AUTHORS’ RESPONSE TO REFEREES

The authors are grateful to the referees for their generally supportive comments on this work. The suggestions and comments made to the manuscript have been taken into full consideration when producing the revised version of the manuscript, and they have helped to improve the quality of the paper. The response to each of the suggestions and comments is written below in a point-by-point manner, with reviewers’ comments in bold and the replies in non-bold. Moreover, a revised manuscript with the changes tracked for easier reference is provided.

REFEREE#1

Overall

This paper presents real-time measurements of PM1 inorganic and organic species from a continental background site in the Western Mediterranean Basin over a 10 month period. Variability in PM1 components was attributed to boundary layer changes, air mass origin and meteorological conditions at the local, regional and continental scales. Only organic aerosol, which was composed mostly of oxygenated organic aerosol, exhibited marked diurnal cycles suggested to be associated with biogenic aerosol formation in the summer. In winter, both organic and inorganic aerosols show diurnal variations influenced by boundary layer dynamics.

This is a relevant paper for ACP and would be of interest to ACP readers. The paper is well written, with clear study objectives, logically presented and articulated conclusions. I have a number of minor revisions and comments that are recommended before acceptance to ACP.

My comments are as follows:

p. 28812, L1-7 - should indicate that the ACSM is built upon the same technology as the AMS, to make it clear that they are not completely separate methods.

As suggested, the text has been modified as follows:

"The ACSM is built upon the same technology as the AMS, in which an aerodynamic particle focusing lens is combined with high vacuum thermal particle vaporization, electron impact ionization, and mass spectrometry. Modifications in the ACSM design (e.g. lack of particle sizing chamber and components, use of simple and compact RGA mass spectrometer detector), however, allow it to be smaller, lower cost, and simpler to operate than the AMS (Ng et al., 2011)."

p. 28812 L 27 – 'elevated emissions of anthropogenic emissions occur' – what is meant by this? Emissions in this region have increased over time and over what time period?

The authors agree that this sentence is unclear, therefore we have eliminated it in the new version of the manuscript.

p. 28813, L14-20 and Section 2.4 – It would be helpful for the reader to know the directions from which these influences are from without having to go to Supplementary, especially if unfamiliar with this region. The authors may want to consider putting Fig.S1 into the main manuscript. In addition, although the authors do refer to Ripoll et al. (2014b) in Section 2.1 for a site description, it would be helpful to have at least a brief description here especially in the context of the types of air masses and frequency intercepted at this site.

A detailed description of the site and its influences is included in the methods section (and extended as suggested as described below). The authors wanted to keep the introduction relatively short, nevertheless the text in the introduction has been changed as follows to better clarify the location of the site:

"In this study we deployed an ACSM at a high altitude site (Montsec, 1570 m a.s.l.) in the NE of the Iberian Peninsula (42º 03' N, 0º 44' E), representative of the continental background conditions of the Western Mediterranean Basin (WMB) (Ripoll et al., 2014)."

Regarding the Fig. S1, the authors acknowledge the inconveniences of having to go to Supplementary to know details of the site, but we decided to include Fig. S1 in the supplement in order to reduce the number of figures in the main text. Moreover, a similar figure is already published in the main text of Ripoll et al. (2014), as it is referred to in the main text (page 28814 line 5).

As suggested by the reviewer, an additional description of the site has been added in Section 2.1. Moreover, Section 2.4 (which described the air mass scenarios) has been put together with Section 2.1 so that now Section 2.1 describes a whole picture of the site. The text is now as follows:

"Montsec site (MSC) is located on the highest part of the Montsec d'Ares mountain, at an altitude of 1570 m a.s.l., in a plain near to the edge of a 1000 m cliff to the south, with no wind obstructions present around. It is located in the NE of the Iberian Peninsula (42º03'N, 0º43'E), 50 km S of the Pyrenees and 140 km NW of Barcelona (Fig.S1). A detailed description of this site can be found in Ripoll et al. (2014).

The daily classification of atmospheric episodes affecting MSC was made following the procedure described by Ripoll et al. (2014) using HYSPLIT model from the NOAA Air Resources Laboratory (ARL). Air masses reaching MSC are mainly from the Atlantic (62% of the days) all along the year. From March to October, North African (NAF) episodes are more frequent (17% of the days) and very often are alternated with the summer regional (SREG) scenarios (12% of the days). The winter regional (WREG) scenarios are detected from October to March (11% of the days), as well as the European (EU) episodes (11% of the days). Conversely, the Mediterranean (MED) episodes are detected sporadically (4% of the days).

