Road proximity influences indoor exposures to ambient fine particle mass and components

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A R T I C L E  I N F O

Article history:
Received 23 February 2018
Received in revised form 5 September 2018
Accepted 7 September 2018
Available online 17 September 2018

Keywords:
Indoor exposure
Fine particle (PM2.5)
Metal elements
Traffic
Road proximity

A B S T R A C T

Exposure to traffic-related PM2.5 mass and its components can affect human health. Meanwhile, indoor concentrations are better exposure predictors as compared to outdoor concentrations because individuals spend the majority of their time indoors. We estimated the impact of traffic emissions on indoor PM2.5 mass and its species as a function of road proximity in Massachusetts. A linear regression model was built using 662 indoor samples and 580 ambient samples. Analysis shows that indoor exposures to traffic-related particles increased dramatically with road proximity. We defined relative concentration decrease, \( R(\alpha) \), as the ratio of the indoor concentration at perpendicular distance \( \alpha \) in meters from the closest major road to the indoor concentration at 1800 m from the major road. \( R(13) \) values for PM2.5 mass and Black Carbon (BC) were 1.3 (95%CI: 1.4, 1.6) and 2.1 (95%CI: 1.3, 2.8) for A12 roads, and 1.3 (95%CI: 1.2, 1.4) and 1.2 (95%CI: 1.1, 1.3) for A3 roads. \( R(\alpha) \) values were also estimated for Fe, Mn, Mo, Sr and Ti for A12 roads, and Ca, Cu, Fe, Mn, Mo, Ni, Si, Sr, V and Zn for A3 roads. \( R(\alpha) \) values for species associated mainly with brakes, tires or road dust (e.g., Mn, Mo and Sr) were higher than others. For A12 roads, \( R(13) \) values for Mn and Mo were 10.9 (95%CI: 0.9, 20.9) and 6.5 (95%CI: 1.4, 11.5), and ranged from 1.3 to 2.1 for other species; for A3 roads, \( R(13) \) values for Mn, Mo and Sr were 1.9 (95%CI: 1.1, 2.9), 1.8 (95%CI: 1.1, 2.4), and 8.5 (95%CI: 5.9, 10.9), and ranged from 1.2 to 1.6 for others. Our results indicate a significant impact of local traffic emissions on indoor air, which depends on road proximity. Thus road proximity which has been used in many epidemiological studies is a reasonable exposure metric.

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1. Introduction

Particulate pollution is a major public health risk worldwide. Of particulate pollutants, PM2.5 - fine particles defined as particulate matter with aerodynamic diameter lower than 2.5 μm is derived from various sources (WHO, 2000; Tucker, 2000). Traffic-derived PM2.5 is recognized as an important contributor to outdoor concentrations (Gertler et al., 2000). Exposure to traffic-related PM2.5 has been associated with a wide range of human adverse health effects including asthma onset and exacerbation, lung growth deficits, increased blood pressure, inflammation and left ventricular mass index, decreased high-frequency heart rate variability, coronary heart disease hospitalizations and mortality, and low birth weight (Carlsten et al., 2011; Gan et al., 2011; Gauderman et al., 2005, 2007; Hoffman et al., 2012; Jerrett et al., 2008; McConnnell et al., 2006, 2010; Van Hee et al., 2010; Wilhelm et al., 2012; Williams et al., 2009; Zanobetti et al., 2010; Zeka et al., 2006). PM2.5 consists of numerous components, including organic and elemental carbon, metals, and ions. In terms of sources, PM-associated elements usually originate from road dust and crustal material (e.g., Ca, Fe, Ti, Al, Si), oil combustion (e.g., Ni and V), traffic (e.g., BC, Zn and Cu), wood burning (e.g., K), sea/road salt (e.g., Na and Cl) and regional pollution (e.g., S) (Bell et al., 2010; Dai et al., 2016).

