

The authors would like to thank the anonymous referee # 2 for taking the time to review the manuscript. We thank for validating our work and for providing us with valuable insights that allowed us to improve the manuscript.

Below you will find the list of the referees' observations (bold), right after, each of the author's responses (normal font) and the respective changes made to the manuscript (italic), highlighting the sections that were modified.

Minor comments

- 1. Fig. 1. The detail map of the sampling site has wrong color scale (opposite to the map of South America) and must be corrected.**

We thank the reviewer for this remark, the colors of the scales were modified for coherence with the adjacent plot.

- 2. Line 212: Typo - should be "hopanes" instead of "hopaes"**

We thank the referee for spotting this typing error. It is now corrected.

- 3. Line 225, Multisite PMF: the dimensions of the final matrix should be mentioned, number of variables used in final run is missing.**

The dimensions of the final matrix were included in the description of the methodology.

"For this purpose, in order to combine both datasets as one (EA-LP) the dates of the La Paz dataset were shifted in time by two years and then appended to El Alto's dataset, thus avoiding repeated dates and composing a single input matrix for PMF that respected the natural seasonal variability of the original datasets. The dimensions of the resulting matrix were 185 rows (samples) x 40 columns (species). The multisite approach stands on the hypothesis that the major sources contributing to PM₁₀ in both sites are similar and display similar chemical profiles, which has been verified within the single site solutions."

- 4. Line 240, fuel fingerprint: The units in results of chemical analysis of fuel in SI are apparently wrong, must be corrected. Moreover, the reviewer is surprised by high concentrations of arsenic in gasoline in comparison with lead and zinc. Is that really correct?**

We thank the referee for spotting the missing magnitude correction. Concentrations are indeed 2 orders of magnitude lower and were corrected in the SI as displayed below:

Table S3. Species analyzed from fuel samples collected at La Paz and El Alto

	<i>Sample#</i>	<i>Al</i>	<i>Cr</i>	<i>Mn</i>	<i>Fe</i>	<i>Co</i>	<i>Ni</i>	<i>Cu</i>	<i>Zn</i>	<i>As</i>	<i>Ag</i>	<i>Cd</i>	<i>Pb</i>
		mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l
<i>gasoline</i>	1	13.7	3.96	63.0	2.81	0.05	0.23	0	0.85	4.00	0.12	0.002	0.10
<i>gasoline</i>	2	8.23	3.84	67.3	0.00	0.03	0.10	0	1.68	3.47	0.09	0.003	0.10
<i>gasoline</i>	3	8.54	3.51	61.7	0.00	0.03	0.07	0	1.71	3.07	0.01	0.004	0.10
<i>diesel</i>	4	14.9	9.82	0.66	0.00	0.02	0.13	0	7.85	4.00	0.01	0.005	0.25
<i>diesel</i>	5	15.8	8.81	0.41	0.00	0.02	0.13	0	6.55	3.74	0.00	0.007	0.27
<i>diesel</i>	6	35.0	10.2	0.91	11.6	0.02	0.11	0	7.49	4.36	0.01	0.011	0.28

Nevertheless, the ratios between the species are maintained, and are the values measured from the samples collected on site.

5. Line 250: Fig. number is missing in this line.

This constitutes a typing error that is now corrected. "Fig. 2" We thank the referee for spotting it.

6. Line 278 -280, equation 4. The equation contents mistakes. Sulphates are corrected for sea salt sulphates, but no sea salt is added. In addition, crustal element mass calculation is a bit strange, why calcium is mentioned twice and only part of other elements are corrected for oxygen, the same is valid for phosphor.