The boundary layer height was calculated using the Global Data Assimilation System (GDAS) model from the NOAA Air Resources Laboratory (http://www.ready.noaa.gov/READYamet.php) (Fig.S2)."

p. 28814, L 15-17 – this sentence is unclear. Could fix by putting (the Relative Ionization Efficiency (RIE)) all in brackets. Or reword.

The sentence has been reworded as follows:

"Since calibration of IEs for all species is not feasible, the Relative Ionization Efficiency (RIE) (compared to that of nitrate) is used (Jimenez et al., 2003)."

p. 28814 L 23 – Does having only 1 IE calibration limit the evaluation of the accuracy of this instrument? Please indicate uncertainties in the ACSM measurements. 62 minute time resolution is a strange sampling interval - Is this the time resolution used in the analyses?

The ACSM was calibrated several times through the sampling period and the values in the manuscript are the average values. In order to clarify this, the text in the manuscript has been modified as follows:

"Several calibrations were conducted throughout the sampling period, and average values of 2.2 x 10^{-11} for nitrate IE and 5.4 for RIE for ammonium were used for the whole dataset."

The detection limits for ammonium, organics, sulfate, nitrate, and chloride are 0.28 µg/m3, 0.15 µg/m3, 0.024 µg/m3, 0.012 µg/m3, and 0.011 µg/m3, respectively (Ng et al., 2011). The time resolution in the ACSM cannot be set as a chosen time interval, but it is the result of selected settings. In the present study, the sampling settings chosen were: 1 scan open (aerosol sampling) and 1 scan filtered repeated, 12 times with a mass spectrum scan speed of 1 s amu^{-1}, following the recommendations from the manufacturer at the time when the sampling was performed for a low-concentrations environment, and this resulted in a time interval between data points of 62 minutes.

p. 28816, L 22 – please describe what 'conventional real-time monitors are'? Thermos? Are they trace level instruments for a background site like this one.

The gaseous monitors are trace level instruments. More information on this has been added to the text as follows:

"Gaseous pollutants (O_3, NO, NO_2, CO and SO_2) were measured using real-time monitors belonging to the Department of Environment of the Autonomous Government of Catalonia. NO and NO_2 concentrations were measured using a Thermo Scientific instrument, Model 42i-TL; CO using a Teledyne 300 EU Gas filter correlation analyzer; O_3 using a MCV 48AV UV photometry analyzer; and SO_2 using a Teledyne 100 EU UV fluorescence analyzer.”

p. 28817, L9 change arithmetical to arithmetic.

The text has been changed as follows:

“Average concentrations shown in the whole paper are arithmetic averages unless otherwise specified.”

Fig S2 – please add a bottom x axis label – I presume this is UTC? In the manuscript please provide the conversion from UTC to local time.

The x axis label has been added to Fig. S2 as follows:

“Hour of the day (UTC)”

The conversion from UTC to local time has been included in the manuscript in section 3.2.1, as follows:

“Minimum ozone concentrations were recorded between 8:00 and 9:00 UTC (Coordinated Universal Time, which is local time - 1:00 and local summer time - 2:00), whereas maximum concentrations were measured between 16:00 and 17:00 UTC.”

Fig S3, figure caption, last line needs a rewrite.

The sentence has been changed as follows:

“Data points correspond to hourly values. Equations and red lines correspond to linear regression fits.”

p. 28818, L9-11, Please define summer and winter periods in the text. Why were these exact dates chosen?

The text has been changed as follows:

“For the sake of brevity only summer (14 Jul 11 – 24 Sep 11) and winter (10 Jan 12 – 7 Mar 12) hourly variation will be discussed …”

These exact dates were chosen because of the data availability, since in fall and spring ACSM data for some periods were not available.

p. 28819, L 24 – within the uncertainties of what?

The authors meant that the slopes were very similar when using OA with RIE of 1.4 and using OA divided by 1.54. Therefore the difference between slopes was likely lower than the differences in the slope when considering the uncertainties of the measurements (both the ACSM+BC concentrations and the co-located measurements). The sentence has been modified as follows:

“The resulting slopes were very similar and hence OA concentrations reported in the present paper were not corrected since further research is needed to better estimate the RIE for OA in the ACSM.”

Fig 2 –I find it difficult to determine the average concentrations in the stacked bar chart – fractional contributions are even difficult to pull out numbers, although it is stated in the text. One must refer to the supplementary to get the numbers. Can these be stated in the Figure or the text?