As people spend 85–90% of their day indoors (Chen and Zhao, 2016),
studies used sulfur as a tracer to approximate the indoor PM$_{2.5}$ mass and its composition as a function of road proximity and indoor exposures. However, existing studies to investigate the effect of road proximity on indoor traffic-related exposures are limited, especially for smaller numbers of homes. For example, Baxter et al. (2008, 2008) investigated the influence of traffic-related parameters on indoor concentrations of PM$_{2.5}$ mass and elemental carbon in Boston, MA, a study which only included 16 and 43 homes, and Martuzevicius et al. (2008) confirmed the importance of traffic components in the indoor environment in a study of 6 homes. In addition, particle infiltration factor, which varies by housing characteristics and human activities, is a key parameter for modelling (Meng et al., 2009; Morawska et al., 2001; Tang et al., 2017; Van Der Zee et al., 2016). Previous studies used sulfur as a tracer to approximate the infiltration of PM$_{2.5}$ mass from outdoor sources (Gaffin et al., 2017; Sarnat et al., 2002; Wallace and Williams, 2005). Having many indoor measurements would be essential to investigate the effect of road proximity on indoor influence of traffic-related particles, accounting for the difference in infiltration behaviours.

In this study, we estimate the impact of traffic emissions on indoor PM$_{2.5}$ mass and its composition as a function of road proximity using 662 samples collected at 340 homes in Massachusetts. This study examines the appropriateness of exposure metrics such as road proximity based on numerous locations and samples, and therefore is of particular importance to epidemiological studies on health impacts of traffic-related particle exposures.

2. Materials and methods

2.1. Indoor and outdoor measurements

Our analysis was based on 662 indoor PM$_{2.5}$ samples collected at 340 homes in Massachusetts during 2006–2011 as a part of two cohort studies: the Normative Aging Study (NAS) (Baja et al., 2010); and the Diabetes, Cardiac Disease, and Pollution Vulnerability Study (DCDPV) (Hoffman et al., 2012). Each indoor sample was collected on a Teflon filter at a flow rate of 1.8 L/min in the main activity room (typically the family room or living room) for 5 or 7 days (Hoffman et al., 2012; Baja et al., 2010). After collection, the sampler was returned to the lab for pollutant concentration analysis. For each home, 1–7 samples were collected during different seasons (summer, winter or transition). About 65% of homes were only collected once (Hoffman et al., 2012; Baja et al., 2010). In addition, 580 daily ambient PM$_{2.5}$ filters were collected at the central monitoring Supersite located on the roof of the Countway Library of the Harvard Medical School in downtown Boston. The Supersite is located on the roof of a 6 story building about 60 m from the nearest major road. Several studies have described that the Supersite has non-significant effect of local point sources (Kang et al., 2010; Tang et al., 2017).

Both indoor and Supersite filters were analysed for PM$_{2.5}$ mass, Black Carbon (BC), and trace elements. Teflon filters were weighed using an electronic microbalance (Model MT-5, Mettler Toledo) to and after sample collection. Subsequently, indoor BC concentrations were determined by measuring the blackness of the particles collected on the filter using a smoke stain reflectometer (model EEL m43d, Diffusion System Ltd., United Kingdom). Additionally, samples were analysed for 48 trace elements from Na to Pb by energy-dispersive x-ray fluorescence spectrometer (Epsilon 5, PANAlytical, The Netherlands). As each indoor concentration was an integrated 5 or 7 day measurement, we calculated the concurrent Supersite concentration by taking the average of daily concentrations for the same days.

2.2. Traffic-related parameters

National Emission Inventories data in 2008 and 2011 (NEI, 2008; NEI, 2011) shows that, annual PM$_{2.5}$ emission from roadway traffic (which included exhaust, brake, and tire wear emissions from light and heavy duty diesel and gasoline vehicles, as well as suspended road dusts) was above 16 000 tons during our study period in Massachusetts, which contributed nearly half of the total PM$_{2.5}$ emission in Massachusetts. Therefore, road proximity is probably to be a reasonable predictor for indoor exposures. In order to examine this, we considered the perpendicular distance (in meters) between a home and the closest of three major road types as a surrogate for traffic exposure. The definitions of these road types from the US Census (Census Feature Class Codes) are as follows: an A1 road is a primary road with limited access via onramps and exit ramps; an A2 road is a primary road without limited access, and; an A3 road is a secondary and connecting road. The perpendicular distance of each home to the center of the nearest major road was calculated using Street Map™ North America ArcGIS 10 Data and Maps. We did not consider the elevation difference as the elevation variability can be ignored compared with the Latitude and longitude in Massachusetts. We then estimated the width of each road based on the information of the number of lanes and got the distance to the boundary after subtracting half of the width. Traffic densities, speed, and diesel vehicle fraction are likely to differ between A1, A2, and A3 roads. This may result in different traffic-related emissions and thus different indoor exposures. We classified major roads into two categories, A12, (combining categories A1 and A2 to represent primary roads), and A3 (secondary and connecting road), instead of three types. Previous studies have shown that particle levels diminish to background levels 100–300 m from major roads (Wilker et al., 2014; Zhou and Levy, 2007). As the traffic density of A12 road is generally much higher than that of A3, we neglected the influence of A3 when the distance from a residence to an A3 road was less than 200 m closer than the distance of the residence to an A12 road. Otherwise, we only considered the influence of an A3 road.

