

Interactive comment on “Source apportionment of PM_{2.5} in Seoul, Korea” by J.-B. Heo et al.

J.-B. Heo et al.

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Author comments

Response to comments by referee #1:

General remarks:

Referee: More PMF diagnostics need to be provided. This comprises the report of Q-values as a function of the number of factors, as well as a discussion of the model errors, $e_{i,j}$, as a function of time and species. This will make the choice of the number of factors more reproducible. In addition, please provide the factor profiles and time series from the 8- and 10-factor PMF (supporting material).

Answer: In this study, in order to select modeling parameters and the number of factors, we used the mathematical PMF diagnosis and interpretable testing of the plausibility of PMF solutions. Mathematical PMF diagnoses (model error, Q, rotational ambiguity, rotmat, etc.) were based on Lee et. al., 1999. Figure 1 shows the results of the Q-value for the different number of factors and the FPEAK values and the variation of IM, IS, and rotational freedom for the different number of factors chosen in PMF. As the number of factors approaches to a critical value, IM and IS will drop clearly. Also, choosing the maximum element in rotmat will be the worst case in rotational freedom. There was a significant increase

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in maximum rotmat element from nine factors to ten factors (Figure 1). Therefore, nine factors were found to be the most reasonable results.

We added this information about the mathematical PMF diagnosis into the manuscript and the factor profiles and time series from the 8- and 10-factor PMF to the supporting material section.

Referee: As far as I am aware, no novel concepts are included in the present analysis and hence even more emphasis should be put on the interpretations of the statistical results provided by this (standard) analysis. The identification and interpretation of factors as sources lacks support. The estimated factors and the corresponding time series need to be validated by ancillary data (by collocated trace gas measurements CO, NO_x etc.), numerical comparisons with literature source profiles, results of PMF-independent modeling approach etc.

Answer: As reviewer pointed out, we validated the estimated factor profiles and time series through the collocated trace gas measurements and the numerical comparisons with literature source profiles. We added the following paragraphs to the manuscript.

EC/OC ratio in diesel and gasoline engines exhaust is usually much higher for diesel engines than for gasoline engines in some previous source apportionment studies (Table 1) and motor vehicle emission profiles studies (Table 2). In this study, the PMF resolved EC/OC ratios for diesel and gasoline engines were 1.4 and 0.4, respectively and this result agreed well with the previous studies listed in Tables 1 and 2. In general, gasoline-fueled vehicles have higher CO emission, while diesel vehicles tend to have higher NO₂ emissions. According to Pearson correlation analysis between the daily source contributions of motor vehicles deduced from PMF and the gaseous concentration data (CO and NO₂) (Table 3), there was a high correlation between gasoline vehicle factor and CO concentration, while diesel factor was highly correlated with NO₂ concentration compared to CO concentration.

The molar ratio of NH₄⁺ to SO₄²⁻ was 2.5 for secondary sulfate factor resolved from PMF, suggesting that (NH₄)₂SO₄ was dominated sulfate form in PM_{2.5} of Seoul. In addition, the resolved secondary sulfate factor contribution was strongly correlated with the daily concentration of HNO₃ compared to the other factors. In general, photochemical products such as HNO₃ and secondary sulfate could be strongly influenced by SO₂ and NO_x emissions from large point sources and average concentration of these products tends to be high during warmer season. The high (NH₄)₂SO₄ production in summer leaves small amount of NH₃ for the conversion of HNO₃ to NH₄NO₃ resulting in the high correlation between the secondary sulfate factor and HNO₃.

As shown in Figure 2, daily factor contributions were analyzed using wind speeds to improve the PMF resolved factors. Figure 2 showed that most primary factor contributions increased as wind speed decreased (or as the atmospheric condition became stagnant), especially for the motor vehicles (gasoline

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and diesel factors). However, the contributions of soil and aged sea salt factors increased as wind speed increased possibly due to the resuspension and mixing in the air from sources during the strong wind conditions.

Referee: Many of resolved sources reported here have a non-constant emission profile over time. It is not very plausible that this PMF assumption (constant emission profiles) is fully valid for a three-year period, e.g. for biomass burning. How stable is the PMF solution with respect to PMFs on yearly data subsets (as an example)?

