

We would like to thank the reviewer for their comments that helped to improve the manuscript. Hereafter, we have responded to the various comments. Some of the comments were directly answered within the text of the manuscript and the modified text is reproduced below in quotation marks.

Reviewer comments:

Reviewer #1: The manuscript presents a source apportionment study in the city of Montreal, Canada and looks at associated health risks. Daily filter samples were used during a 3-month period and analysed for a comprehensive chemical composition, including a number of organic molecular markers to better identify sources. Further the study utilises a chemical transport model to identify source regions and evaluates the health risks of measured components.

In my opinion the manuscript represents a good contribution to existing literature and the topic is relevant. The scientific quality is sound, and the analysis has been performed and presented with care. The structure of the manuscript, the results and presentation are clear. Thus, I believe, the manuscript is worth publication in ACP/EGUsphere, however, I do have some comments below:

Major comments:

- 1. The study took place during the Covid Pandemic in 2020 but there is no mention of what impact this may have had on the outcome of the study. Even though, from what I can find, Montreal was not in a lockdown during that period, activities will have altered and thus might have influenced local and transboundary pollution. I think this point needs to be addressed.**

Answer: We agree with the reviewer on the importance of mentioning that this study took place during the COVID-19 pandemic in 2020. During our sampling period, Montréal was in partial lockdown where public spaces (e.g., bars, gyms, cinemas, museums, libraries and casinos) were closed due to the possibility of a second wave of the COVID-19

pandemic. We have mentioned in **lines 300-302** in the manuscript that the concentrations of PM_{2.5} in 2020 were not too different in comparison with the previous years (2018 and 2019) for data for the same dates of the year (13 August to 11 November) and locations. Thus, the characteristics of the sources of PM_{2.5} identified in this study are likely to be similar to other years. The comment of the referee was taken into consideration and the following paragraph was added in the text:

“It is important to mention that during our sampling period, Montréal was in partial lockdown where public spaces (e.g., bars, gyms, cinemas, museums, libraries and casinos) were closed due to the possibility of a second wave of the COVID-19 pandemic. Primary and some secondary schools were opened during that period. While these considerations suggest that the results presented here are also applicable to pre-and post-pandemic conditions, further studies are needed before generalizing the results of this study other periods.”

2. Summary needs to be clearer in what species have been used for source apportionment. The sentence starting “this source apportionment study, which examined...” (line 15) sounds like the large suite of organic markers are the chemicals used for source apportionment. This section needs reworking to be clearer.

Answer: The comment of the reviewer was taken into consideration and the paragraph was updated in the text:

“This source apportionment study, which examined the main contributing sources to PM_{2.5} using a larger suite of organic molecular markers than other Canadian studies, is the first of its sort in Canada. A focus of this study was on quantifying previously unresolved sources of PM_{2.5} through the inclusion in the PMF analysis of additional organic molecular markers beyond those measured typically by the Canadian government’s National Air Pollution Surveillance Program (NAPS). The organic species included in the PMF model were comprised of six n-alkanes, two fatty acids, one dicarboxylic acid, two biogenic secondary organic aerosols (SOA) tracers and hopane.”

3. Section 2.3 Enrichment factors and respective results: The enrichment factors were calculated with Al as reference element, however, later in the text (line 442, p19) there is mention of Aluminium production. Will this impact the Al concentrations within Montreal and thus is Al an appropriate reference element?

Answer: The enrichment factor (EF) is a qualitative method that is used to differentiate between natural and anthropogenic sources of metals in the samples. EF (Eq. 1) is defined as the ratio of the considered element concentration (C_x) to the reference element concentration (C_{ref}) in $PM_{2.5}$ divided by the same ratio for crustal material retrieved from the upper crust (Mason & Moore, 1982). Typical reference elements used in the literature are Al, Ti, and Fe (Rodriguez-Espinosa et al., 2017; Amil et al., 2016). An EF value close to 1 indicates an element originates from crustal materials while an EF higher than 10 indicates a strong anthropogenic source (Esmailirad et al., 2020).

$$EF = \left(\frac{C_x}{C_{ref}} \right)_{air} / \left(\frac{C_x}{C_{ref}} \right)_{crust} \quad (\text{Eq. 1})$$

We have reported in the manuscript the results using Al as a reference. We have also checked the results using Ti and Fe, and all three reference elements pointed to the same results (Fig. 1 in this authors' comment). We agree with the reviewer that we have mentioned in the manuscript ([lines 442-444](#)) that aluminum production and industrial processes related to metallurgy contribute to air pollution in Québec, although this part of manuscript is focused on sulphur emissions. To clarify, we have added the following sentence to the manuscript:

“Although based on the weak correlation between sulfate and Al and strong correlation of Al with crustal elements, we believe that aluminium production is not an important source of particulate aluminum at our site.”

It is important to mention that in the updated version of the manuscript, we have decided to remove the section concerning the EF based on the recommendation of the second reviewer.