2.3. Statistical analysis

As 35% homes have multiple measurements, we used a mixed linear regression model to examine the relationship between indoor concentrations and the perpendicular distance to the nearest major road. Considering that sulfur is a regional pollutant and exhibits little spatial variability, which can be ignored within a small geographical area (Brown et al., 2008a, 2008b; Tang et al., 2017), we used the sulfur concentration measured at the Supersite site as a
surrogate for the sulfur concentration outside homes. The indoor/outdoor sulfur ratio was used to estimate the magnitude of outdoor PM$_{2.5}$ penetrating indoors (Sarnat et al., 2002). Assuming no other local source with direct impact on indoor PM$_{2.5}$ exposure, indoor PM$_{2.5}$ of outdoor origin in these homes can be considered from two sources: local road traffic and general urban emission. According to previous studies, meteorological conditions have effects on PM$_{2.5}$ (Wang and Ogawa, 2015). We tried seasonality, temperature and wind speed as candidate predictors in our model. And finally, we chose seasonality and wind speed as predictors in our model, because the Bayesian Information Criterion (BIC) value was the lowest when we adjusted for the two predictors (i.e., seasonality and wind speed). The model is formulated as follows:

\[
C_{ij} = \beta_0 + \gamma_{aij} + \beta_{3j} \times \log (a_j) \times (SI/SO)_{ij} + \beta_{2i} \times CC_{ij} \times (SI/SO)_{ij} + \beta_{3j} \times \cos d + \beta_{4j} \times \log (ws) + e_{ij}
\]

\[
(1)
\]

where, $C_{ij}$ is the indoor concentration of a species $i$ in home $j$; $SI$ and $SO$ are the concurrent indoor and outdoor sulfur concentrations, respectively; $CC_{ij}$ is the Supersite concentration which represents the impact of general urban emissions of species $i$ for home $j$; $\alpha_i$ is the perpendicular distance (in meters) from home $j$ to the nearest major road; $\cos d$ is a continuous variable which represents the seasonality, $(\cos d = \cos (2\times \pi \times d/365.25), d$ is a continuous variable and equal to 1 when the date is 01/01/1960 (it is the default value in SAS), and $d$ increases by 1 as date increases (Piegorsch and Bailer, 2005); and $ws$ is the wind speed obtained from the weather station at Boston Logan airport. $\beta_{3j} \times \log (a_j) \times (SI/SO)_{ij}$ represents the concentration of a species $i$ that penetrated indoors as a result of traffic emissions from the nearest major road; $\beta_{3j} \times CC_{ij} \times (SI/SO)_{ij}$ represents the concentration of species $i$ that penetrated indoors as a result of general urban emissions; $\beta_{3j} \times \cos d$ represents the influence of season; $\beta_{4j} \times \log (ws)$ represents the influence of wind speed; $\beta_{4j}$ is intercept which represents the concentration of species originating indoors; $\gamma_{aij}$ is the random intercept which accounts for the distribution of indoor sources in different homes; $e_{ij}$ is the random error. Examination of linear regression assumptions including the distributions of variables as well as the random errors, and are shown in Supplementary data.

Although the terms $\log (a_j) \times (SI/SO)_{ij}$ and $CC_{ij} \times (SI/SO)_{ij}$ all include the sulfur ratio, they are only moderately correlated with a correlation coefficient of less than 0.25.

As models were formulated for A12 and A3 roads separately using different exclusion criteria as stated in section 2.2, the observations were different for two models. Furthermore, we excluded the homes with $SI/SO$ ratios higher than 1.2 from further analysis, since they might have indoor sources of sulfur that make it impossible to use sulfur as a tracer of outdoor particles. Similarly, $CC_{ij}/CC_{ij}$ ratios higher than unity suggest the presence of important indoor sources of a species $i$ and these homes were also excluded from the analysis. Finally, we excluded about 30% homes from the original dataset. Therefore, this model is only predictive for homes that don’t have substantial indoor sources.