Answer: There could be variability in profiles over time. PMF is in effect finding the average profiles over the set of samples collected. In the case of biomass burning, it is hard to see as much variation over time as there is from just the burning conditions. Depending on whether the biomass is flaming or smoldering, there is a substantial difference in the OC and K ratio. However, the profile is still dominated by these species with some EC in there. Thus, the question should really be whether or not there are sufficient differences over time for the factor to split into two factors. We do not see that so although there is some likely variability in the profile, we take that into account by the inflation of the uncertainties in the data points.

Referee: Throughout the distribution of OC to the different factors representing primary as well as secondary components, it would be interesting give estimates for the amount of secondary OC (SOC) and primary OC (POC) and compare them the results from other approaches.

Answer: We agree that it would be interesting to give the estimates for the amount of SOC and POC as well as the evaluation of atmospheric aerosol aging, but these kinds of studies are beyond the scope of this paper. We have been conducting further study for the empirical estimates of SOC and POC, including a multiple regression model employing markers (gas concentrations and particle data) and a carbon mass balance using carbon and CO data, and it will be explored in a future publication.

Specific comments:

Referee: p. 20429, lines 18-19: The reference list here should be extended. The reviewer remains in doubt, if the authors are aware of work carried out by other groups (e.g. PMF applications in the AMS community by Ulbrich et al., 2008, and Lanz et al., 2007). Furthermore, PMF developments should be concisely re- and overviewed in the context of other receptor models (e.g. COPREM by Wahlin, 2002, and the most recent developments by Lingwall et al., 2008).

Answer: As a reviewer pointed out, we added these references.

Referee: p. 20430, line 5: However, chemical profiles are needed to verify the factor analytical results.

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Answer: We changed this sentence to “Factor analysis is a different but highly effective tool that can be used to apportion sources without directly interacting with the chemical source profiles.

Referee: p. 20435: What was assumed for the height (of the polluted planetary boundary layer) of the geophysical grid cell?

Answer: In this study, we used half of the mixing heights for the trajectory starting heights in Seoul, Korea during the study period. Kim et al. (2007) reported that the mixing heights were approximately less than 300 m under stable conditions, and about 2-3 km of the mixing heights were observed under well-mixed atmospheric conditions. The spatial variation of the mixing heights calculated using HYSPLIT model 4.8 versions were about 200 m \sim 2 km at the sampling site. Therefore, we used the back-trajectories generated at four different starting heights (100, 500, 1000, and 1500 m) for every atmospheric condition.

Referee: p. 20437, line 6: How were these standard deviations calculated precisely? In the corresponding Fig. 3, please plot these std dev. with a color and both, the upper and low confidence limit.

Answer: Fig. 3 showed the resolved factor chemical profiles for PM_{2.5} and the error bars representing the computed error estimates.

We changed the following sentence “The PMF-deduced source profiles (predicted values \pm standard deviation) and contributions are presented in Figs. 3 and 4, respectively” to “The PMF-deduced source profiles (predicted values and error bars) and source contributions are presented in Figs. 3 and 4, respectively.”

Referee: p. 20437, line 13: add OH/radiation

Answer: This was added.

Referee: p. 20437, line 13: nighttime chemistry via NO₃(g)

Answer: This was added.

Referee: p. 20438, line 18-22: the same applies for diesel emissions. The “diesel emission-factor” however does not show such a behavior.

Answer: This can be explained by large road and building constructions and numerous diesel vehicles operating within 500 m of the sampling site during the beginning of this study through the fall of 2003 and during the period of 2006. This increased activity resulted in higher contributions of diesel emission factor during spring and fall than during winter.

Referee: Supplementary material, Fig. S1 and Fig. S4: Please indicate the average absolute mass concentration for both plots. “Residue” shown in Fig. S4 is 14.4%. After the PMF analysis and reconstruction

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of the data, the “Residue” diminished to only 3.1%. Please explain and/or rewrite the Figure captions and re-label. Was the $PM_{2.5}$ residue (probably water and mineral dust) also included as a species in the data matrix ($x\{i, j\}$)? Does the particle mass shown in Fig. 2 include the residue? It further might be confusing that there is more calculated “Secondary nitrate” (20%) (Fig. S1) than measured nitrate (Fig. S4). I suggest to re-label the factors.