The authors agree with the suggestion and we have included the concentrations in the Fig. 2 as follows:

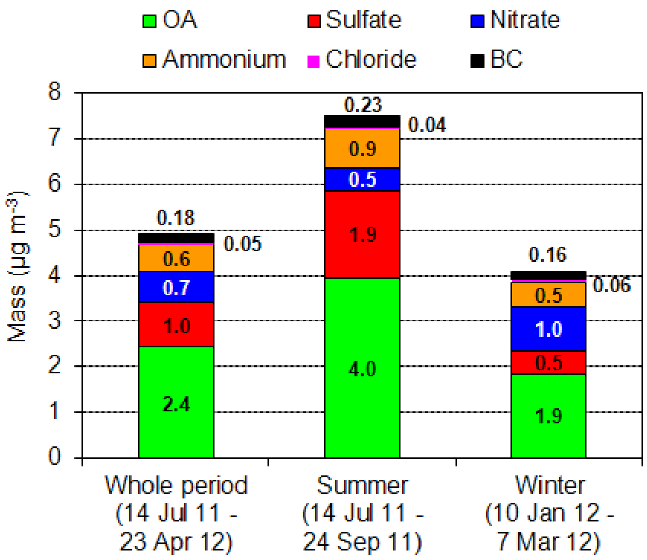


Fig. 1 Average concentrations of PM_1 chemical species measured at Montsec during the whole study, in summer and in winter.

p. 28820, L 7-12 – the summer maximum of PM components (except nitrate) is stated here to be due to photochemistry, but how does this reconcile with earlier statements (p. 28818, L17-23) that attribute seasonal PM1 mass concentrations principally to variations in the PBL? I see that seasonal differences must be due to both physical and chemical processes that change as a function of season.

The seasonal variation is indeed due to both physical and chemical processes that change as a function of season. In order to explain it clearly, the following sentence has been included at the end of the section 3.1.

“Moreover, the summer maximum has been ascribed to the higher photochemistry in the atmosphere that enhances the formation of secondary inorganic and organic aerosols (Querol et al., 1999).”

p. 28821, L 28-29 – Is there any evidence to support this hypothesis? For example, are there m/z markers from the ACSM indicate biogenic influence, perhaps during specific ‘biogenic events’ even though PMF was not able to pull out a biogenic factor. Are the increases in OA related to wind direction and transport patterns from areas dominanted by biogenics? In which direction are the biogenic sources predominantly

located? What time does the boundary layer reach the site in summer – is the increase in OA reflective of this?

The only evidence from these results is that diurnal cycle of OA during summer is completely different from the diurnal cycle of BC, sulfate, ammonium and nitrate (Fig. 4). If the increase of OA was due to the boundary layer effect, it would be also reflected in the rest of the chemical components. Moreover, if the OA increase was owing to anthropogenic emissions, it would be also reflected in the rest of the chemical components since they have anthropogenic origin as well. Therefore, the midday increase of OA is more likely due to the photooxidation of BVOCs.

Fig 4 – the blue colours showing nitrate and NOx are difficult to discern from each other.

The colours have been changed in the new version of the manuscript as follows:

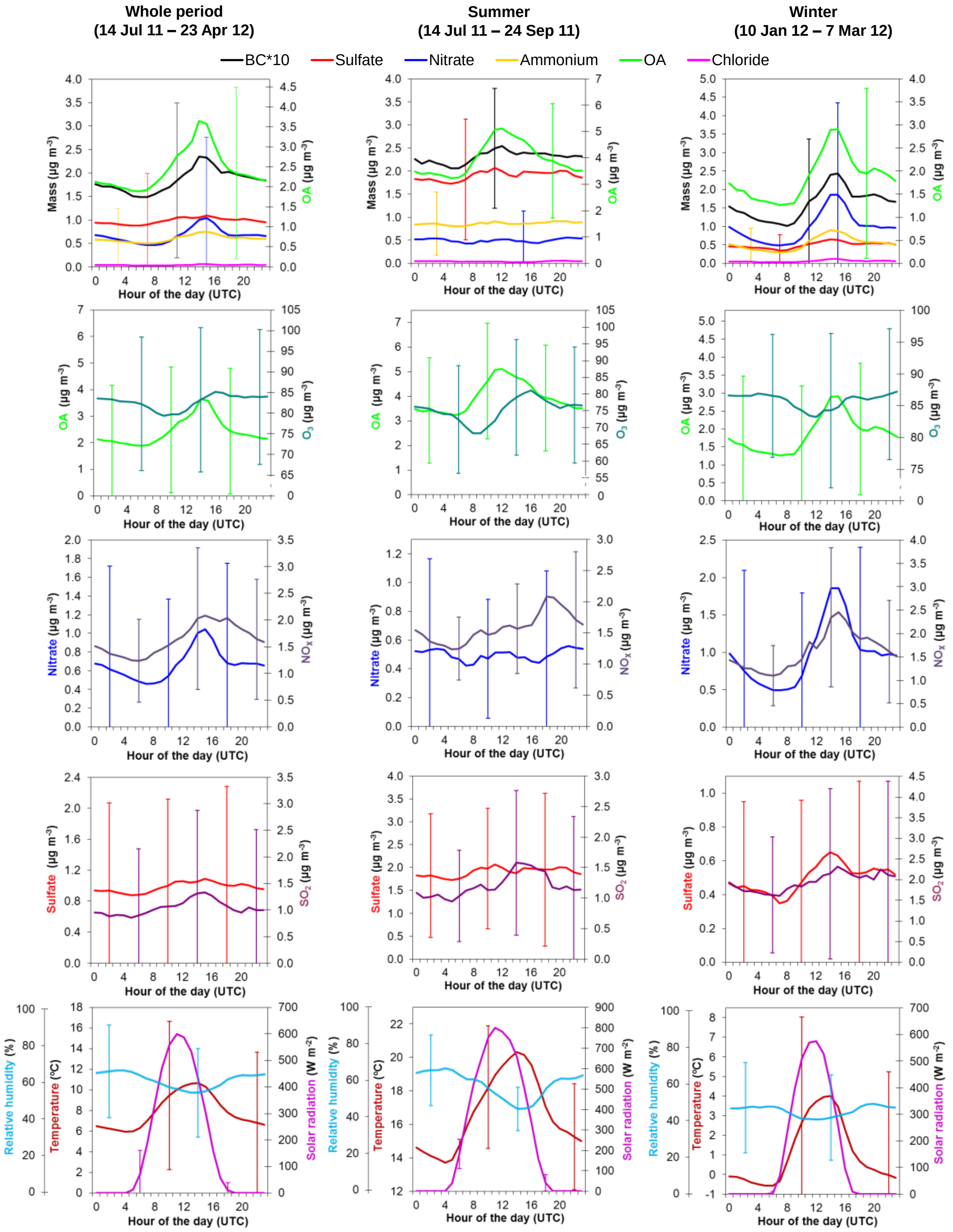


Fig. 2 Diurnal cycles of PM1 chemical species (black carbon (BC), sulfate, nitrate, ammonium, chloride and OA), gaseous pollutants (ozone (O3), nitrogen oxides (NOx), and sulfur dioxide (SO2)), and meteorological parameters (relative humidity, temperature and solar radiation) averaged for the whole period, summer and winter. Variation bars indicate ± standard deviation.

p. 28822, L10, Please clarify how SOA formation is compared to another site in a meaningful way using the difference between day minus night. Over what time period? How is dilution accounted for?

The authors' reasoning was that the average daytime minus night OA concentrations in summer was the SOA formation near to the site (recently-produced SOA) from biogenic emissions because if this SOA increase was from anthropogenic emissions or from long-range transport it would be also reflected in the rest of the chemical components. So, if we compare the summer diurnal cycle of OA obtained at MSC with that obtained at Puy-de-Dôme (Freney et al., 2011), the midday increase is higher at MSC than at Puy-de-Dôme. The differences in dilution between the two sites have not been calculated, but the difference in the daily pattern is evident enough to make such a statement (higher SOA formation at MSC), although this difference cannot be accurately quantified as pointed out by the reviewer.

L 13 – higher SOA in Mediterranean environments compared to what?

The authors referred to the European region. In order to clarify this, the text has been changed as follows:

"This is in agreement with the modeled SOA emissions over Europe, which identified higher SOA concentrations in Mediterranean environments (Bessagnet et al., 2008)."

p. 28822, L15 – do you mean Boreal forested areas?, Don't know what Boral areas are.

We apologize for the typo (Boral instead of Boreal). The Boreal areas are indeed Boreal forested areas. To make it clear "forested" has been added to the text as follows:

"This higher SOA formation is probably due to the higher emissions of BVOCs in the Mediterranean forested areas (up to 3 times higher than Boreal forested areas)…"

Fig 5 – Meteorological situations in the caption need to be identified (acronyms and descriptions in the text need to be defined) as in Fig 3. Also very difficult to discern between the EU and the WREG background colours; please adjust colours.

The acronyms have been added to the Fig. 5 caption and the colours have been changed as follows:

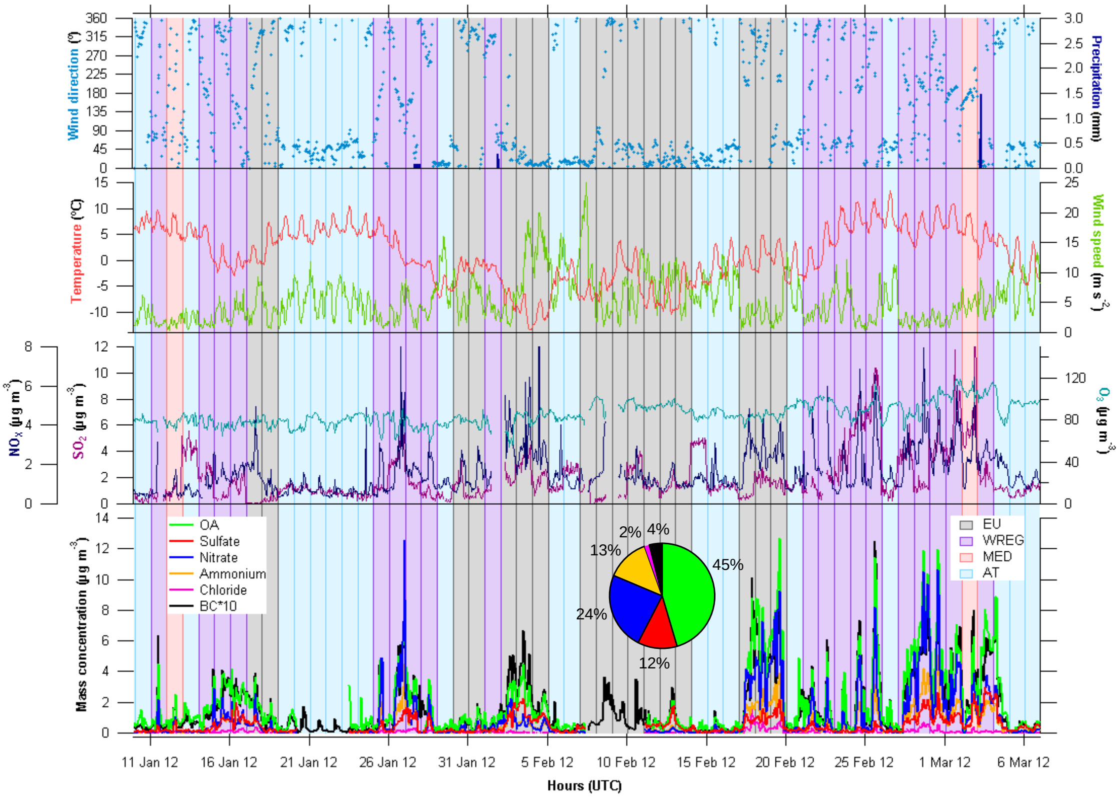


Fig. 3 Time series of wind direction (WD) and speed (WS), temperature (T), precipitation (PP), concentrations of nitrogen oxides (NO_x), sulfur dioxide (SO_2), ozone (O_3), and PM_1 chemical species (organics, sulfate, nitrate, ammonium, chloride and black carbon (BC)) in winter (10 Jan 12 – 7 Mar 12). Background colors correspond to daily classification of atmospheric episodes (European (EU), winter regional (WREG), Mediterranean (MED) and Atlantic (AT)) and the pie chart correspond to the average chemical composition for the winter period.

p. 28823, L7-9, In both winter and summer the site is in the FT all night and through convection (less in winter) the boundary layer eventually reaches site elevation (except Oct-Jan). However, how do these mechanisms explain the diurnal behaviour in winter, but not in summer? Are mountain breezes not prevalent during the summer?

The principal difference between mountain breezes dynamics in summer and in winter is that in summer these local processes are masked by the synoptic circulation, and therefore the diurnal variation in summer is dominated by long-range and/or regional transport. Moreover, summer regional recirculation of air masses over the WMB (Millan et al., 1997), induced by an abrupt orography, can cause the injection of aged pollution aerosols into the free troposphere, where they attain a longer life-time and/or accumulate in high reservoir layers. As a result, nocturnal aerosol concentrations increase in summer.

p. 28823, L25-38 – If the comparison here is with the Atlantic data, the plots do show increased background concentrations of PM1 components, but I don't see how the midday increments are lower; please clarify. The amplitudes appear greater. This whole paragraph is confusing when using the words 'In contrast' and 'On the other hand' – must be very careful to explicitly say what is being compared. Also it is impossible to look at daily variations in Fig 5 eg. 17-19 Feb 2012 – reader can determine this kind of variation on such a plot.

In this paragraph the authors are comparing the diurnal cycle of PM_1 components under winter regional conditions, which is described in the previous paragraph and for this reason the paragraph (p. 28823, L20) start with "In contrast", with the diurnal cycles under Atlantic advections and European episodes. In order to make it less confusing the paragraph has changed as follows:

"The study of the daily cycles as a function of air mass origin (Fig.S7) showed clear diurnal patterns under winter regional episodes, as mentioned above, and less marked daily patterns when MSC is affected by Atlantic advections and long-range transport from mainland Europe. Under Atlantic episodes the concentrations of PM_1 components were very low and the standard deviations with respect to the average pattern were quite high, resulting in unclear diurnal patterns compared to those under winter regional conditions. During European episodes, which can be more intense at high altitude layers (Ripoll et al., 2015; Sicard et al., 2011), background concentrations of PM_1 components were higher and the midday increment was lower compared to those under winter regional conditions, resulting in less marked daily patterns…"

REFEREE#2

Overall

This paper presents long-term ACSM data from a remote mountain site in Spain. This is a relatively new instrument, but as the ACTRIS network matures, datasets of this nature are becoming more commonplace. Nevertheless, the work here could be considered novel because it is the first time this has been presented in this specific environment. The fact that it is such a remote site makes it important when considering regional transport and transformations. The paper is largely well-written and most of the comments I have are of a technical nature.