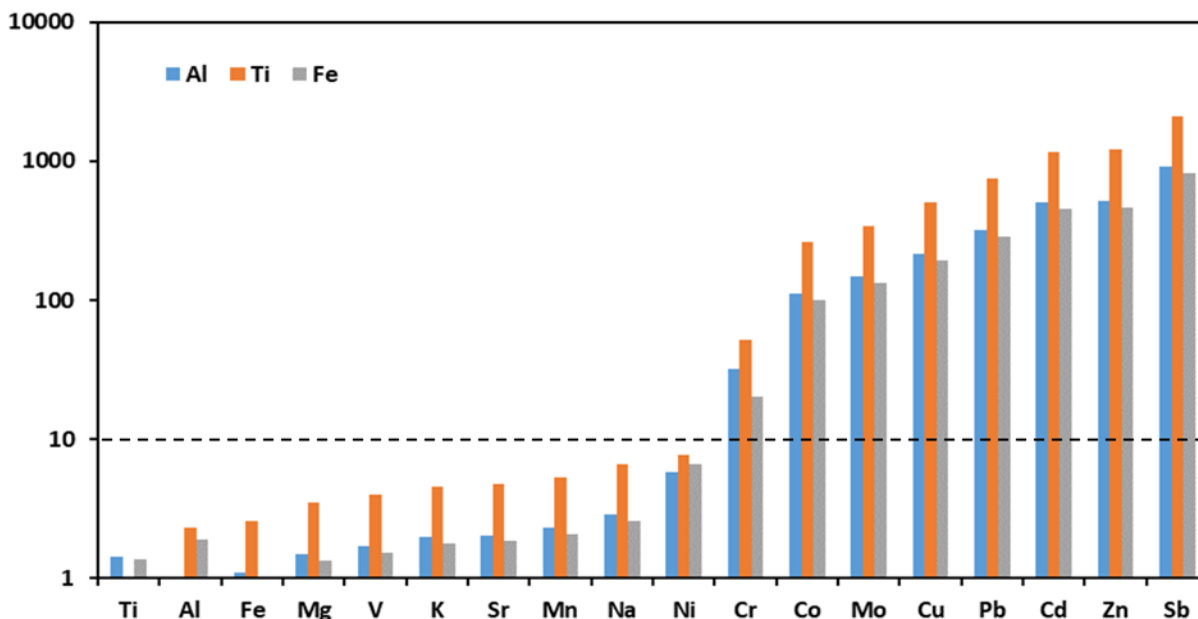


Fig. 1: Enrichment factor of selected elements in PM_{2.5} using Al, Ti and Fe as a reference element. An EF higher than 10 indicates an anthropogenic source.

References

Amil, N., Latif, M.T., Khan, F., Mohamad, M., 2016. Atmos. Chem. Phys. Seasonal variability of PM_{2.5} composition and sources in the Klang Valley urban-industrial environment 5357–5381. <https://doi.org/10.5194/acp-16-5357-2016>

Esmailirad, S., Lai, A., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., Uzu, G., Daellenbach, K., Canonaco, F., Hassankhany, H., Arhami, M., Baltensperger, U., Prévôt, A.S.H., Schauer, J.J., Jaffrezo, J.L., Hosseini, V., El Haddad, I., 2020. Source apportionment of fine particulate matter in a Middle Eastern Metropolis, Tehran-Iran, using PMF with organic and inorganic markers. *Sci. Total Environ.* 705, 135330. <https://doi.org/10.1016/j.scitotenv.2019.135330>

Mason, B., & Moore, C.B. Principles of Geochemistry, fourth edition. Wiley. 1982.

Rodriguez-Espinosa, P.F., Flores-Rangel, R.M., Mugica-Alvarez, V., Morales-Garcia, S.S., 2017. Sources of trace metals in PM₁₀ from a petrochemical industrial complex in Northern Mexico. *Air Qual. Atmos. Heal.* 10, 69–84. <https://doi.org/10.1007/s11869-016-0409-0>

4. Section 2.5 Source apportionment: Given this is a source apportionment study as per title I think there needs to be some more information on the source apportionment: a) Uncertainties are important in PMF analysis and therefore it would be useful to know how uncertainties were calculated. I could not find these calculations in the method section or method reference Fakhri et al. (2023). Can this please be detailed, possible in the supplementary material. b) It would be good to list the species used in PMF and the percentage below LOD, maybe this can be indicated in table 3 or given in the appendix. c) Figure 5 on page 21 should include the concentration of species and % of species in each factor as both information is useful for identifying the factors. Possibly this should also include confidence intervals that should be available through bootstrap. d) in the supplement it would be useful to also display Q/Qexp. e) It would be good to have an idea of the residuals as well.

Answer: The comment of the reviewer was taken into consideration and more information and references were added to the manuscript and the supplementary information. With respect to point (a) in the above comment, in the present work, samples below the detection limit (DL) were replaced by half of the DL and were given an uncertainty of 5/6 times the detection limit (Polissar et al., 1998). Missing samples were replaced by the median value of that species and were given an uncertainty of 4 times the median value (Polissar et al., 1998). When the concentration was greater than the DL, the uncertainty was calculated according to the US EPA guidelines (USEPA, 2014; Liu et al., 2018; Lee et al., 2022; Park et al., 2019): $\sqrt{(Concentration \times 0.1)^2 + (0.5 \times DL)^2}$. After screening the integrity of the input data, 27 species were included in the PMF model (lines 218-219). The overall number of samples (80 samples) and the number of species complies with the ratio of at least 3:1, as proposed by Belis et al., (2019). When the S/N ratio was less than 0.2, the PM species were classified as "bad," "weak" when the S/N ratio was between 0.2 and 2, and "strong" when the S/N ratio was greater than 2 (Esmailirad et al., 2020). The bad species are excluded from the analysis while the uncertainty for the weak species is tripled. PM_{2.5} was designated as a "total variable" and was automatically classified as "weak". All the included species were successfully modeled by PMF with their concentrations reconstructed accurately and were qualified as "strong" except for nitrate which presented a S/N ratio of 0.9 and was defined as "weak".

With respect to point (b), all species included in the PMF analysis were above DL for all samples except for some elements (between 1 and 12% were below the DL). The following table (Table 1) was added in the supplementary information of all the species included in PMF and the percentage below LOD.

Table 1: Species included in PMF.