Answer: Table 4 showed the average absolute factor contributions resolved by PMF in this study. As a reviewer pointed out, we changed Fig. S1 to Table 4 in supplementary material and re-labeled the factors.

Linguistic/technical corrections:

Referee: p. 20437, line 20: “slightly higher”, rewrite

Answer: This was changed.

Referee: p. 20437, line 27-29: “Two types of motor vehicles... were separated at the sampling site”, rewrite

Answer: This was changed.

Referee: p. 20434, line 6: possibly “wind directions”

Answer: This was changed.

Referee: p. 20447: “Malinowsk” or “Malinowski”

Answer: This was changed.

Referee: p. 20455, Fig. 5: the lower confidence level is not plotted.

Answer: We plotted the lower confidence level in Fig. 5.

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Anonymous referee #2

Referee: Detailed PM_{2.5} speciation data description needed to be included, such as time series plot, it will be interesting to see the comparison of PM_{2.5} species composition contribution pie chart versus figure S1, PMF source contribution pie chart.

Answer: As a reviewer pointed out, we incorporated the detailed PM_{2.5} speciation data description into the text by adding new subsection, “3.1. PM_{2.5} mass concentrations and chemical compositions”

Referee: In the PMF results description, the ratios between key species, such as NO₃/NH₄ ratio in nitrate

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factor, the SO₄/NH₄ ratio in sulfate factor, and by comparing that to the source profiles that are well established, the author can demonstrate the quality of their calculation.

Answer: As described above (reply to referee #1), we added the ratios between key species in nitrate and sulfate factors resolved by PMF.

Referee: Can the author provide the correlation among the gasoline vehicle, diesel vehicle, and road salt and 2 stroke-engine? The correlation among them seems very low from Fig. 4, which is surprising. It is probably beyond the factor analysis capability to resolve such highly correlated source. What are the authors thoughts on this?

Answer: Pearson correlation coefficients among the gasoline vehicle, diesel vehicle, and road salt and 2 stroke-engine were 0.10 for gasoline vehicle vs. diesel vehicle, 0.32 for gasoline vehicle vs. road salt and 2 stroke-engine, and 0.24 for diesel vehicle vs. road salt and 2 stroke-engine. In this study, diesel vehicle was additionally contributed by off-road diesel engines in road and building constructions near the sampling site and the contribution of road salt and 2 stroke-engine highly increased in winter so that correlation among the different types of motor vehicles would be low.

Referee: Usually a number of source categories in the emission inventory are reflected in one source resolved from PMF. For example, there are EC and trace metals in the secondary sulfate sources. Can the author make an estimate the pure source contribution of one single source (For example, the pure sulfate and ammonium contribution for the resolved secondary sources)?

Answer: In PMF, resolved source profiles have more processed characteristic result in part from atmospheric mixing and condensation of oxidized compounds, so that the resolved PMF factors do not represent a single pure source. Without additional "marker" species in the measurements such as molecular organic carbon and triple-isotope (¹⁶O, ¹⁷O, ¹⁸O) in aerosol sulfate and nitrate, it is difficult to separate primary and secondary aerosol contributions from the resolved secondary sources.

Referee: If the regulators need to make an effective control strategy for PM based on these results, they need the confidence of the resolved sources. Can the author provide the rank of confidence the resolved sources, not only based on the modeled output uncertainties, but based on the source composition and the variation of the source contributions?

Answer: As a reviewer pointed out, we added the rank of confidence for the resolved sources to conclusion.

Anonymous Referee #3

Referee: I agree with the authors that if reliable source profiles (preferably local ones) are not available, it

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is better to use statistical models like PMF model. Still, when identifying factors the authors used literature results from abroad. Since emission profiles might be different in Seoul as pointed out by the authors, the authors should pay extra attention on the identification of the contributors. For example, based on Ministry of Environment (MOE), Korea, during that sampling period, the majority of the bus fleet in Seoul has changed fuel from diesel to natural gas and control equipments such as DPF and DOC have been installed to a large fraction of heavy duty diesel vehicles. Also MOE has set a new emission standard for vehicles effective on 2006 (Homepage of MOE, Korea). Thus, it is highly probable the emission composition of diesel vehicles in Seoul be different from the literature results cited in the manuscript.

