteorology at residence	
	Wind speed	0.27
	Relative humidity	0.16
	Ambient air pollution at Downtown Los Angeles monitoring site	
	Chloride ion (Cl ⁻)	0.24
	Chlorine (element, Cl)	0.20
Fuel Oil	Outdoor air pollution at residence	
	0 ₃	- 0.18
	NO ₂	0.17
Crustal	Outdoor air pollution and meteorology at residence	
	NO ₂	0.39
	PM ₁₀	0.24
	Relative humidity	-0.48
	Precipitation	-0.16

Values in bold font represent significant p values at p < 0.05 level.

consistently applied throughout the study). Despite the low smoking prevalence and these design restrictions, secondhand smoking was still identified as the source with the largest contribution to personal $PM_{2.5}$ exposures in our study. The mass contributions of this source (and measured ETS concentrations) did not show any clear changes over time as the study progressed, suggesting that recruitment decisions did not significantly influence our findings. This source has been found in other personal $PM_{2.5}$ monitoring studies [34, 35, 59]. Minguillón et al. [34] identified cigarette sources in the personal, indoor and outdoor environments for the pregnant women living in Barcelona, Spain. Özkaynak et al. [35] found nicotine mainly in the personal sample and indoor environment for the nonsmoking

residents in Riverside, California. Using a real-time PM_{2.5} monitor in personal and residential environment, Zhang et al. [59] found SHS distribution in children with and without self-reported SHS exposure in New York City, where children from smoking families had six times that of SHS exposure for children from non-smoking families. All these studies, including the current one, demonstrate that SHS source is a common finding in the personal monitoring studies. This suggests that secondhand (and possibly thirdhand) smoke exposure remains an issue of public health concern especially during pregnancy and in the context of our population, despite the success of large-scale bans on public smoking. We found that participants living in apartments tended to have marginally higher exposure to secondhand smoking than those living in detached houses. This could suggest greater potential of secondhand smoke infiltration from adjacent units in an apartment building or from visitors smoking [60-62]. Anecdotal evidence from the study (data not shown) also suggests secondhand smoke exposure could be occurring outside of the residence, during public transit or while commuting, and warrants further investigation.

We also found both fresh and aged sea salt—sources of outdoor origin—contributed to personal PM_{2.5} exposures in our study with high loading of Na, Mg, Cl (fresh only), and S (aged only). Previous work identified sea salt or marine aerosol sources with similar loading profiles [16, 23, 63]. Despite only having access to average wind direction data over the 48-hr monitoring period (not an ideal wind measure compared to most frequent wind direction for example), fresh sea salt mass contributions were higher with westerly winds and higher wind speeds, which provides greater potential for aerosolization and airborne transport of sea salt particles from the Pacific Ocean. Habre et al. [23] found sea salt mass contributions to PM2.5 mass in southern California to be highest in coastal communities. As fresh sea salt ages and undergoes photochemical reactions that also lead to secondary O₃ formation, chlorine is lost and sulfates are formed [23, 64]. Thus, aged sea salt resembles fresh sea salt in its loading profiles, except with S substituting Cl. Lower wind speed, stagnant conditions, and warmer temperatures provide more chemical reaction time and contribute to the loss of CI and formation of O₃ as fresh sea salt ages [65, 66]. Given the mild, coastal southern California climate year-round, it is not surprising to see both outdoor sea salt sources contributing to personal PM_{2.5} exposures of pregnant women in our study.

As for the crustal source, crustal elements such as Al, Ca, and Ti are naturally present in the earth's crust. They originate outdoors and can enter the indoor environment as windblown dust or as dust tracked indoors on residents' shoes. Once indoors, crustal materials will typically settle and get resuspended as indoor sources (or emissions of indoor origin) when disturbed by human movement or other activities (i.e., vacuuming). Therefore, the presence of more occupants in a household provides greater opportunities for re-suspension of crustal dust, similar to earlier studies [26]. Despite crustal being positively correlated with the traffic source, it did not have any loadings of known traffic-related markers (BC, Ba, Zn, Cu) suggesting it was not reflecting a road dust signal [67]. As such, crustal can be thought of as a predominantly indoor origin source despite the possibility of our participants getting exposed to crustal dust outside of their homes, in daily commutes, and outdoor activities.

