

We would like to thank the editor for their comments that helped to improve the manuscript. Hereafter, we have responded to the various comments. The comments were directly answered within the text of the manuscript (in track changes mode and highlighted in yellow) and the modified text is reproduced below in quotation marks.

Minor Points

- 1) It would be beneficial to add a short discussion about the advantages and disadvantages of increasing the number of tracers both from a measurement and information standpoint. Something along the lines of an abbreviated version of the discussion on page 21 and 22 of the combined response document (response to the first point of referee 2). The conclusions could be a good section for this.**

Answer: The comment was taken into consideration and the conclusion was updated in the manuscript (line 687-704):

"This work examines PM_{2.5} sources in Montréal using detailed chemical speciation data collected over a 3-month period (August-November 2020). The chemical composition data included concentrations of the major components of PM_{2.5} such as OC, EC, water-soluble ions, and elements. These species, along with a large suite of organic tracers were used as inputs in a source apportionment model (PMF) to identify and quantify the sources of PM_{2.5}. In Canada, the NAPS program only provides data on organic compounds that can be measured by ion chromatography, which limits the available measurements to a small subset of polar organic compounds. Performing PMF analysis without organic species, or only a few polar organics, not only over- and underestimates some sources but also neglects some sources (Fakhri et al., 2023). On the other hand, performing PMF analysis with only organic species is valuable for understanding organic aerosol chemistry, but it neglects important sources that contribute to the PM mass such as secondary sulfate, secondary nitrate, sea salt and crustal dust. 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 source apportionment studies, is the first of its sort in Canada. Furthermore, a focus 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 from the GC-MS analyses were namely, 6 n-alkanes, 2 fatty acids, 1 dicarboxylic acid, 2 biogenic secondary organic aerosols (SOA) tracers and hopane. This study demonstrates that having a small set of speciated organic tracers included in PMF input matrices is beneficial for understanding the sources of PM_{2.5} in Canada."

2) Referee 1 had several questions regarding presence of trace elements in some of the factors (pages 12, 15, 18 of the combined response document). The replies in the response document are informative and should be included in the revised manuscript (SI ok) to clarify the results for future readers.

Answer: The comment was taken into consideration and the following paragraphs were added to the manuscript (line 549-556 and line 568-572):

"Upon close examination of the PMF factor profiles, one notices some very small mixing of the traffic exhaust, road dust and crustal dust factors, which is a limitation of this study. However, 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, only 2% of Fe was allocated to the road dust factor while the amount of Al was 4%. Moreover, for the traffic exhaust factor, these values were 2% and 6% for Fe and Al, respectively. It is also possible that these metals are truly 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."

"A small percentage of Cu, Sb and Fe are attributed to Biogenic SOA. Specifically, Fe, Sb, and Cu were 5%, 6%, and 5%, respectively. Fe was allocated in much higher proportion 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 also included biogenic SOA tracers in the PMF analysis and in their biogenic SOA profile one also notices small amounts of metals/elements."

3) Sect 3.6.1: The possibility that the marine source is from road salt should be addressed in the main text not only the supplementary information. Are there any implications in road salt vs sea salt for the chemical mass closure?

The comment was taken into consideration and the following paragraphs were added to the manuscript (**line 520-529** and **line 172-178**):

"A marine factor was characterized by the ions Na^+ (46%), Cl^- (69%) and NO_3^- (30%), contributing to 11% of the $\text{PM}_{2.5}$. The Cl^-/Na^+ calculated for this factor was 0.95, which is lower than the ratio of 1.80 reported for fresh sea salt and is indicative of aged sea salt (Petit et al., 2019; Seinfeld and Pandis, 2016). The presence of high nitrate loading in the profile is also consistent with the presence of aged marine salt. The observed chloride depletion is due to the reaction of nitric and sulfuric acid with NaCl particles (Seinfeld and Pandis, 2016). While the factor has been tentatively identified as "marine", there is some evidence that this factor may originate, at least partially, from road salt. The marine factor exhibits relatively high concentrations for multiple wind directions including from the west and southwest (Fig. S10), and thus, the marine factor pollution rose resembles to some extent that of road dust. It is also notable that the marine factor exhibits its highest concentrations in November when minimum temperatures were below freezing, and some snowfall occurred. Based on these findings, we suggest that further work is needed to evaluate the contribution of road salt to $\text{PM}_{2.5}$ in Montréal."