Answer: As a reviewer pointed out, we identified the factor profiles resolved by PMF after taking into account to the emission change of diesel vehicles. In Figure 3, the temporal trend of alteration of diesel vehicles by pollution control facilities and the ratio of increasing CNG buses to the total running buses in Seoul between 2003 and 2006 were shown. In 2006, the ratio of the installation of emission reduction devices to total registered diesel vehicles in Seoul was approximately 3%, and the change of total registered running diesel buses to CNG buses in Seoul were about 3,456 – approximately 38%. There was also a huge volume of traffic including diesel buses and trucks which were not equipped with pollution control devices from the outside to the inside of Seoul. Therefore, in this study period, the emission change of diesel vehicles couldn't be a significant influence on the results of the PMF resolved factor profile.

Referee: Related to the problem of the fuel change and control equipment installation on the heavy duty diesel vehicles, is the PMF results still valid with changing emission profiles?

Answer: As mentioned above, we have carefully validated the estimated factor profiles and time series through the emission change of diesel vehicles and the collocated trace gas measurements. We think that diesel vehicle factor might be insignificantly influenced by the emission changes which were not so big in the period of 2003-2006. The chemical components and organic molecular markers in $PM_{2.5}$ have been continuously monitored over the period 2007-2008 to examine more accurate effect of emission change for air quality improvement in Seoul. Using these results, we will make further study on the variations of primary combustion sources of $PM_{2.5}$ with emission changes in Seoul.

Referee: As the authors have suggested in section 3.1, it is very hard to separate contributions between gasoline and diesel vehicles. I wonder if there two sources are put into one, whether the performance of the PMF model enhances.

Answer: In this study, we examined different numbers of factors to obtain the most appropriate solution and nine factors gave reasonable results (Please see the response to referee #1). When the numbers of factors decreased, the PMF model results were not enhanced and also gasoline and diesel vehicles were not separated.

Referee: Organic carbons (OCs) can be divided into primary and secondary OC. When the authors apply the PMF model, is this considered? One thing that makes the analysis difficult is that the secondary OC might have two contributors, local and regional. Thus, without separating this factor, the factor analysis results based on foreign literature results might be misleading.

Answer: Up to date, without additional “marker” species in the measurements such as molecular organic carbon, it is difficult to separate primary and secondary OC from the organic carbons, and there is a large uncertainty in the SOC estimates calculated by EC trace method and the empirical method. In PMF, the OC and EC components reflect mixing of primary and secondary pollutants, so that the resolved PMF factors do not represent a single pure source. Therefore, the consideration of secondary OC contributed from local and regional sources are beyond the scope of this paper. As mentioned above, we have been conducting further study for the estimates of SOC and POC, and it will be explored in a future publication.

Referee: The authors applied the backward trajectory analysis for five days with the starting heights of 100, 500, 1000, and 1500 m. I do not think for five day trajectories, the results for the height of 100 or 500 m are not that reliable. I think with these low starting heights a large fraction of the trajectories might touch surface, especially on the Yellow Sea. It might explain why major identified sulfate ‘source’ region is on the Yellow Sea.

Answer: Figure 5 showed the results of cluster analysis for air mass back-trajectories of four different starting heights at the sampling site. As shown in Figure 5, the trajectories of low starting heights did not touch surface. Furthermore, in order to increase the precision of PSCF model results, we had excluded the back-trajectories touching surface in this study.

Referee: Regarding to the backward trajectory, I suggest the trajectory analysis result should be shown. Also, it is interesting that in north of Beijing, there is a major contributing region for ‘secondary nitrate’ but no ‘secondary sulfate’ region is shown. The authors suggested high ammonia concentration might cause it, but that explanation dose not explain the phenomenon north of Beijing.