Finally, we found that fuel oil and traffic sources contributed to personal PM_{2.5} exposures as well. Similar to previous studies, the fuel oil source had high loadings of Ni and V which are known tracers of heavy residual fuel oil combustion in industrial applications, certain heavy-duty machinery, and in marine engine emissions [68–72]. BC serves as a traffic marker especially for diesel engines [16, 26], while Zn, Ba, and Fe are related to motor vehicle exhaust emission, brake and tire wear, and diesel additives [73, 74]. Traffic is an important source of air pollution in Los



Fig. 2 Relationship between source mass contributions (y-axis) and environmental or home characteristics. a Secondhand smoking and home type, **b** aged sea salt and window opening time in the 48-h monitoring period, **c** fresh sea salt and average wind direction in the 48-h monitoring period, and **d** crustal and number of household occupants (*The relationships in (**b d**) were significant with Kruskal-Wallis test p value < 0.05) $\stackrel{\leftarrow}{\to}$ $\frac{Boxplot}{Mean}$.

Angeles, CA [23, 25]. Our study shows that greater concentrations of traffic-related pollutants outdoors (CALINE NO_x and ambient NO_2) and spending more time near primary roads (in actual activity spaces) correlate with greater personal exposure to the $PM_{2.5}$ traffic source. It is also important to note that the traffic source had high loadings of tailpipe combustion markers (BC) and non-exhaust brake and tire wear markers (Ba and Zn, respectively) potentially capturing both tailpipe and non-tailpipe traffic exposures.

The strengths of this study include the personal $PM_{2.5}$ measurements and detailed chemical composition analysis that allowed us to apportion the major sources of personal $PM_{2.5}$ exposure during a critical time window of pregnancy. By integrating concurrently collected questionnaire data and geospatially modeled environmental exposures in activity spaces (from GPS) and in the residential neighborhood, the results further corroborate these sources, their determinants, and origin (primarily indoor vs outdoor). However, the PMF model only explained a portion of the variability in personal $PM_{2.5}$ mass concentrations (R^2 0.48). One possible reason could be that we did not measure organic carbon (OC) species on the Teflon filters in this study which are known to contribute a large fraction of indoor $PM_{2.5}$ mass concentrations [26, 75]. It was also possible that some of most volatile OC species and ammonium nitrate among others

volatilized off the filters; we found higher temperature was associated with lower personal $PM_{2.5}$ mass in bivariate analysis [38]. We also did not measure volatile organic compounds or other species that could potentially contribute significantly to personal air pollution exposures (in different phases, and not just in $PM_{2.5}$). The sample size of the study, while considered large in personal monitoring settings, and the short monitoring period may limit the generalizability and representativeness of personal $PM_{2.5}$ exposures beyond this study area and across the full pregnancy and postpartum periods.

CONCLUSION

This is one of the few studies to conduct a thorough characterization of sources impacting personal $PM_{2.5}$ exposures of predominantly Hispanic and low-income women during pregnancy in an environmental health disparities context. Our findings show the diversity of sources that impact personal $PM_{2.5}$ exposures in pregnancy in Los Angeles, CA. With ~ 77% of personal exposures contributed by indoor sources, the findings highlight the importance of the indoor environment contributions to total $PM_{2.5}$ exposures during pregnancy and the potentially incomplete understanding of this population's exposures by solely relying on outdoor air pollution measures. These results may

inform source-specific health investigations and subsequent interventions to reduce exposure to potentially more harmful components of $PM_{2.5}$.

DATA AVAILABILITY

The datasets generated during and/or analyzed during the current study are not publicly available due to human subjects research protections but are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

Yan Xu: Formal analysis; Investigation; Methodology; Visualization; Roles/Writing original draft; Writing—review & editing. Karl O'Sharkey: Investigation; Writing—review & editing. Jane Cabison: Data curation; Investigation; Methodology; Project administration; Writing—review & editing. Marisela Rosales: Data curation; Investigation; Project administration; Writing—review & editing. Thomas Chavez, Mark Johnson, and Tingyu Yang: Data curation; Writing—review & editing. Seung-Hyun Cho and Ryan Chartier: Data curation; Writing—review & editing. Brendan Grubbs: Data curation; Writing review & editing. Nathana Lurvey and Deborah Lerner: Data curation; Writing—review & editing. Fred Lurmann: Data curation; Writing—review & editing. Shohreh F. Farzan: Investigation; Writing—review & editing. Carrie Breton: Data curation; Funding acquisition; Investigation; Project administration; Resources; Writing—review & editing. John Wilson: Investigation; Investigation; Investigation; Methodology; Project administration; Resources; Supervision; Visualization; Writing—review & editing.

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COMPETING INTERESTS

The authors declare no competing interests.

ETHICAL APPROVAL

Study procedures were approved by the USC IRB and all participants completed written informed consent at first study visit (IRB: HS-16-00530).

ADDITIONAL INFORMATION

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