Species	% of data below the DL	Species	% of data below the DL
OC	-	Levogluconan	-
EC	-	7 α [H]-21 β [H]-Hopane	-
Na ⁺	-	Hexadecanoic acid	-
Cl ⁻	-	Octadecanoic acid	-
NH ₄ ⁺	-	C20	-
NO ₃ ⁻	-	C21	-
SO ₄ ²⁻	-	C24	-
Al	3	C25	-
Fe	1	C27	-
Ti	-	C29	-
Cu	4	Oxalic acid	-
Sb	3	Pinic acid	-
Cd	5	Cis-pinonic acid	-
Co	12		

With respect to point (c), Figure 5 was updated (Fig. 2 below).

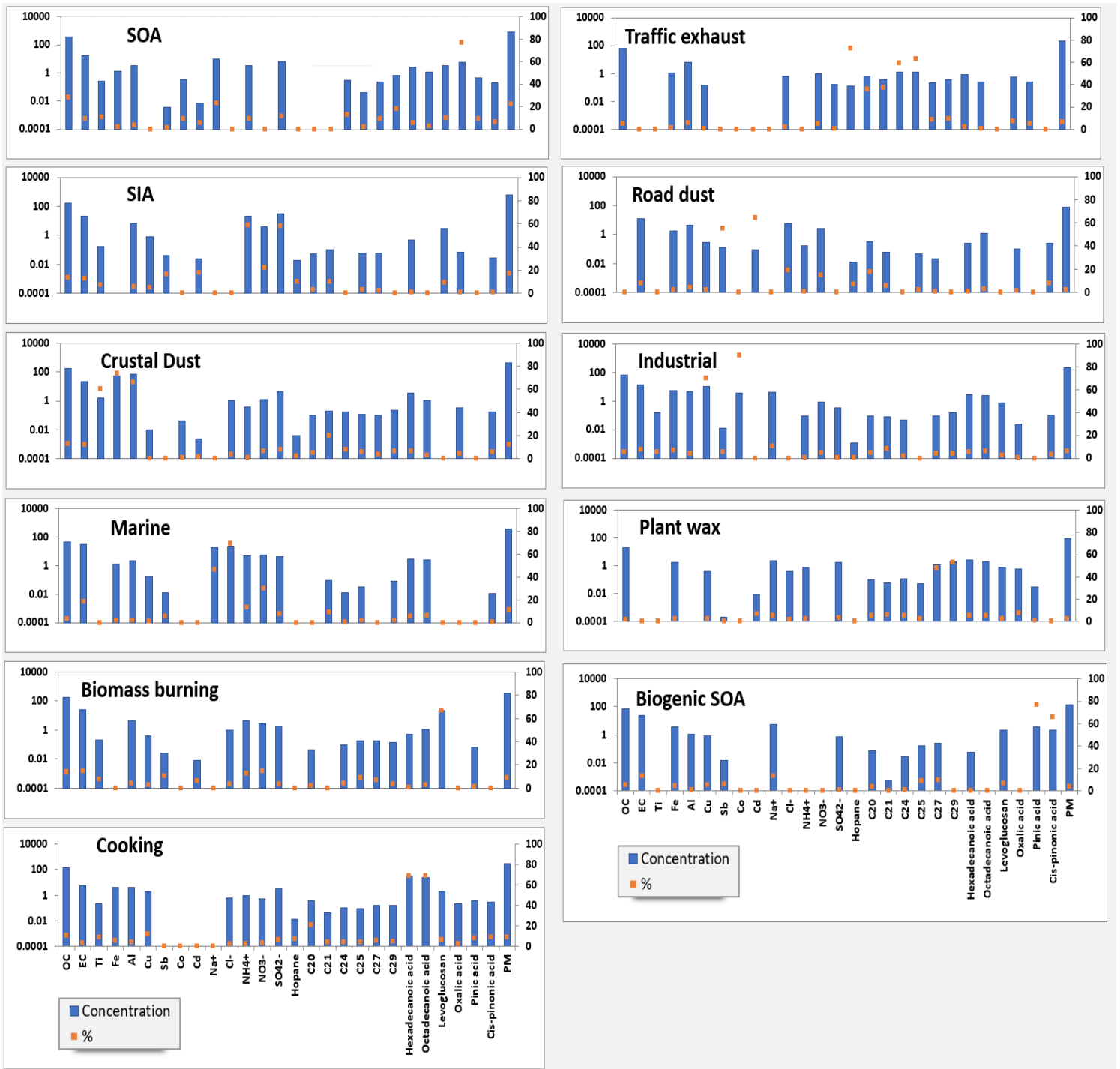


Fig. 2: Profiles of the eleven factors identified from the PMF model. The left axis corresponds to concentration (blue bars) and the right axis percentage (orange markers). Units of concentration are ng/m^3

With respect to points (d) and (e), in the reviewer's comment above, the objective function Q in PMF is considered as a critical parameter. PMF minimizes it when determining factor contributions and profiles. Since the number of factors in PMF is unknown, we started with the minimum number of factors (which is 2) and we started increasing this number. To select the appropriate number of factors, different mathematical diagnostic methods were investigated such as the maximum individual mean (IM) and the maximum individual standard deviation (IS) (as described in the Supplementary Information). Graphical representations of IM and IS statistics along with the Q -value showed generally a constant decrease of their values when increasing the number of factors and a stabilization starting with the 11-factor solution, which suggests that 11 is the optimal number of solutions. However, we have also used our understanding of the probable sources impacting the sampling site and the species characteristics to choose the most suitable number of factors.