Answer: The trajectory analysis results for four different starting heights were shown in Figure 5, and we added this to the supporting section. The PSCF plot of secondary nitrate factor showed that the eastern provinces of China had the highest source probability, while the north of Beijing was the moderate potential source region. Up to date, model performances of source-receptor relationship for NH_4NO_3 in East Asia are relatively poor, with lower correlation and with almost all models tending to overestimate NO_3^- and NH_4^+ levels due to larger uncertainties in either emission estimation or complex chemical mechanism. Therefore, it might be possible that the north of Beijing have the moderate source probability.

Referee: In Fig. 7, a large fraction of trajectories have moved over the northeastern region including Beijing which is the heavy coal consumption area with industrial activities. Still, according to the PSCF

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result, this region is not the major contributing region for 'secondary sulfate'. Please explain.

Answer: In this study, according to the comparison between the chemical components of PM_{2.5} and regional air mass transport pattern, the SO₄²⁻ concentrations were highly elevated when air mass originating from the southeastern China/or southwestern marine passed through the Korean peninsula (data not shown). Sun et al. (2006) reported that the concentrations of secondary species (NH₄⁺, SO₄²⁻, and NO₃⁻) in ambient fine particles were much higher when the air masses originated from provinces located south of Beijing than any other direction in haze-fog episodes in Beijing. Also, from the study on "Sulfur Dioxide Emission Trading in China (Wang et al., 2003)", the identified key geographic areas affected by acid rain whose major source might be SO₂ emission were similar to the major contributing region for "secondary sulfate."

Referee: Regarding to the PSCF result, there are several 3-D chemical transport modeling result in the region some of them were dealing with the transport pattern in Korean peninsula. Also, there might be several existing receptor modeling results for Seoul. The authors should give a literature survey result in the manuscript.

Answer: In this study, we carefully identified the potential source regions deduced from the PSCF model by comparing with literature results, and also references of several receptor modeling results were added in the manuscript.

Referee: I could not find a description on where and how the authors have measured the meteorological parameters. I presume they measure these at the aerosol sampling site. I am curious about how representative the wind measurement data were for Seoul proper and surrounding area.

Answer: We added "Meteorological data including temperature, relative humidity, wind speed, and wind direction were recorded every 5 min using a meteorological tower (Davis Instrument) at the sampling site" to the manuscript.

In this study, we used the conditional probability functions (CPF) to assess the impacts of local point sources from various wind directions to the receptor site so that the surface wind direction data measured on sampling site were needed. The wind measurement data collected at the sampling site were not representative for Seoul area, but for receptor site.

Referee: Related to the wind data, I think the authors have tried too much to get the result from the CPF modeling. For example, they interpreted the southeasterly air flow have transported aged sea salt from the Yellow Sea though the river. However, from Fig. 1, the Yellow Sea is on the west of Seoul and the authors stated based on the trajectory analysis that the winds were from west (both southwest and northwest). Thus, without detailed background information, it is hard to envision the relationship between

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the local wind be different from the major wind direction.

Answer: CPF modeling is a powerful approach for identifying the location and the directionality of local point sources whose emissions significantly impact the receptor site. The interpretation of CPF result might be typically more ambiguous for urban-scale mobile/area sources and regional sources. In regional cases, because day-of-arrival surface winds could not clearly capture the source locations governing long-range transport, the PSCF model was applied to identify the more distant regional source influences in this study. Additionally, if there was ambiguity about the interpretation of CPF modeling for local sources such as aged sea salt, we applied the function of wind speeds to verify the effects of local sources (see Fig. 2).

On some specific points:

Referee: I think the manuscript be more easily readable if English be checked by a native speaker.

Answer: This manuscript had been checked by native speakers.

Referee: The authors use 'source' but it might be misleading. For example, there is no 'secondary nitrate source'. I suggest the authors use either 'factor' or 'contributor'.

Answer: As a reviewer pointed out, we changed "source" to "factor."

Referee: Also, it might be misleading to the terms like 'secondary sulfate' and 'secondary nitrate'. These terms imply that there are primary sulfate and nitrate sources.

Answer: We changed "secondary sulfate" and "secondary nitrate" to "sulfate factor" and "nitrate factor", respectively.

