Response to reviewer comments for the paper "Effect of vehicular traffic, remote sources and new particle formation events on the activation properties of cloud condensation nuclei in the Brazilian megacity of São Paulo".

We thank the reviewer for the valuable suggestions to improve our manuscript. We agree with the comments and have made all the suggested modifications. Below, our responses to each of the comments (in black) are provided (in blue). In the revised manuscript, we have highlighted the newly added text in yellow.

1. Abstract and Conclusion sections: "Our results show that particulate matter from local pollution sources, mainly local road traffic emissions, has a greater effect on CCN activation parameters than those from remote sources."

"In summary, local pollution sources, represented mainly by vehicular traffic emissions, had greater negative effects on activation parameters than did remote sources."

According to the authors, local sources are even not able to be discriminated in this study. So how can they conclude local sources had greater negative effect on CCN activation? Moreover, in figure 9, 10, 11 and 12, there are no AR or Dact data about local sources at all. How can they make any conclusion about local source effect? This is my major concern.

Response: In order to assess the main sources of aerosol pollution in the MASP, we carried out a positive matrix factorization (PMF) analysis, using total concentrations of trace elements and BC and carbon monoxide (CO) average concentrations (Fig. S4), as described below. In addition, some markers of vehicular traffic, such as Cr, Cu, Ti and BC, were plotted against carbon monoxide, measured on the main road Marginal Tietê. As can be seen in Figure S5, discussed below, those markers showed strong correlations with CO, which is associated with vehicular emission in urban areas. Our findings are in line with those of many studies, carried out in the MASP, which identified the vehicular traffic as the major local pollutant source in MASP. Those conclusions are in agreement with the great number of vehicles (7 million) in this area and the higher hourly mean vehicular traffic density during the daytime (8000 automobiles, 1500 motorcycles, 220 trucks and 90 buses, on Marginal Tietê). The follow paragraph has been included on page 12, lines 7-19, in the revised manuscript.

In order to assess the source apportionment of aerosol mass concentration, a PMF analysis was applied to trace-element, BC and carbon monoxide (CO) concentrations. The CO is an important marker associated with vehicular emissions in urban areas, its concentrations being measured at the Marginal Tietê. Our analysis showed that vehicular traffic had a great influence on all days, mainly during the daytime, and vehicular emissions + biomass burning had an influence only on days 6, 12 and 13, whereas sea-salt + industrial emissions had an influence only on days 7 and 8 (Figure S4 in the Supplementary Information).

Various studies have identified vehicular traffic as the main local pollutant source in MASP. The contribution of vehicular traffic is higher during the daytime and lower during the nighttime. Some species, such as Cr, Ti, Cu and BC, are frequently used as markers for vehicular traffic (Andrade et al., 2012). In this study, those compounds showed a strong correlation with CO during daytime. We also observed that BC and Cu correlated strongly with CO during nighttime, as illustrated in Figure S5 (Supplementary Information). In addition, the strong correlation between vehicular traffic marker species and CO confirms that vehicular traffic was the predominant daytime source on all days. The strong correlation of BC and Cu with CO at nighttime can be related to vehicular emission + biomass-burning events, as demonstrated by the PMF analysis.

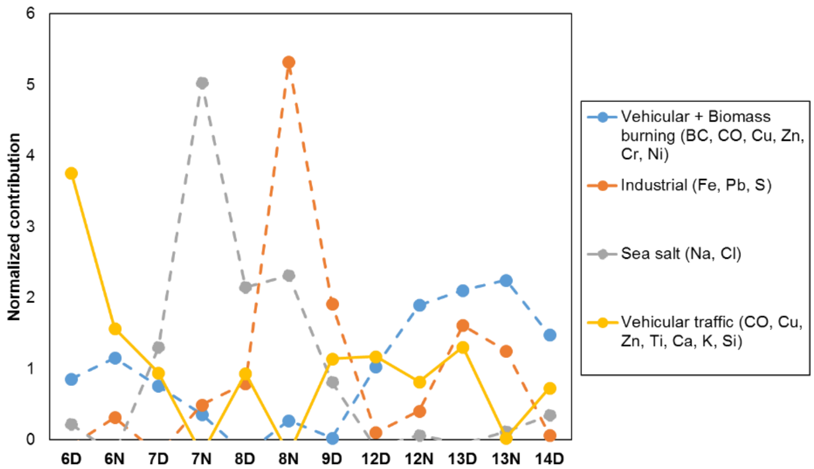

Figure S4. Source apportionment analysis performed by positive matrix factorization (PMF) model, using elemental, carbon monoxide and BC concentrations. Vehicular traffic showed major contributions in most days during daytime mainly, whereas vehicular + biomass burning had main contributions on days 6, 12 and 13. Sea salt and industrial apportionment was most pronounced during days 7 and 8.