To ensure robust results in PMF, several points were taken into consideration:

- The authors ensured that the uncertainty-scaled residuals of all the species are generally normally distributed with residuals varying between -3 and +3.
- The authors checked that all the species are well modeled with high determination coefficients (R^2) between observed and predicted observations.
- The authors examined the Q/Q_{exp} values for the different species and ensured that this value was lower than 2 as recommended in the EPA PMF manual (USEPA, 2014). For each species, the Q/Q_{exp} is the sum of the squares of the scaled residuals for that species divided by the overall Q_{exp} divided by the number of strong species. Thus, examining Q/Q_{exp} is an efficient way to understand the residuals of a PMF solution.
- The authors compared the resulting source profiles against the literature.
- The variation of Q/Q_{exp} ratio from 3 to 14 factors is now provided in the supplementary information as requested by the reviewer.
- Lastly, the robustness of the PMF solution was tested by the two-error estimation method (bootstrap and displacement) as instructed in the PMF manual to ensure the solution was stable (Table S2 in the supplementary information) (USEPA, 2014).

References

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Park, M. Bin, Lee, T.J., Lee, E.S., Kim, D.S., 2019. Enhancing source identification of hourly PM_{2.5} data in Seoul based on a dataset segmentation scheme by positive matrix factorization (PMF). *Atmos. Pollut. Res.* 10, 1042–1059. <https://doi.org/10.1016/j.apr.2019.01.013>

Polissar, A. V., Hopke, P.K., Paatero, P., Malm, W.C., Sisler, J.F., 1998. Atmospheric aerosol over Alaska. 2. Elemental composition and sources. *J. Geophys. Res. Atmos.* 103.

USEPA, 2014. EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide. U.S. Environmental Protection Agency Office of Research and Development Washington, DC 20460.

Minor Comments:

P3L71: “...elements in the PM_{2.5} are investigated...” “the” should be deleted.

Answer: The sentence was corrected in the text.

P12Figure 2 and respective section 3.1: It is not entirely clear if the concentrations given are for the period of 13/Aug-11/Nov for all years and sites or just for the year 2020; if just for the year 2020, it might be useful to only use the same period in previous years also or indicate clearer if this is not the case.

Answer: The caption in Figure 2 was changed to make it clearer. Only data for same dates of the year (13 August to 11 November) were used.

P13Section3.2L340onwards: I think it would be useful to include more information on this in the supplementary, like a figure or what EC/OC min is used and how it was derived, and also a reference of the method used.

Answer: The comment of the reviewer was taken into consideration and more information and references were added to the manuscript and the supplementary information. The time series plot of the OC/EC ratio was also added in the supplementary information (Fig. 3 in this authors' comment). The text added to the supplementary information is quoted below.

“While EC is derived only from combustion processes, organic carbon (OC) is produced by both primary and secondary sources. Several studies have estimated the contribution of secondary organic carbon (SOC) by employing the OC/EC minimum ratio method and the following equation (Castro et al., 1999; Shivani et al., 2019; Cesari et al., 2018; Calvo et al., 2018; Joseph et al., 2012).

$$\text{SOC} = \text{OC}_{\text{total}} - \text{EC} \times \left(\frac{\text{OC}}{\text{EC}} \right)_{\text{min}} \quad (\text{Eq. 2})$$

In the first step, the OC/EC ratio is calculated for each sample, and $(OC/EC)_{\min}$ is the minimum ratio observed in the samples. In this study, $(OC/EC)_{\min}$ was 2.22. In the second step, the measured OC (OC_{total}) and EC for each sample are used with the minimum to calculate the SOC following the equation above. ”

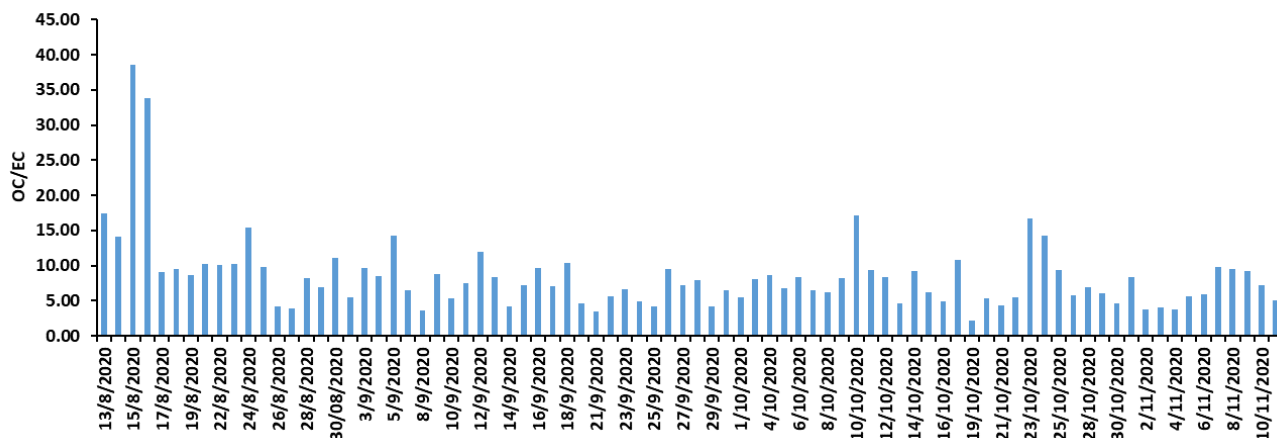


Fig. 3: The temporal variation of OC/EC ratio for the sampling period.

References

Calvo, A.I., Pont, V., Liousse, C., Dupré, B., Mariscal, A., Zouiten, C., Gardrat, E., Castera, P., Lacaux, C.G., Castro, A., Fraile, R., 2008. Chemical composition of urban aerosols in Toulouse, France during CAPITOUL experiment. *Meteorol. Atmos. Phys.* 102, 307–323.

Castro, L.M., Pio, C.A., Harrison, R.M., et al., 1999. Carbonaceous aerosol in urban and rural European atmospheres: estimation of secondary organic carbon concentrations. *Atmos. Env.* 33 (17), 2771-2781.