In the chemical mass closure calculation, we assume that Na^+ originates from sea salt, and the ratios between Na^+ and other ions (SO_4^{2-} , Ca^{2+} , K^+ , and Mg^{2+}) in sea salt aerosol are the same as those for seawater. However, it is possible that some Na^+ originates from road salt, which is principally composed of NaCl , with a small amount of CaCl_2 (Charbonneau, 2006). In this case, the contributions of SO_4^{2-} , K^+ , and Mg^{2+} would be overestimated, and the true concentration of the "road/sea salt" component would be less than that calculated. In the extreme case of the component being derived entirely from road salt, the overestimation in the concentration of this component would be approximately 20%, given the preceding equations. This error is relatively small because SO_4^{2-} , K^+ , and Mg^{2+} have relatively small concentrations in sea salt..

4) Sect 3.6.2: Please add to the main text a version of the text provided in the referee response document (pg 34) regarding the potential sources of sulfate within Québec and how this could explain the similarities in contribution from the US and Québec.

The comment was taken into consideration and the following paragraph was added in the manuscript (**line 627-633**):

“Regarding the sources of sulphate in Québec, it is somewhat surprising that the contributions from the province and the US are essentially the same (35% vs. 33%) given that there are no coal-fired powerplants in Québec while coal is still used at some powerplants in the US. This finding indicates that it is important to consider other sources that contribute to sulphate regionally. Specifically, aluminum production is a major industry in Quebec that emits large amounts of SO₂ (NPRID, 2022). Nearly, 70% of North American aluminum is produced in Québec. In addition, other industries involving smelting and metallurgy in Québec emit SO₂. When also considering the recent decreased use of coal in the US (USEIA, 2022), these alternate sources of sulphate appear to be relatively important in Québec.”

Technical

1) Line 77: Please include the information on the specific species that were included in PMF like is already present in the abstract.

Answer: The comment was taken into consideration and the sentence was updated in the text (**line 80-83**):

“One objective of this work is to investigate previously unresolved PM sources in Montréal, by using some selected organic markers, namely six n-alkanes, hopane, two fatty acids, one dicarboxylic acid, and two biogenic secondary organic aerosols tracers and hopane in the PMF model.”

2) Line 91: Please list how long samples were typically stored before analysis.

Answer: The comment was taken into consideration and the following sentence was added to the manuscript (line 98-99):

“Collected filters were also stored at -20 °C until analysis. Organic species and elements were immediately quantified following the field campaign (i.e., within 3 months). Analyses of the water-soluble ions, sugars, OC, and EC were performed a year after the field campaign.”

3) Line 175: CF has not yet been defined. I also encourage you to consider including an equation that shows the calculation explicitly so that it is clear how CF is being used.

Answer: The comment was taken into consideration and the paragraph was updated to (line 190-196):

“To account for unmeasured O, N, S, and H atoms in OM, the conversion factor (CF) from OC to OM was derived using the equation $OM = CF \times OC$. The method used to calculate the CF sums all the PM components while systematically varying the OM/OC conversion (Genga et al., 2017). 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.”

4) Figure 4: Please clarify in the caption what the percent is referring to (i.e., percent of what?).

Answer: The comment was taken into consideration and the caption in the manuscript was updated to:

“Figure 4. Profiles of the eleven factors identified from the PMF model. The left axis corresponds to the concentration of each species (blue bars) and the right axis corresponds to the percentage of each species (orange markers). Units of concentration are ng/m³.”

5) Line 606: Since “other” is the largest contribution, please revise to clarify that Québec represents the highest of the apportioned contribution.

Answer: The comment was taken into consideration and the conclusion was updated in the manuscript (line 634-636):

“On the other hand, anthropogenic dust emissions from Québec presented the highest apportioned contribution to total dust concentrations among the three regions studied, and the concentrations dropped by 16% when emissions from Québec were excluded and by 10% when US emissions were excluded.”

6) Figure S6: Please include the meaning of the red box in the caption.

Answer: The comment was taken into consideration and the caption in the SI was updated to:

“Fig. S6: The temporal variation of nitrate concentrations for the sampling period at the MTL site. The red box indicates the period (end of October and November) where the nitrate concentrations were higher in comparison with the warmer months.”

7) Figure S10: Thank you for adding this figure. I think it is helpful and adds to the manuscript. However, the figure is currently very challenging to interpret. I recommend selecting different colors (please pay attention to color blind accessibility). It would be helpful to consider using a scale that can more intuitively be interpreted in terms of increasing mass.

Answer: The comment was taken into consideration and Figure S10 was updated in the SI.