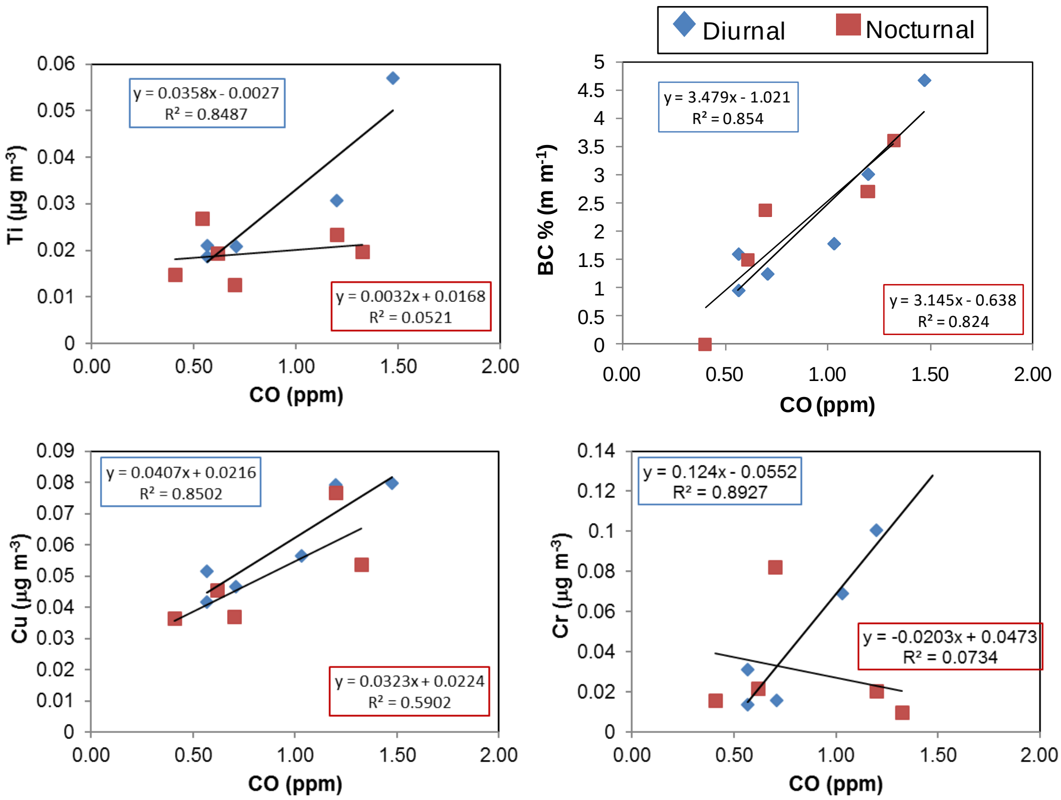

Figure S5. Elemental concentrations and BC versus carbon monoxide (CO) variability. The strong correlation between vehicular markers species and CO during daytime confirm the major apportionment of this source to aerosol during daytime mainly.

3. page 17 line 20-22,

"As can be seen in Fig.12c, diurnal periods with NPF showed lower AR values than did biomass burning and non-event days. However, AR values were similar among nights after NPF with those non-event and sea salt influence nights and also biomass burning after NPF events."

---In daytime NPF AR is about 0.05 lower than Biomass burning and non-event. Comparing with this, I would say AR in the night after NPF is also lower (not similar!) than those non-event and sea salt influence nights. Or I would say AR in all nighttime classes were just slightly different.

4. page 17 line 23-25

"Lower AR for nocturnal periods with sea-salt influence after NPF can be attributed to smaller particle size, whereas sea-salt events in these nocturnal periods occurred after low-O3 NPF events (on days 7 and 8)."

This sentence does not make sense at all. AR for nocturnal periods with sea salt + NPF is not low. Instead, it is the second highest among all.

Response: Questions 3 and 4 are addressed in the paragraph on page 16, lines 25-30, in the revised manuscript, as follows:

As can be seen in Figure 12b, days with vehicular traffic + NPF showed lower AR values than did those with vehicular traffic + biomass burning and those with vehicular traffic only, without daytime events. In addition, AR values were slightly different at nighttime. The nighttime increase in AR after NPF events was related to particle growth (Fig. 12b). In relation to D_{act}, during nighttime, biomass-burning events promoted slightly higher D_{act} values than did NPF and sea-salt transport, which is attributable to lower particle hygroscopicity. Although there was a lack of statistical significance, we observed a tendency for biomass burning to affect the air masses arriving at the MASP by decreasing the activation properties.

6. I suggest to use daytime and nighttime, which are more commonly used in the literature, to replace diurnal and nocturnal throughout the manuscript.

Response: In all instances, the terms nocturnal and diurnal have been replaced with nighttime and daytime in the revised manuscript.

7. I suggest to shorten section 2.6 and 3.4

Response: These sections have been reviewed and shortened in the revised manuscript.