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Joseph, A.E., Unnikrishnan, S., Kumar, R., 2012. Chemical characterization and mass closure of fine aerosol for different land use patterns in Mumbai city. *Aerosol Air Qual. Res.* 12, 61–72.

Shivani, Gadi, R., Sharma, S.K., Mandal, T.K., 2019. Seasonal variation, source apportionment and source attributed health risk of fine carbonaceous aerosols over National Capital Region, India. *Chemosphere* 237, 124500. <https://doi.org/10.1016/j.chemosphere.2019.124500>

P17L397-398: Sentence: “No correlation was found between Cu...” - is this finding confirmed by the source apportionment or is it that Cu has a more dominant source but still has a brake wear component?

Answer: The correlation between trace metals provides qualitative information on the sources of the measured elements. In this study, we have looked at the Pearson correlation coefficient (R) between Cu and the elements Cd and Sb. No correlation was found between Cu and the elements Cd and Sb ($R < 0.01$, $p < 0.05$); indicating that brake wear debris was not an important source of Cu in Montréal for our study. Moreover, we have included the elements Cu, Sb and Cd in the source apportionment analysis. As the reviewer likely knows, PMF accounts for the possibility of multiple sources. In this study, 70% of Cu was attributed to the industrial emission factor. The road dust factor was characterized by high loadings of Cd (69%) and Sb (58%). These two elements are linked to non-exhaust vehicle emissions, particularly from brake-wear debris (Thorpe and Harrison, 2008; Lin et al., 2015). The road dust factor only contained 2% of the Cu. Thus, the PMF results confirm the conclusion from the correlation analysis that brake wear debris was not an important source of Cu in Montréal for our study.

References

Thorpe, A., Harrison, R.M. Sources and properties of non-exhaust particulate matter from road traffic: A review. *Sci. Total Environ.* 400, 270–282. <https://doi.org/10.1016/j.scitotenv.2008.06.007>, 2008.

Lin, Y.C., Tsai, C.J., Wu, Y.C., Zhang, R., Chi, K.H., Huang, Y.T., Lin, S.H., Hsu, S.C. Characteristics of trace metals in traffic-derived particles in Hsuehshan Tunnel, Taiwan: Size distribution, potential source, and fingerprinting metal ratio. *Atmos. Chem. Phys.* 15, 4117–4130. <https://doi.org/10.5194/acp-15-4117-2015>, 2015.

P17L406-407: Sentence: “Lastly, no corelation was found between...” - this needs a reference for Zn, Pb and Sb, Cl as incinerator traces.

Answer: The comment of the reviewer was taken into consideration and two references were added to the text. The references are provided below.

References:

Riffault, V., Arndt, J., Marris, H., Mbengue, S., Setyan, A., Alleman, L.Y., Deboudt, K., Flament, P., Augustin, P., Delbarre, H., Wenger, J. 2015. Fine and ultrafine particles in the vicinity of industrial activities: A review. *Crit. Rev. Environ. Sci. Technol.* 45, 2305–2356. <https://doi.org/10.1080/10643389.2015.1025636>.

Rahn, K.A., Huang, S., 1999. A graphical technique for distinguishing soil and atmospheric deposition in biomonitors from the plant material. *Sci. Total Environ.* 232, 79–104. [https://doi.org/10.1016/S0048-9697\(99\)00112-6](https://doi.org/10.1016/S0048-9697(99)00112-6)

P18section3.5 This references the mass closure results. I think the mass closure should be mentioned in the text or even the methodology.

Answer: The detailed mass closure methodology was moved from the supplementary information to the main text of the manuscript. The text moved to the main text is quoted below.

“The term "chemical mass closure" refers to the reconstruction of the measured weighed mass using just the chemical composition. It is done by comparing the combined masses of the chemical species to the gravimetric particulate matter mass (m_{grav}), wherein the reconstructed PM_{2.5} mass (m_{chem}) is defined as the sum of organic matter (OM), EC, crustal matter, sea salt, secondary inorganic aerosol (SIA), and other elements that are not taken into account as minerals (Chow et al., 2015).

A chemical mass closure study was performed using the chemical composition measurements to estimate the contributions of the different components to the total PM_{2.5} mass concentration following the method reported by Fakhri et al. (2023). Briefly, the contribution of sea salt is calculated by summing the six major ions (Sciare et al., 2005):

$$[\text{Sea salt}] = [\text{Na}^+] + [\text{Cl}^-] + [\text{ss} - \text{Mg}^{2+}] + [\text{ss} - \text{K}^+] + [\text{ss} - \text{Ca}^{2+}] + [\text{ss} - \text{SO}_4^{2-}] \quad (\text{Eq. 1})$$

Ionic constituents such as K^+ , Ca^{2+} , Mg^{2+} and SO_4^{2-} are derived from both marine and non-marine sources. Therefore, it is necessary to discriminate sea salt (ss) from non-sea salt (nss) contributions. Assuming that all sodium ions are of marine origin, the sea salt contribution can be calculated based on sea water composition as shown in Eqs. 2 - 5

(Genga et al., 2017; Sciare et al., 2005). Furthermore, non-sea salt potassium, calcium, magnesium and sulfate (nss-K⁺, nss-Ca²⁺, nss-Mg²⁺ and nss-SO₄²⁻) are calculated by subtracting the sea-salt fraction (ss-K⁺, ss-Ca²⁺, ss-Mg²⁺ and ss-SO₄²⁻, respectively) from the total concentration of the ions (K⁺, Ca²⁺, Mg²⁺ and SO₄²⁻, respectively).

$$[ss - SO_4^{2-}] = 0.252 \times [Na^+] \quad (\text{Eq. 2})$$

$$[ss - Ca^{2+}] = 0.038 \times [Na^+] \quad (\text{Eq. 3})$$

$$[ss - K^+] = 0.036 \times [Na^+] \quad (\text{Eq. 4})$$

$$[ss - Mg^{2+}] = 0.119 \times [Na^+] \quad (\text{Eq. 5})$$

In addition, secondary inorganic aerosol (SIA) is represented by the sum of nss-SO₄²⁻, NH₄⁺ and NO₃⁻. To take bound water into account a hydration multiplication factor of 1.29 was applied to convert the dry inorganic concentrations (SIA and sea salt) into hydrated species (Sciare et al., 2005; Genga et al., 2017).

