

Anonymous Referee #1

The authors thank Referee #1 for highlighting the originality of the present article. The reporting of response factors is indeed essential for ACSM measurements. In this respect, a dedicated table and a small paragraph will be added in the revised manuscript:

“Throughout the measuring period, three Response Factor (RF) calibrations and one $(\text{NH}_4)_2\text{SO}_4$ calibrations were performed, summarized in Table 1. The low drift of the obtained slopes allowed the use of an average response factor of $2.72 \cdot 10^{-11}$ (with an standard deviation of $\pm 13\%$),”

Table 1. Response factors obtained through IE calibrations from June 2011 to May 2013

Date	Response Factor	RIE_{NH_4}	RIE_{SO_4}
16/11/2011	$2.31 \cdot 10^{-11}$	6	-
09/10/2012	$2.98 \cdot 10^{-11}$	4.8	-
15/05/2013	$2.84 \cdot 10^{-11}$	6.84	1.25
Average	$2.72 \cdot 10^{-11}$	5.88	-
Standard deviation	13%	17%	-

Anonymous Referee #2

The authors thank Referee #2 for all comments that have been raised.

The thorough evaluation of such novel instrumentation is much needed. A European intercomparison exercise has been carried out at SIRTa in November 2013, involving 13 ACSMs, and represents a unique opportunity to assess the reproducibility of ACSM instruments (Crenn et al., in preparation). Some lines will be added on this subject:

“A comprehensive determination of the overall uncertainty (as well as PM_1 components) associated to ACSM-derived measurements has been carried out in November 2013 through an inter-comparison exercise (Crenn et al., in preparation; Fröhlich et al., 2015). Here, the consistency of ACSM measurements has been assessed from the comparison with co-located measurements, as described in Section 3.”

With the presented dataset, we chose to compare the ACSM with collocated instruments at different timescales (high/low temporal resolution) over different temporal windows (short-/long-term). In this context, we only used PM_1 TEOM-FDMS data, PM_1 PILS-IC data and $\text{PM}_{2.5}$ filters.

The source apportionment of the organic fraction over long-term periods is scientifically very interesting, and although this topic is not covered in this article, it will be emphasized in the conclusion as a perspective:

“In parallel, the long-term characterization of the organic fraction would surely lead to a better assessment of aerosol sources and some (trans-)formation processes of secondary pollution in the Ile-de-France area.”

Please also note that a dedicated paragraph on this subject is already present in section 2.4.

Specific comments

1. Page 24227, line 21. ACSM measures PM_1 aerosols (unless they have a $\text{PM}_{2.5}$ lens). Do the authors mean that they used a $\text{PM}_{2.5}$ cyclone at the inlet?

AR: Yes, we used a PM_{2.5} cyclone inlet (in order to prevent coarse particles from reaching the PM₁ aerodynamic lens of the ACSM). It will be clearly stated in the manuscript as follows:

“Briefly, PM_{2.5} aerosols are sampled at 3 L/min (from a PM_{2.5} cyclone inlet) and then sub-sampled at 85 mL/min (volumetric flow) through an aerodynamic lens”

2. Page 24228, line 6. Technically, for ACSM, it should be “response factor” (see Ng et al), not “ionization efficiency”. Also, what is the variation in the response factor throughout the whole campaign? How often as calibration performed? What is the data saving interval? (every 30mins?)

AR: Yes, we agree. As also mentioned by Referee#1, more information should be given on this RF calibration. In this respect, a dedicated table and a small paragraph will be added in the revised manuscript (please see above our response to Referee#1)

The temporal resolution is indeed about 30 min, which will be specified in the manuscript as follows:

“A total number of ~26,000 ACSM data points (with a temporal resolution of 30 min) were collected from June 2011 to June 2013”

3. Page 24230, line 21. Are these TEOM data?

AR: Yes. It will be clearly stated in the manuscript:

“Hourly PM_{2.5} data from TEOM-FDMS measurements were retrieved from the three stations representative of the Paris urban background”

4. Page 24233, nitrate discussion. It is not clear how “no overestimation of ACSM nitrate is observed at high concentrations” would suggest “the ability of the Middlebrook algorithm to properly correct our ACSM collection efficiencies”. If “proper” CE means that concentration of the species measured by the ACSM is in agreement with other measurements, then all other species (sulfate, etc) should also be in agreement with other measurements? (but this is not quite the case).

AR: The collection efficiency is influenced by the mass fraction of ammonium nitrate (Middlebrook et al., 2012). To assess the validity of this correction, we used PM₁ PILS-IC measurements. The quasi perfect match between the 2 instruments (slope of 0.99) underlines the “ability of the Middlebrook algorithm to properly correct our ACSM collection efficiencies”. Then, although filters measurements can be influenced by some sampling artefacts and that sampling was performed at PM_{2.5}, the good slope (0.91) and r^2 lead to the same conclusion.

A different behavior is observed for sulphate, as it may be influenced by other types of artifacts that are not fully understood yet (Crenn et al., in prep; Frohlich et al., 2015). It is also to note that the determination of sulphate concentrations is furthermore subject to the determination of a specific Relative Ion Efficiency, independently of the choice of adequate CEs.

5. Page 24233-24234. It is not clear why the authors compared the PM1 ACSM data to PM2.5 filter data. It appears that PM1 filter data are available (see Figure 3). They should compare ACSM data to PM1 filter data.

AR: Figure 3 refers to the comparison with PM1 TEOM-FDMS. Unfortunately, there was no PM1 filter measurement available. This will be more clearly specified in the revised manuscript.