General comments:

The paper has a very large number of figures, so it is expected that many will be in the form of supplementary material rather than in the article itself. However, I sometimes feel that some of the most pertinent and interesting figures are actually the ones in the supplement rather than the main article. As a case in point, Figures S2 and S3 are referred to 3 times each in the text, whereas figures 7 and 8 aren't referenced at all. I would suggest that the authors consider moving these into the main article and the ones less important to the conclusions of the paper moved out.

The authors acknowledge the inconveniences of using too many figures in a manuscript and sometimes it is difficult to decide which figures are the most relevant. We decided to put Fig. S3 in the supplementary material because a similar comparison is shown in Fig. 1. Furthermore, figures 7 and 8 (which are figures 6 and 7 in the new version of the manuscript) are referenced 2 times in section 3.3. Nevertheless, as figure 6 was referenced only 1 time, authors decided to move it to the supplementary material.

I think the finding that the agreement with other instruments was improved when the OA was scaled is an important one and perhaps underplayed. On a technical level, this will have implications for all other ACSM (and potentially AMS datasets). Further investigation of this and exercising care in the quantitative use of ACSM OA data in the meantime should be key recommendations of this work.

The AMS RIEs are better understood, this issue is specific to the ACSM only. Nevertheless, the recommendation of further work on the quantification of OA is now included in the revised conclusions section as follows:

"Discrepancies of OA determined by ACSM with co-located measurements pointed to an overestimation by the ACSM probably caused by the use of the default RIE for OA, which could be lower than the actual one. Further research is needed to better address this issue."

<u>**Minor/technical comments:**</u>

P28815, L2: How close is 'very close'? The authors should be specific here.

As the experimental determination of the RIE for sulfate could not be done until one year later, the authors were not sure to specify exactly the RIE value. Nevertheless, the RIE for sulfate has been included in the new version of the manuscript as follows:

"RIE for sulfate was experimentally determined one year later and was found to be 1.26, although the default value was used for the current dataset."

P28815, L16: What other instruments are being referred to here?

The authors referred to OPC and SMPS, as it is shown in section 3.1

P28816, L4: 'Standard' gravimetric analysis is referred to, but there are many important procedural variables to consider, such as sample conditioning. More detail should be supplied here, or if a specific procedure was being followed (e.g. FRM), this should be referred to.

The details on the gravimetric standard used are supplied in the new version of the manuscript as follows:

"Daily PM_1 mass concentrations were determined by off-line gravimetric procedures according to the EN 12341 standard (CEN, 1999), i.e. at 20ºC temperature and 50% relative humidity. Furthermore, PM_1 chemical composition…"

P28816, L10: The model number of the OPC should be 1.107, not 1107. The refractive index assumed in its calibration should also be specified, as this is likely to have a significant effect on the quantitative volume concentrations.

The quantitative volume/mass concentration depends strongly on the refractive index indeed, for this reason calibration of the OPC is necessary. The OPC used in the present study is calibrated once a year by the manufacturer which use different lattices PSL refraction index of 1.59. Nevertheless, we recalibrate the instrument very frequently by comparing OPC measurements with gravimetric mass concentrations.

In order to clarify this, the text has been changed as follows:

"Real-time PM_1 mass concentrations were continuously measured by an optical particle counter (OPC, Model GRIMM 1.107 calibrated with different latex PSL refraction index 1.59)."

P28817, L1: The model number of the classifier unit (e.g. 3080) should also be given.

The model of the classifier unit has been added in the text as follows:

"The SMPS system comprises a classifier unit (Model TSI 3080) and a differential mobility analyzer (DMA, Model TSI 3081) connected to…"

P28817, L16: As I understand, HYSPLIT does not calculate the boundary layer height as such; it is a part of the NCEP reanalysis, that it can report it as a diagnostic. Regardless, more information should be given on how this quantity is derived and how it should be interpreted because clearly, a boundary layer height below the altitude of the site is not representative of the mixed layer at the site itself.