It should be noted that we only measured the outdoor concentrations in Supersite and we used the Supersite concentrations as surrogate for the outdoor concentration from the impact of general urban emissions. This is discussed in the Supplementary data.

Analyses were performed using SAS software package 9.3 (SAS Inc. NC, USA). When $p$-value is less than 0.05, we reject the null hypothesis and consider the result as significant. When $0.05 \leq p$-value < 1, we consider the results as marginally significant.

3. Results

3.1. Indoor and outdoor concentrations

Table 1 shows the indoor and outdoor concentrations of PM$_{2.5}$, BC and selected elements (Al, Br, Ca, Cl, Cr, Fe, K, Mg, Mn, Mo, Na, Ni, S, Sb, Si, Sn, Sr, Ti, V and Zn). The method detection limits (MDL) from method blank results are shown in Table S1 in Supplementary data. We kept the concentrations even they are below the MDLs. This is because MDL is not good criteria to estimate each sample's significance. Even the concentrations below MDLs have physical meanings, which means the home is far away from the major road. Indoor concentrations of selected species showed high variability across homes. The averages of indoor PM$_{2.5}$ and BC concentrations were $7.708 \pm 4.025$ and $0.438 \pm 0.271 \mu g/m^3$, ranging between 0.204-23.955 and 0.006-1641 $\mu g/m^3$, respectively. The averages of outdoor PM$_{2.5}$ and BC concentrations at the Supersite were $8.659 \pm 3.439$ and $0.626 \pm 0.176 \mu g/m^3$, ranging between 2.494-21.033 and 0.201-1.192 $\mu g/m^3$, respectively. Supersite concentrations of the PM$_{2.5}$ and selected elements are all significantly higher than the corresponding indoor concentrations on average ($p < 0.01$ for T-test results for all selected species), indicating the importance of the outdoor sources for these species.

Tables S2 and S3 in the Supplementary data show the Pearson correlation coefficients ($r$) between pollutants for Supersite and indoor respectively. According to Table S2, r values were no less than 0.7 between Al and Ca, Al and Si, Al and Ti, Ca and Fe, Ca and Si, Cu and K, Cu and Sr, K and Sr, K and Ti, Mg and Na, Sr and Ti, Sr and V, Sr and Zn. This indicates strong linear relationships between these pairs. One reason for this is that the strong correlated pairs are mainly from the same source. For example, Al, Ca and Si usually originate from road dust and crustal material; Sr and Ti from road or urban dust re-suspension. According to Table S3, all of the r values were less than 0.7 (the largest $r$ is 0.57, between Ca and Sr), indicating no strong linear relationship between these pollutants. A possible reason for this is that a main portion of pollutants are from outdoor, which decay differently with distance. In addition, there might be sources indoor for some pollutants, which can also weaken the linear relationship between the indoor and outdoor concentrations.

3.2. Influence of road proximity on indoor concentrations

Fig. 1 shows the distribution of perpendicular distances to the nearest A3 and A12 roads for homes included in our model. Fig. 1 shows that there is a large range of distances from homes to the nearest A3 and A12. Distance from homes to the nearest A3 ranged from 3 m to 1886 m, with mean value of 215 ± 265 m, and median of 119 m. Distance from homes to the nearest A12 ranged from 13 m to 1991 m, with mean value of 828 ± 527 m, and median of 753 m. It should be noted that the influence of A12 roads on indoor concentrations was small when homes are far away from them. Therefore, when we modelled the influence of proximity to A12 roads, we only included 462 samples within 2000 m from A12 roads. Homes outside this range are mostly in rural areas where land use and micro-meteorology differ from those in urban and suburban areas where most of our study homes are located. Distance to the nearest A12 roads for all the homes has a mean value of 1869 ± 2177 m, and median value of 1 191 m, with a range from 13 m to 12 586 m as shown in Figure S5 in the Supplementary data.