The contribution of crustal matter (CM) (Eq. 6) was estimated by summing the concentrations of aluminum, silicon, calcium, iron, and titanium in their oxide forms (Huang et al., 2014). The coefficients in front of the elements correspond to the additional mass due to oxygen in the minerals. Silicon was not measured in this study and was indirectly determined by multiplying the measured aluminum concentration by a factor of 3.41 (Esmailirad et al., 2020). This factor is obtained from the ratio of Si and Al in the Earth's crust following Mason and Moore (1982).

$$[CM] = 2.2 [Al] + 2.49 [Si] + 1.63 [Ca] + 2.42 [Fe] + 1.94 [Ti] \quad (\text{Eq. 6})$$

To find the optimal CF to calculate OM from OC, the factor was varied from 1.2 to 2.1. The Pearson correlation (R) calculated between the reconstructed PM_{2.5} and the measured mass did not change significantly (0.978-0.979), but the highest correlation and the slope closest to 1 was obtained with CF=1.6. The results of chemical mass closure study are shown in Fig. S5."

References

Chow, J.C., Lowenthal, D.H., Chen, L.W.A., Wang, X., Watson, J.G., 2015. Mass reconstruction methods for PM_{2.5}: a review. *Air Qual. Atmos. Heal.* 8, 243–263. <https://doi.org/10.1007/s11869-015-0338-3>

Huang, X.H.H., Bian, Q., Ng, W.M., Louie, P.K.K., Yu, J.Z., 2014. Characterization of PM_{2.5} major components and source investigation in suburban Hong Kong: A one year monitoring study. *Aerosol Air Qual. Res.* 14, 237–250.

Fakhri, N., Fadel, M., Öztürk, F., Keleş, M., Iakovides, M., Pikridas, M., Abdallah, C., Karam, C., Sciare, J., Hayes, P.L., Afif, C., 2023. Comprehensive chemical characterization of PM_{2.5} in the large East Mediterranean-Middle East city of Beirut, Lebanon. *J. Environ. Sci.* 133, 118–137. <https://doi.org/10.1016/j.jes.2022.07.010>

Genga, A., Ielpo, P., Siciliano, T., Siciliano, M., 2017. Carbonaceous particles and aerosol mass closure in PM_{2.5} collected in a port city. *Atmos. Res.* 183, 245–254.

Mason, B., Moore, C.B., 1982. *Principles of Geochemistry*, fourth edition. Wiley.

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P20L463 onwards: the traffic exhaust factor still has some Al in it and Fe, thus Might there still be some mixing with road dust/crustal dust? Especially as the road dust has less aluminium than the traffic exhaust – see also comment about PMF in general as the factor profiles in the figure would benefit from more information.

Answer: We agree with the reviewer that there may be some very small mixing of the traffic exhaust, road dust and crustal dust factors, which is a limitation of this study, but the amount of mixing is very minor and should not impact the conclusions drawn from these results. In this study, PMF allocated 76% of Fe and 68% of Al to the crustal dust factor. In comparison, if we look closely at the results (Fig. 4 below), only 2% of Fe was

in the road dust factor while the amount of Al was 4%. Moreover, Fe was 2% in the traffic exhaust factor and Al was 6%. These are therefore very small amounts which may indicate a slight mixing of the factors. However, it is also possible that these metals are actually associated with the identified sources. Previous literature has found Fe and Al-containing particles in vehicle exhaust (Golokhvast et al., 2015; Wang et al., 2021). It is also logical that road dust would contain some crustal elements.

References

- Golokhvast, K.S., Chernyshev, V. V., Chaika, V. V., Ugay, S.M., Zelinskaya, E. V., Tsatsakis, A.M., Karakitsios, S.P., Sarigiannis, D.A., 2015. Size-segregated emissions and metal content of vehicle-emitted particles as a function of mileage: Implications to population exposure. *Environ. Res.* 142, 479–485. <https://doi.org/10.1016/j.envres.2015.07.018>
- Wang, J.M., Jeong, C-H., Hilker, N., Healy, R.M., Sofowote, U., Debosz, J., Su, Y., Munoz, A., Evans. G.J., 2021. Quantifying metal emissions from vehicular traffic using real world emission factors. *Environ. Poll.* 268, 115805. <https://doi.org/10.1016/j.envpol.2020.115805>