The National Centers for Environmental Prediction (NCEP) runs indeed a series of computer analyses and forecasts operationally. One of the operational systems is the GDAS (Global Data Assimilation System). At NOAA's Air Resources Laboratory (ARL) GRADS model output are used for calculating stability time series and therefore obtain boundary layer height.

In order to clarify this, the text, now moved to section 2.1, has been changed as follows:

"The daily classification of atmospheric episodes affecting MSC was made following the procedure described by Ripoll et al. (2014) using HYSPLIT model from the NOAA Air Resources Laboratory (ARL). Air masses reaching MSC are mainly from the Atlantic (62% of the days) all along the year. From March to October, North African (NAF) episodes are more frequent (17% of the days) and very often are alternated with the summer regional (SREG) scenarios (12% of the days). The winter regional (WREG) scenarios are detected from October to March (11% of the days), as well as the European (EU) episodes (11% of the days). Conversely, the Mediterranean (MED) episodes are detected sporadically (4% of the days).

The boundary layer height was calculated using the Global Data Assimilation System (GDAS) model from the NOAA Air Resources Laboratory (http://www.ready.noaa.gov/READYamet.php) (Fig.S2)."

P28818, L2: The slopes and intercepts from the regressions should be given here rather than referring to the supplement.

The slopes and intercepts have been included in the new version of the manuscript as follows:

"The scatter plots of ACSM plus BC concentrations versus PM_1 concentrations from the OPC and SMPS showed strong correlations (R^2=0.72 and R^2=0.87, respectively) and slopes close to unity (slope=0.94 and intercept=0.09 for the ACSM+BC vs. OPC, and slope=1.21 and intercept=0.86 for the ACSM+BC vs. SMPS) (Fig.S3)."

P28819, L19: The other site and period of the measurements should be specified, such that the other study can be identified in the future.

The authors agree that the other study can be identified in the future so we added the name of the site and the measurements period as follows:

"Similar series of intercomparisons with a similar discrepancy for OA has been found for a one-year dataset (June 2012 – July 2013) with the same instrument at Montseny site (Minguillón et al., 2015) and…"

Figure (general). The text and lines are too small. Please make these larger.

The authors acknowledge the inconveniences of using figures with so many variables because it makes the figure too busy, however we tried to make them as big as possible.

REFEREE#3

Overall

The paper describes almost 1 year of aerosol chemical composition measurements in the Western Mediterranean Basin. Seasonal and dial trends were presented for specific aerosol components as well as

statistical source apportionment analysis was performed. The latter allows evaluating and acknowledging the significant contribution from secondary aerosol formation. It is a very nice and well written study, enhancing the knowledge on aerosol composition and different sources. The topic and presentation is suitable for ACP and I would recommend publishing it after some minor improvements listed below.

<u>Specific comments:</u>

Abstract: PM1 is not defined

The definition of PM1 has been included in the new version of the manuscript as follows:

"…and organic submicron aerosols (aerosols with an aerodynamic diameter less than 1 µm) from a continental...**"**

Page 10, Line 2: SMPS is not measuring PM1, define an upper mobility and aerodynamic diameters for SMPS and discuss the differences from PM1, maybe, the disagreement won't be as large then.

The SMPS is not measuring PM_1 indeed, for this reason the range of aerodynamic diameters measured are described in section 2.3 (P28816, L28-29) as follows:

"…a scanning mobility particle sizer (SMPS) was installed to measure particle number size distribution of mobility diameters 11-350 nm in the summer campaign, and 8-450 nm in the winter campaign."

And only the data from the winter campaign was used for the comparison with the ACSM, since the range of aerodynamic diameters measured was wider, as it is pointed in the text (P28817, L2):

"The SMPS data for the winter campaign were also used to estimate the mass concentration to compare with the ACSM data."

However, a phrase has been added in section 3.1 as follows:

"The differences in the particle size range measured by the different instruments needs to be considered when assessing these comparisons."

Fig. 3: Chloride is not visible in the pie chart.

The authors agree that it is hard to see chloride from the pie chart of the Fig. 3, and we changed a bit the figure but as chloride is only 1% of the total mass it is difficult to improve much more the figure.

Page 13, Lines 28-30: Midday increase is up to 10 µg/m3, can it really be biogenic? Could you provide reference for such high biogenic secondary concentrations?

The authors do not understand where the 10 µg/m^3 quoted by the reviewer comes from, since such a number does not appear in the text nor in the figures. The midday increase is described in p28822, L1-4 as follows:

"Despite the marked diurnal cycle of OA regardless of air mass origin, the average increase during the day with respect to average concentrations during the night was higher under summer regional (**2.6 µg m^{-3}**) and North African (**3.0 µg m^{-3}**) episodes than during Atlantic advections (**1.3 µg m^{-3}**)(Fig.S6)."