The modelling results are presented in Table 2. With A12 roads, significant relationships between indoor concentrations and perpendicular distance were found for PM$_{2.5}$, BC, Fe and Mo, and marginally significant for Mn and Ti. With A3 roads, significant relationships were found for PM$_{2.5}$, BC, Ca, Fe, Mn, Mo, Ni, Si, Sr and...
Zn, and marginally significant for V. The species with significant relationships of indoor concentration to the log of distance to the nearest major road (log distance) were more likely associated with traffic emissions. According to previous studies, BC is most strongly associated with direct tailpipe emissions; Zn with direct tailpipe emissions and with tire wear; Fe, Ca, Si and Mn with re-suspended road dust; Ni and V from on-road diesel fuel combustion (Amato et al., 2011; Cadle et al., 1997; Garg et al., 2000; Gertler et al., 2002; Kam et al., 2012; Lough et al., 2005; Ning et al., 2008), Al (soil), K (wood burning), Na and Cl (sea salt) (Dai et al., 2016) (see Table 3).

3.3. Relationship between relative concentration decrease and road proximity

From the results of the linear model shown in Table 2, we estimated the decrease concentrations of indoor species of outdoor origin as a function of the log distance. The effect of the log distance was examined only for the species with significant or marginally significant log(\(a\))/\(S_i/S_o\) slope values (\(\beta_i\)) which were associated with traffic. For a simpler interpretation of the results, we define the relative concentration decrease of a species \(i\) at a distance to major road, \(R_i(\alpha)\), which is given by the following equation:

\[
R_i(\alpha) = \frac{C_i(\alpha)}{C_i(1800)}
\]

Where, \(C_i(\alpha)\) and \(C_i(1800)\) are the indoor concentrations of species \(i\) at distance to major road of \(\alpha\) and 1800 m, respectively. These concentrations were calculated using equation (1) with average Supersite concentration, average indoor/outdoor sulfur ratio, average wind speed, and winter season. A few homes with over 1800 m were excluded for a better interpretation: one home for an A3 road and 20 homes for A12 roads were excluded by this consideration.

The results for A12 and A3 roads are presented in Figs. 2 and 3, respectively. \(R_i(\alpha)\) values representing perpendicular distance (\(\alpha\)) to the nearest major road ranged from 13 to 1800 m for A12, and from 3 to 1800 m for A3. For A12 roads, the \(R_i(13)\) values of PM\(_{2.5}\) and BC were 1.3 (95%CI: 1.1, 1.5) and 2.1 (95%CI: 1.3, 2.8) respectively. This indicates that indoor concentrations of PM\(_{2.5}\) and BC at a home located 13 m from a road are 1.3 (95%CI: 1.1, 1.5) and 2.1 (95%CI: 1.3, 2.8) times those of a home located 1800 m from a road; the \(R_i(13)\) values of Mn (10.9 (95%CI: 0.9, 20.9)) and Mo (6.5 (95%CI: 1.4, 11.5))

### Table 1
Concentrations of indoor and outdoor PM\(_{2.5}\), BC and selected elements.

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Indoor concentration (µg/m(^2))</th>
<th>Supersite concentration (µg/m(^2))</th>
<th>Ratio of Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>N Mean ± SE</td>
<td>Median</td>
<td>Min Max</td>
</tr>
<tr>
<td>PM(_{2.5})</td>
<td>650 7.708 ± 4.025</td>
<td>6.72</td>
<td>0.204</td>
</tr>
<tr>
<td>BC</td>
<td>598 0.438 ± 0.271</td>
<td>0.422</td>
<td>0.006</td>
</tr>
<tr>
<td>S</td>
<td>662 0.583 ± 2.027</td>
<td>0.377</td>
<td>0.000</td>
</tr>
<tr>
<td>Al</td>
<td>652 0.008 ± 0.011</td>
<td>0.004</td>
<td>0.000</td>
</tr>
<tr>
<td>Br</td>
<td>656 0.001 ± 0.001</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Ca</td>
<td>634 0.021 ± 0.011</td>
<td>0.019</td>
<td>0.000</td>
</tr>
<tr>
<td>Cl</td>
<td>624 0.016 ± 0.020</td>
<td>0.008</td>
<td>0.000</td>
</tr>
<tr>
<td>Cr</td>
<td>640 0.000 ± 0.000</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Cu</td>
<td>631 0.002 ± 0.002</td>
<td>0.002</td>
<td>0.000</td>
</tr>
<tr>
<td>Fe</td>
<td>654 0.022 ± 0.013</td>
<td>0.019</td>
<td>0.000</td>
</tr>
<tr>
<td>K</td>
<td>623 0.031 ± 0.018</td>
<td>0.026</td>
<td>0.000</td>
</tr>
<tr>
<td>Mg</td>
<td>645 0.005 ± 0.008</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Mn</td>
<td>659 0.001 ± 0.001</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Mo</td>
<td>636 0.001 ± 0.001</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Na</td>
<td>648 0.087 ± 0.066</td>
<td>0.072</td>
<td>0.000</td>
</tr>
<tr>
<td>Ni</td>
<td>656 0.001 ± 0.001</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Pb</td>
<td>652 0.002 ± 0.002</td>
<td>0.001</td>
<td>0.000</td>
</tr>
<tr>
<td>Sb</td>
<td>639 0.001 ± 0.002</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Si</td>
<td>639 0.028 ± 0.022</td>
<td>0.023</td>
<td>0.000</td>
</tr>
<tr>
<td>Sn</td>
<td>556 0.000 ± 0.000</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Sr</td>
<td>662 0.000 ± 0.000</td>
<td>0.000</td>
<td>0.000</td>
</tr>
<tr>
<td>Ti</td>
<td>645 0.002 ± 0.001</td>
<td>0.001</td>
<td>0.000</td>
</tr>
<tr>
<td>V</td>
<td>652 0.001 ± 0.001</td>
<td>0.001</td>
<td>0.000</td>
</tr>
<tr>
<td>Zn</td>
<td>647 0.005 ± 0.003</td>
<td>0.004</td>
<td>0.000</td>
</tr>
</tbody>
</table>