We agree with the referee, for which a more detailed description of the terms composing the equation was included in the main text. The form of the equation was modified based on a deeper bibliography research, which now includes a sea salt term and the oxygen correction for all crustal elements

"The reconstruction of the measured PM₁₀ mass resulted from the mass closure of the major components of PM, as described in Favez (2010), Putaud (2004), Seinfeld & Pandis (1998), Chan et al., (1997), Pérez (2008), and Cesari et al., (2016). Thus,

$$\text{PM(recons)} = \{(1.8[\text{OC}])\} + \{[\text{EC}]\} + \{([\text{SO}_4^{2-}] - 0.252[\text{Na}^+] + [\text{NO}_3^-] + [\text{NH}_4^-])\} + \{2.54[\text{Na}^+]\} + \left\{1.15 \cdot \left((1.89[\text{Al}]) + (2.14 \cdot (2.65[\text{Al}])) + 1.67[\text{Ti}] + (1.4 \cdot ([\text{Ca}] - [\text{Ca}^{2+}])) + (1.2 \cdot ([\text{K}] - [\text{K}^+])) + 1.36[\text{Fe}] + (1.5[\text{Ca}^{2+}] + 2.5[\text{Mg}^{2+}]) \right) \right\} \quad (4)$$

Where the first curly bracket accounts for the organic matter, the third one accounts for the sum of the mass of secondary inorganic aerosol particles (non-sea-salt sulfate, nitrate, and ammonium), the fourth accounts for sea salt, and the fifth curly bracket accounts for the mass of the main components of crustal material: Al₂O₃, SiO₂, TiO₂, CaO, K₂O, FeO and Fe₂O₃ (multiplied by 1.15 to take into account sodium and magnesium oxides), and the mass of unmeasured carbonates. "

7. Line 281 – 284: Water absorbed in aerosol and adsorbed on the filter can be also part of unresolved mass.

We thank the referee for the correction, also brought up by referee #1, which is now corrected.

"Average PM₁₀ (recons.)/PM₁₀ (meas.) ratios of 0.91 in El Alto and 0.82 in La Paz were found. The remaining unidentified mass fraction may be attributed to the loss of volatile organic matter and secondary aerosols post-weighing, during the transport of the filter fractions to be analyzed. The difference can also be associated to the presence of non-measured species (i.e. carbonates) or to the adsorption of water in the aerosol particles or the filter (Pio et al., 2013). Moreover A 10% uncertainty associated with the gravimetry measurements could also have a role in the observed difference".

8. Line 319 - Dust factor in El Alto, the authors mentioned that sampling site was surrounded with dusty surface. Although the sampling site was on the roof of the building, it cannot be excluded that sampling site is influenced by the local dust. It should be mentioned here.

We included a sentence to remark what was suggested by the referee.

"The dust factor has outstanding contributions of 32% in the city of El Alto, becoming the dominant source in this city. Although the volume sampler was placed on the roof of the observatory building, it cannot be excluded that the samples were influenced by the local dust. For La Paz, the vehicular emissions take the lead in terms of percentage contributions (35%). The factors associated to secondary aerosols (secondary sulfate, secondary nitrate, MSA-rich) were responsible for nearly

22% and 24% of total PM (La Paz and El Alto respectively), only a slight difference can be observed between the cities except for the nitrate rich profile.”

- 9. Line 320 – The factors associated to secondary aerosol ... It should be mentioned which factors. Probably sulphates, nitrates and MSA rich factors, but it should be specified.**

A specification was included in the text.

“The factors associated with secondary aerosol particles (secondary sulfate, secondary nitrate, MSA-rich) were responsible for nearly 22% and 24% of total PM (La Paz and El Alto respectively), only a slight difference can be observed between the cities except for the nitrate rich profile.”

- 10. “Line 441 – metals such as Cu, Sn, Sn and Pb. Not sure if one of Sn should be Zn or Sb (Zn seems more probable)**

We thank the reviewer for spotting the typing error. The duplicated Sn was replaced by Sb.

- 11. Line 473 – citation F. Amato et al., 2011 should be Amato et al. 2011**

The format of the reference was corrected

- 12. Line 598 – “natural anthropogenic sources” should be “natural and anthropogenic sources”**

We thank the referee for spotting this typing error that is now corrected: “there is a significant contribution of regional natural *and* anthropogenic sources of PM (Primary and secondary biogenic emissions, and biomass burning)”