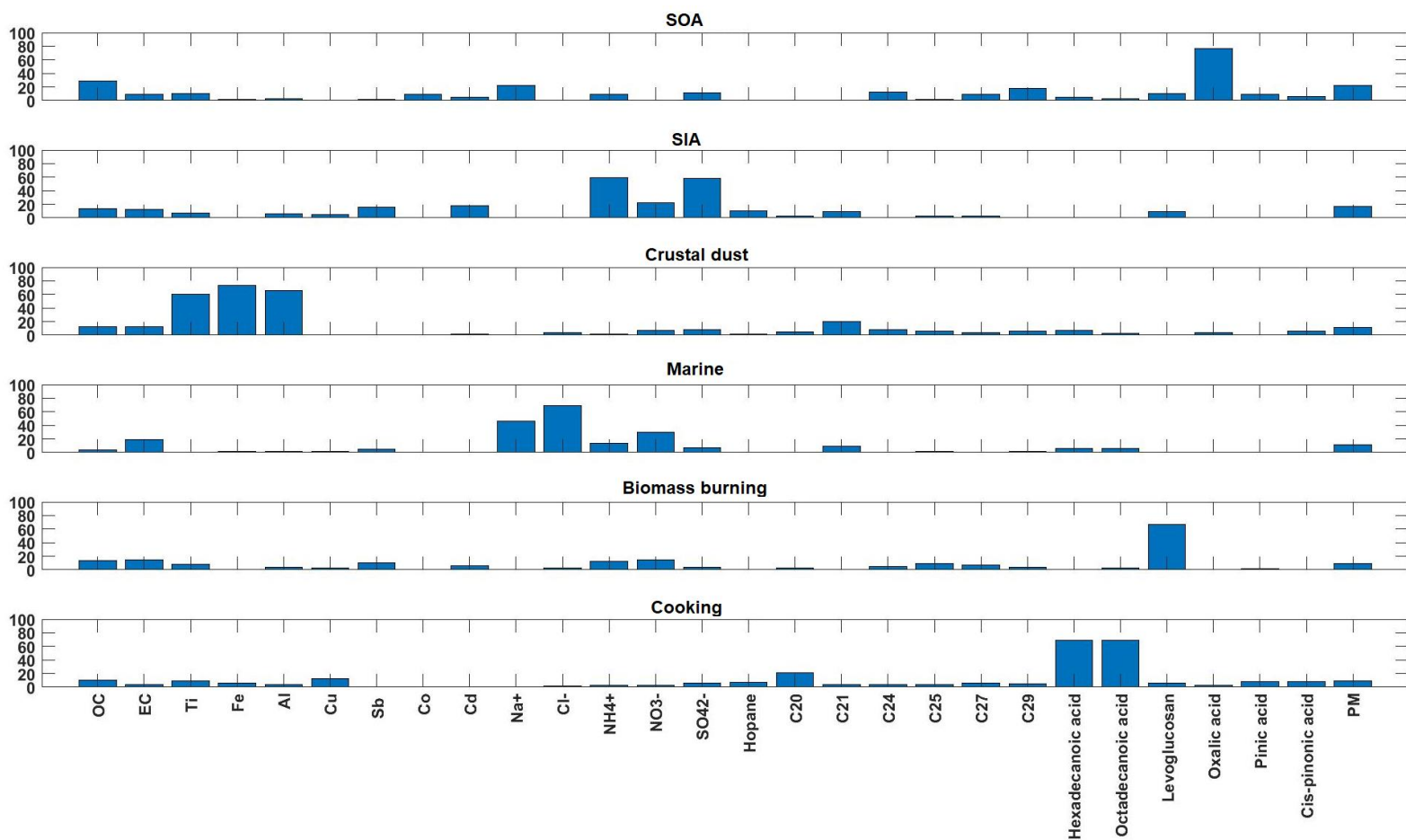


Fig. 4 (Part 1): Profiles of the factors identified from the PMF model. Loading (in percentage) is indicated on the vertical axes.