Footnotes: SE represents standard error; Min and Max represent the minimum and maximum concentrations respectively; Ratio represents the indoor and outdoor ratios for pollutants; N is the number of observations.
were considerably higher than PM2.5 and BC, while the R^2 (13) value of Fe (1.4 (95%CI: 1.0, 1.8)) was lower than BC and similar to PM2.5. For A3 roads, the R^2 (3) values of PM2.5 and BC were 1.5 (95%CI: 1.4, 1.6) and 1.3 (95%CI: 1.1, 1.5), respectively; R^2 (3) values of Mn (2.2 (95%CI: 1.1, 3.5)), Mo (2.0 (95%CI: 1.1, 2.9)) and Sr (11.0 (95%CI: 7.5, 14.0)) were higher; R^2 (3) values of Ni, Fe, Cu, Si, V, Ca and Zn were relatively lower, ranging from 1.2 to 1.7 (95%CI values are shown as dash lines in Fig. 2). In order to compare the different impacts of A12 and A3 roads on indoor concentrations, a dashed dot vertical line indicating the R^2 (13) values for A3 roads was drawn in Fig. 3.

The R^2 (13) values of PM2.5 and BC were 1.3 (95%CI: 1.2, 1.5) and 1.2 (95%CI: 1.1, 1.3) respectively; R^2 (13) values of Mn, Mo and Sr were 1.9 (95%CI: 1.1, 2.9), 18 (95%CI: 1.1, 2.4), and 8.5 (95%CI: 5.9, 10.9); R^2 (13) values of Ni, Fe, Cu, Si, V, Ca and Zn ranged from 1.2 to 1.6 (95%CI values are shown as dash lines in Fig. 3).

3.4. Model validation and sensitivity test

We compared the predicted concentration for indoor PM2.5 and BC of our model and Tang et al. (2017) model with measurement data.
data. The comparison results are shown in Figure S6 in the Supplementary data. For PM$_{2.5}$, $R^2$ values are both 0.81 for our model and Tang et al.’s model. For BC, $R^2$ values are 0.76 and 0.75 for our model and Tang et al.’s model. According to Figure S6, the prediction concentrations by the two models are very close. The comparison results indicate that “the distance to the nearest major road” can be used as a predictor for the indoor traffic-related concentration instead of land use parameters (i.e., road traffic density and urban percentage) in Tang et al. (2017) model.

For sensitivity test, observations were deleted when the indoor PM$_{2.5}$ concentrations were above top 10% percentile. $R_i(13)$ values of PM$_{2.5}$ for A$_{12}$ and A$_3$ changed less than 7.6% (1.3 and 1.2 for A$_{12}$ and A$_3$ respectively compared with 1.3 and 1.3 for the original dataset), indicating the robustness of the results of the model.