Referee: In abstract, section 2.1, and Conclusion, the authors report the sampling started on March 2003, but in the first line of Chapter 3, it was on May 2003. Please correct it.

Answer: We changed "May 2003" to "March 2003."

Referee: In the last paragraph of Chapter 3, the figure number regarding to soil is not Fig. 10 but Fig. 11.

Answer: This was changed.

References

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haze-fog episodes in Beijing. Environmental Science and Technology 40, 3148-2155.

Table 1. Estimated EC-to-OC ratio in some important source profiles resolved by the previous receptor model studies

Resolved source	PMF ¹⁾	CMB-MM ²⁾	CMB ³⁾	This study
Diesel emission	1	1.33	1.24	1.4
Gasoline vehicle	0.5	0.025	0.45	0.4
Biomass burning	0.5	0.12	0.06-0.25	0.02
Soil	0.07	0.06	-	0.02

1) Liu et al., 2006 (study location: southeastern U.S.)

2) Zheng et al., 2002 (study location: southeastern U.S.)

3) Zheng et al., 2006 (study location: Hong Kong)

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Table 2. OC and EC fractions of diesel and gasoline engine particulate matter exhaust

Engine type	OC	EC	EC/OC
Heavy-duty diesel ^a	19±8	75±10	2.4-7.7
Heavy-duty diesel (SPECIATE) ^b	21-36	52-54	1.4-2.7
Light-duty diesel ^c	30±9	61±16	1.1-3.7
Light-duty diesel (SPECIATE) ^b	22-43	51-64	1.2-2.9
Gasoline (hot stabilized) ^a	56±11	25±15	0.2-0.9
Gasoline ("smoker" and "high emitter") ^{a,c}	76±10	7±6	0.01-0.2
Gasoline (cold start) ^a	46±14	42±14	0.5-1.7

a) Fujita et al. (1998) and Watson et al. (1998)

b) SPECIATE database (U.S. Environmental Protection Agency, 1999)

c) Norbeck et al. (1998)

Source: U.S. Environmental Protection Agency (2002).

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Table 3. Pearson correlation coefficients between source contributions and meteorological data and gas concentrations

Factor	Temp	RH	WS	SO ₂	NO ₂	HNO ₃	CO
Diesel emission	0.04	0.00	-0.21	0.35	0.63	0.28	0.51
Secondary nitrate	-0.05	0.05	-0.11	0.38	0.45	0.15	0.36
Industry	0.10	0.09	-0.08	0.29	0.36	0.15	0.23
Gasoline vehicle	-0.06	-0.06	-0.46	0.09	0.54	-0.10	0.60
Secondary sulfate	0.22	0.23	-0.07	0.30	0.23	0.50	0.21
Road salt/2-stroke engine	-0.57	-0.17	-0.05	0.35	0.36	-0.03	0.40
Biomass burning	-0.25	-0.06	-0.06	0.45	0.36	0.15	0.46
Soil	-0.17	-0.23	0.32	0.16	0.05	0.06	0.04
Aged sea salt	-0.29	-0.19	0.37	0.25	-0.01	0.02	0.07

Note that NO₂ and CO were collected in the national ambient air monitoring site in Seoul, distance 900 m from our sampling site, but SO₂ and HNO₃ were directly measured in our sampling site using annular denuder systems.

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Table 4. Average factor contributions of PM_{2.5} mass concentration at the sampling site

	Average factor contribution (standard error mean)	
	Mass contribution ($\mu\text{g}/\text{m}^3$)	% Contribution
Secondary nitrate	9.37 (0.41)	20.9 (0.6)
Secondary sulfate	8.80 (0.40)	20.5 (0.5)
Gasoline vehicle	5.68 (0.17)	17.2 (0.6)
Biomass burning	5.12 (0.21)	12.1 (0.3)
Diesel emission	3.18 (0.15)	8.1 (0.3)
Soil	3.10 (0.23)	7.4 (0.5)
Industry	2.77 (0.13)	6.7 (0.3)
Road salt/2-stroke engine	2.29 (0.16)	5.1 (0.3)
Aged sea salt	0.78 (0.03)	2.2 (0.1)

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