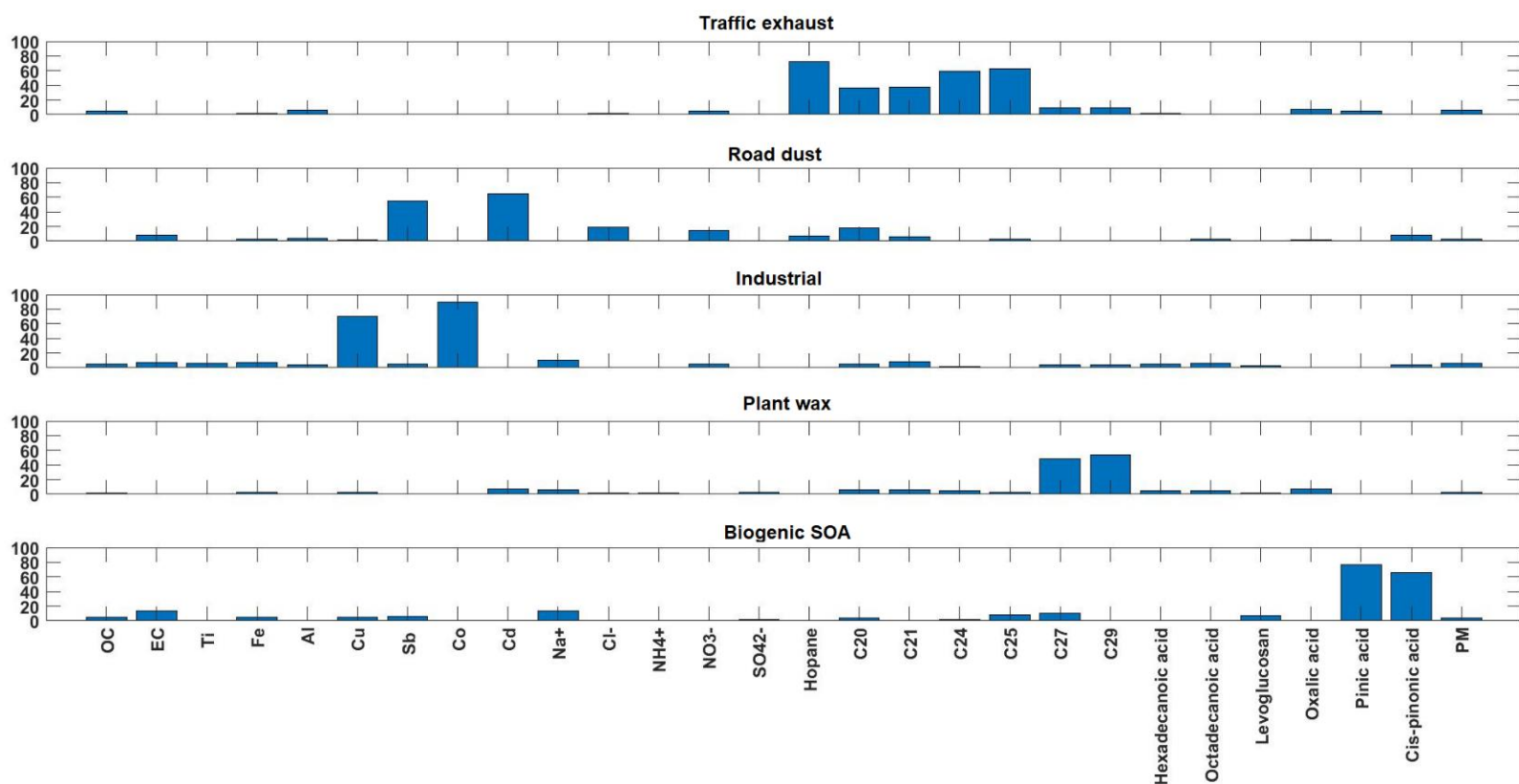


Fig. 4 (Part 2): Profiles of the factors identified from the PMF model. Loading (in percentage) is indicated on the vertical axes.

P20L481 onwards: Similar to the previous comment, I wonder who there is some Cu, Sb, Fe in the biogenic SOA – is there still some mixing? I guess from the supplementary material it sounds like a higher solution split the factors too far, so maybe just a comment or a reference that may have experienced the same issues would be useful.

Answer: We agree with the reviewer that when looking at the biogenic SOA, we notice that there is some Cu, Sb and Fe (below 10%). If we look closely at this factor, the biogenic SOA factor was identified based on high loading of pinic acid (75%) and pinonic acid (66%). On the other hand, Fe was only 5%, Sb was 6% and Cu was 5%. Fe was allocated in much higher proportions to the crustal dust factor, Sb to the road dust factor and Cu to

the industrial factor. Our PMF analysis is consistent with a study reported by Fadel et al. (2023). Fadel and coworkers have also included biogenic SOA tracers in the PMF analysis and in their biogenic SOA profile (Fig. 5) one also notices small amounts of metals/elements.

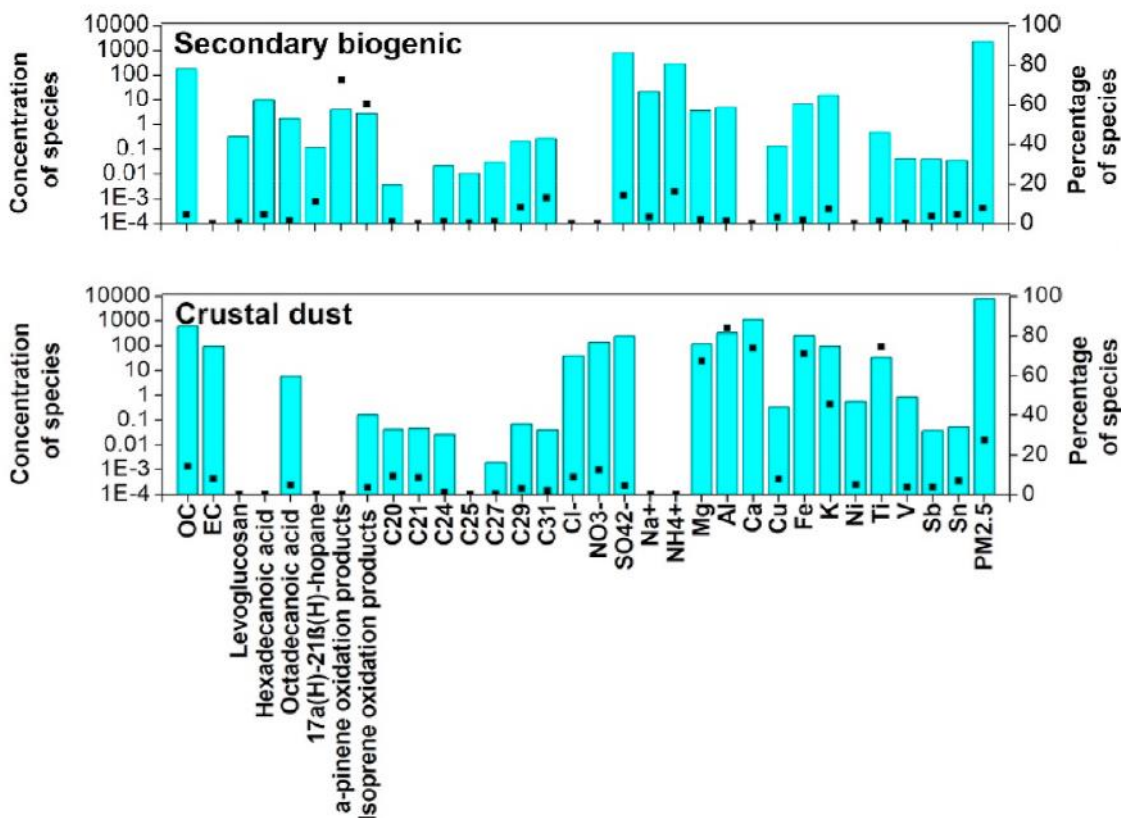


Fig. 5: PM_{2.5} profiles calculated via PMF in Fadel et al. (2023)

Reference:

Fadel, M., Courcot, D., Seigneur, M., Kfoury, A., Oikonomou, K., Sciare, J., Afif, C., 2023. Identification and apportionment of local and long-range sources of PM_{2.5} in two East-Mediterranean sites. Atmos. Pollut. Res. 14, 101622. <https://doi.org/10.1016/j.apr.2022.101622>