4. Discussion

In this study, we investigated the influence of road proximity on the concentration of indoor PM$_{2.5}$ mass and its components, including BC, S, and 21 trace elements. Our analysis was based on 662 indoor samples from 340 homes and 580 ambient samplers collected at the central monitoring Supersite in Massachusetts. We found strong relationships between road proximity and indoor concentrations of traffic-related species. We also found that the impact of traffic emissions on indoor concentrations depends on particle penetration rates as determined by using indoor/outdoor sulfur ratios.

For most of the species, $p$-values of the slope for $\log(a_j) \times (S/SO)$ were smaller for A$_3$ roads than those for A$_{12}$. This is possibly because the homes are much closer to A$_3$ than A$_{12}$ roads (see Fig. 1) however this does not necessarily mean a stronger effect of A$_3$ roads. Furthermore, a linear relationship was established between the relative concentration decrease $R_i(13)$ and the log of the distance to a major road. According to our results, the indoor BC concentration decreased more sharply with distance to A$_{12}$ roads as compared to A$_3$ roads. This is expected since the traffic emissions are generally larger for A$_{12}$ roads. The relative concentration decreases differ by species, possibly due to the relatively different contribution of local traffic emissions to their concentrations. For instance, the BC concentration decrease as a function of road

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Fig. 2. The relationship between relative concentration decrease and the log of perpendicular distance to A$_{12}$ road.
proximity is considerably higher than PM2.5. PM2.5 is largely associated with regional sources, and local traffic contributes a small fraction of its total concentration, while in comparison local traffic emissions contribute a significant fraction of BC concentrations (Masri et al., 2015; Schwartz et al., 2005). Similarly, concentration decreases are sharper for elements (e.g., Mo, Mn, and Sr) which have significant local traffic sources such as the non-tailpipe emissions (brakes and tires) or resuspension (Amato et al., 2011; Ning et al., 2008; Cheung et al., 2010). Another reason for different rates of decrease is that these species are associated with particles in different size distributions, which results in different deposition rates. Species associated with relatively larger particles (e.g., Mo, Mn, and Sr from brakes, tires or road dust) have higher settling velocities and may decrease more sharply, while species associated with submicron particles can travel relatively longer distances (e.g., BC, Cu and Zn from tailpipe emissions) (Dongarra et al., 2009; Kaiser, 2003). The relative concentration decreases of Ni and V for A3 road are very close and relatively low among traffic-related species, with R(13) values equal to 1.3 (95%CI: 1.0, 1.6) and 1.2 (95%CI: 1.0, 1.4) respectively. The reason for this may be that Ni

Fig. 3. The relationship between relative concentration decrease and the log of perpendicular distance to A3 road.
and V are not only emitted from traffic (i.e., on-road diesel fuels combustion), but are also significantly associated with residential fuel oil combustion (Peltier and Lippmann, 2010; Cui et al., 2017). In  
addition, different analytical uncertainties for each species might also influence the results.

As we repeated measurements were made in 35% of the homes, we used mixed linear model including the random intercept $\gamma_0$ to capture the difference of indoor sources in different homes instead of simple linear model which did not account for this difference. The $p$-value of the likelihood ratio test of the two models was less than 0.05, which means the random intercept was significant. Therefore, we cannot ignore the difference of different homes and included the random intercept in our final model.

Though we only considered homes with indoor pollutants originating mainly from outdoors, the indoor sources for some pollutants were significant. This is reflected by a significantly positive $\beta_0$ value, which represents the concentrations of species originating indoors. For PM$_{2.5}$ and BC, these values were similar for different types of major roads (i.e., A3 and A12). This is reassuring because although the contribution of different types of roads on the indoor exposure are different, the indoor emission for the homes closer to A3 and those closer to A12 should be similar.

According to Table S4 and S5, the season and variable lead can affect indoor concentrations of Na, V, and Zn significantly for A12 road, and concentrations of BC, Ca, Cl, Cu, Fe, and Zn for A3 road. This is mainly due to different suspension characteristics and industrial production amounts in different seasons (Suvarapu and Beak, 2016). Wind speed was found to be significantly related to the indoor concentrations of Ca for A12 road, and concentrations of BC, Cu, K, Mg, and Zn for A3 road. The effects of wind speed were negative for most of the above elements. According to previous studies (Stortini et al., 2009; Suvarapu and Beak, 2016), lower wind speed enhances the concentrations due to less diluting capability. However, the effect was positive for Mg. The possible reason is that Mg is a common cation in oceans, and the production of magnesium chloride by evaporation is enhanced at higher wind speed, which can be a source of road dust (Post, 1999; Yu, 2007).