Nevertheless, we would like to underline that previous AMS (PM1) and PILS-IC (PM2.5) measurements in the region of Paris have shown very similar results, emphasizing on the fact that the fraction between 1 and 2.5 μm is not significant in this region (Freutel et al., 2013; Crippa et al., 2013).

6. Page 24233, sulfate comparison. Can the authors compare their PM1 sulfate data and PM2.5 sulfate data to support their hypothesis that sulfate associated with larger particles is a cause for the difference between ACSM and filter sulfate comparison?

AR: Unfortunately, there was no PM1 filter measurement available, and no PM1 vs. PM2.5 sulphate data obtained from the same methodology could be done. Moreover, various reasons can be raised to explain the observed discrepancies between PM1 SO₄ from ACSM measurements and PM2.5 SO₄ from filter sampling, such as size distribution and artifacts peculiar to the sampling technique. All combined, they might lead to compensatory errors, making it difficult to thoroughly determine the contribution of each artifact.

7. Page 24233, line 26. Is it appropriate to compare ACSM OM to PM2.5 OC? Please justify this.

AR: The comparison of ACSM OM with PM2.5 OC does not lead to a thorough conversion factor, although it has been shown that i) a few percentage of OM is located between 1 and 2.5 μm (Favez et al., 2007); and ii) the use of a constant OC-to-OM conversion factor is reasonable throughout the year as it brings to very satisfactory PM mass closure (Bressi et al., 2013). However, the relatively good r^2 obtained after the regression indicates that the temporal variations of ACSM OM make sense; but it does not directly validate absolute concentrations.

8. Page 24235, line 9. What discrepancies?

AR: These discrepancies are linked to an interannual variability observed in Fig. S2. The following sentences will be added for clarity: "The highest observed discrepancies occur with highest measured mass, which may highlight an intensification of pollution episodes. On a broader perspective, this feature is also observed through inter-annual variability of urban background PM2.5 concentrations (Fig. S2)."

9. Page 24236, lines 16. Why such a feature is only observed here but not in previous measurements (Megapoli/Ariparif-Particules projects)?

AR: This feature has not been seen during PARTICULES, essentially due to a low time resolution (daily filter sampling), and has not been investigated during MEGAPOLI. It will be clearly stated in the manuscript:

“, a feature which has not been seen during the AIRPARIF-Particules projects, essentially due to highly time resolved measurements, nor investigated during the MEGAPOLI project”

10. *Page 24241, line 4. This citation is for isoprene SOA. Do the authors expect high contributions of isoprene SOA in the region?*

AR: The determination from on-site measurements of the contribution of biogenic SOA in urban environments can be challenging, because isoprene can be emitted in significant amounts by anthropogenic sources. Von Schneidemesser et al. (2011) and Crippa et al. (2013) estimated a low contribution of isoprene on the OH reactivity in Paris. Modelling studies show conversely a much higher contribution (more than 90%) of biogenic SOA in Paris, especially in summer (Petetin et al., 2014).

11. *Page 24241, line 5. It seems that nitrate and OM have a more different diurnal trends on the weekends. Why?*

AR: Nitrate and OM show indeed a slightly different pattern during the week-end, in winter and summer, respectively. However, the scales are quite zoomed in, and as we do not want to overinterpret the data, we decided to focus on more obvious general trends.

12. *Figure 6. Please states clearly how seasons are defined (which month to which month). I suggest using a different color scheme for the seasons, so as not to be confused with ACSM species. It is not clear what the authors meant by “each data point correspond to 1 ACSM measurement”.*

AR: Seasons were differentiated by the seasonal equinoxes. This will be clearly stated in the manuscript.

By “each data point correspond to 1 ACSM measurement”, we wanted to emphasize on the fact that each average bin has been calculated from 30-min ACSM datapoint. As this may add some confusion; the sentence will be deleted.

13. *Figure 7. What is SO42- in this figure? Sulfate measured by ACSM? PM1 filter sulfate? PM2.5 filter sulfate?*

AR: It corresponds to ACSM (PM1) SO_4^{2-} . It will be clearly stated in the figure caption.

14. *Figure 8, what is the color scale of the wind rose? Why NH4 data are not included in this figure?*

AR: The color scale represents the Joint Probability Function (described in Henry et al., 2009), and corresponds somewhat to the occurrence of any wind direction and speed. A colorbar will be added for clarity.

NH₄ is not included in the NWR because, as it is quasi entirely combined to NO₃ and SO₄, it would lead to redundant information.

15. *Figure 10, why is sulfate data not included here?*

AR: Sulphate presents poor diurnal variations (average of $0.75 \mu\text{g}/\text{m}^3 \pm 2\%$), linked with the fact that sulphate is mostly advected over the Ile-de-France region, and de facto not influenced by potential local sources. This is why we didn't include this specie. More precision will be added: "Sulphate variations are not presented and discussed here because they lead to poor daily variations (average of $0.75 \mu\text{g}/\text{m}^3 \pm 2\%$), which are consistent with its mid-to-long range transport origin"

16. *I think the authors intended to use BC / SO₄ to denote local vs. regional/advected pollution. However, throughout the manuscript, there are multiple sentences noting that this ratio is used to denote local/regional vs. advected PM – this needs to be corrected.*

AR: The use of the BC/SO₄ ratio was intended to assess local, regional, and advected patterns. However, this distinction was, indeed, not evident throughout the manuscript. It will be corrected throughout the manuscript, as follows:

"BC-to-sulphate ratio used here as a proxy of the local / regional / advected contribution of PM"

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