In this study, we only considered the distance to the nearest major roadway rather than considering all roadways for three reasons: (1) Distance to the nearest major road has been used as an exposure metric in many epidemiological studies (Lue et al., 2013; Rosenbloom et al., 2012; Wilker et al., 2013, 2016). The main purpose of our paper is not the prediction for indoor exposures, but to examine if road proximity is a reasonable exposure metric and also in addition, different analytical uncertainties for each species might also influence the results. (2) For the distances from homes to minor roads, there is not as much difference as the distances to the major roads, and the traffic emission is much lower compared to major roads; (3) We also checked with ArcGIS that, the major roads are not overlapping as the minor roads. Even for the second closest major roads for A3/ A12, the distances to homes are mostly at least 150m larger than the nearest one. Other studies (Wilker et al., 2014; Zhou and Levy, 2007) suggest that the influence of a road can be ignored when distance is more than 150m. Therefore, we just look at the distance to the single closest major road to each home.

There are some limitations in our study. One limitation is that as our study focused on a single metropolitan area, our findings may not be generalizable to other geographical areas, where housing characteristics and weather conditions may differ. Another limitation is that we only measured outdoor concentrations at a central site, which may reduce to some bias for the results. However, our regression results show that the impact of distance from Supersite on indoor concentration is not significant. The comparison results of our predicted concentrations with measured concentrations as well as with Tang et al. (2017) model which considered the distance to Supersite also indicated the effectiveness of our model.

The major strengths of this study are as follows: 1) We used a large data set of indoor exposures (662 samples collected at 340 homes), which provided ample statistical power to examine relationships between indoor pollutant concentrations and road proximity; 2) Our pollutants included not only PM$_{2.5}$ mass and BC, but also a wide range of traffic-related species; 3) We used road proximity as surrogate for exposure to traffic, which is more simpler and more intuitive compared with exposure metric in previous studies (e.g., traffic density); 4) A large number of homes are close to main roads, which will enable us to statistically determine concentration gradients near roads where there is rapid change.

5. Conclusions

We investigated the influence of road proximity on the concentration of indoor PM$_{2.5}$ mass and its components based on 662 samples collected at 340 homes in Massachusetts. Strong linear relationships were found between road proximity and indoor concentrations of traffic-related species, indicating that traffic emissions contribute substantially to indoor exposures. A linear relationship was established between the relative concentration decrease $R(x)$ and the log of the distance to a major road. Differences in relative concentration decrease among the examined particle species are due to the relative contribution of local traffic to pollutant concentrations and different size distributions. The $R(13)$ values of PM$_{2.5}$ mass and BC for A12 roads were 1.3 (95%CI: 1.1, 1.5) and 2.1 (95%CI: 1.3, 2.9), respectively; and the $R(13)$ values of PM$_{2.5}$ mass and BC for A3 roads were 1.3 (95%CI: 1.2, 1.4) and 1.2 (95%CI: 1.1, 1.3), respectively. Relative concentration decreases for Mo, Mn and Sr, which are mainly from brakes, tires or road dust, were even more dramatic: for A12 roads, the relative concentration decrease values of these elements were 3.1–8.4 times of other species; and for A3 roads, the values were 1.2–9.2 times of others. These sharp gradients merit our attention since they suggest that individuals living near busy roads, and not necessarily only highways, are at higher risk.

Acknowledgments

This research was supported by U.S. EPA, United States (grant numbers R835872 and R834798) and NIEHS, United States (grant number ES 000002) and is a research component of the Massachusetts VA Epidemiology Research and Information Center (MAVERIC). This paper is solely the responsibility of the grantee and does not necessarily represent the official views of the U.S. EPA. Further, U.S. EPA does not endorse the purchase of any commercial products or services mentioned in the publication. We also acknowledge the diabetes study supported by NIEHS, (grant number P01 ES009825).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2018.09.046.

References

Meng, Q.Y., Spector, D., Colome, S., Turpin, B., 2009. Determinants of indoor and personal exposure to PM(2.5) of indoor and outdoor origin during the RIOPA study. Atmos. Environ. 43 (36), 5750–5758.


World Health Organization (WHO), 2000. Air Quality Guidelines for Europe, second ed. WHO Regional Office for Europe, Copenhagen, Denmark (Chapter 7.