Page 15, Lines 13-15: biogenic emissions in winter need more detailed discussion, sources, efficiency?

The authors agree that biogenic emissions in winter are much less important than in summer. However, the average maximum temperature in the lower valleys during winter was around 11ºC which is similar to the temperature reported in DAURE winter campaign at Montseny (Mediterranean forest) and they observed BVOCs emissions (Seco et al., 2011). Moreover, Steinbrecher et al. (2009) showed the seasonal variation of VOC emissions from natural and semi-natural vegetation in Europe, and some biogenic emissions were found in winter. Therefore, biogenic emissions in winter cannot be discarded.

In order to include this in the manuscript, the text has been changed as follows:

"These mountain winds transport anthropogenic emissions from the adjacent valleys and plains to the top of the mountain, with a maximum upslope transport in the afternoon. Moreover, biogenic emissions influence cannot be ruled out as average winter temperatures are high enough for them to occur (Seco et al., 2011; Steinbrecher et al., 2009)."

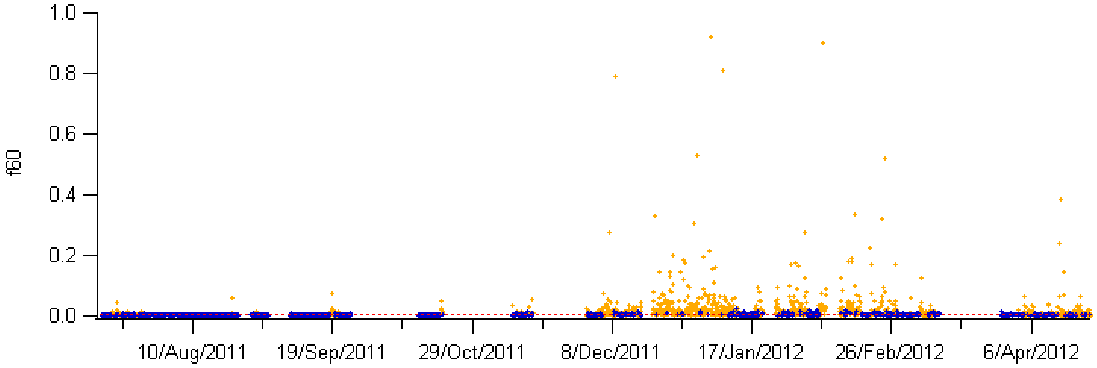
Paragraph 3.3: Could you provide some more arguments or considerations for 2 factor solution, as for now, m/z 60 and 73 are visible in LV-OOA, why it cannot be separated as a factor, did splitting start earlier? Please provide more details and arguments?

The authors have re-considered the solution chosen and hence a different solution is now included in the revised manuscript. We here explain why we chose the 2 factors solution and then we explain why we decided to change this solution in the revised manuscript.

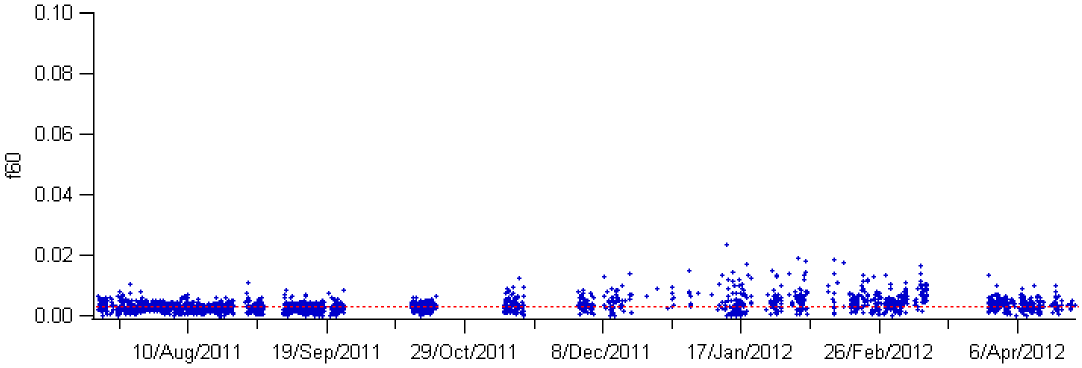
The reasons to choose the 2 factors solution were:

- The investigation of f60 showed that only some data points in winter were above the background level (0.003) found by Cubison et al. (2011), but a large fraction of these data points corresponded to very low concentrations of total OA mass, as it can be seen in the following figures:

a)



b)



Time series of f60 (a) colored in yellow all data points and in blue only the data points with OA concentration >1 µg m^{-3}, and (b) zoom of the f60 data points with OA concentration >1 µg m^{-3}. The red dashed line indicates the background level of 0.003.

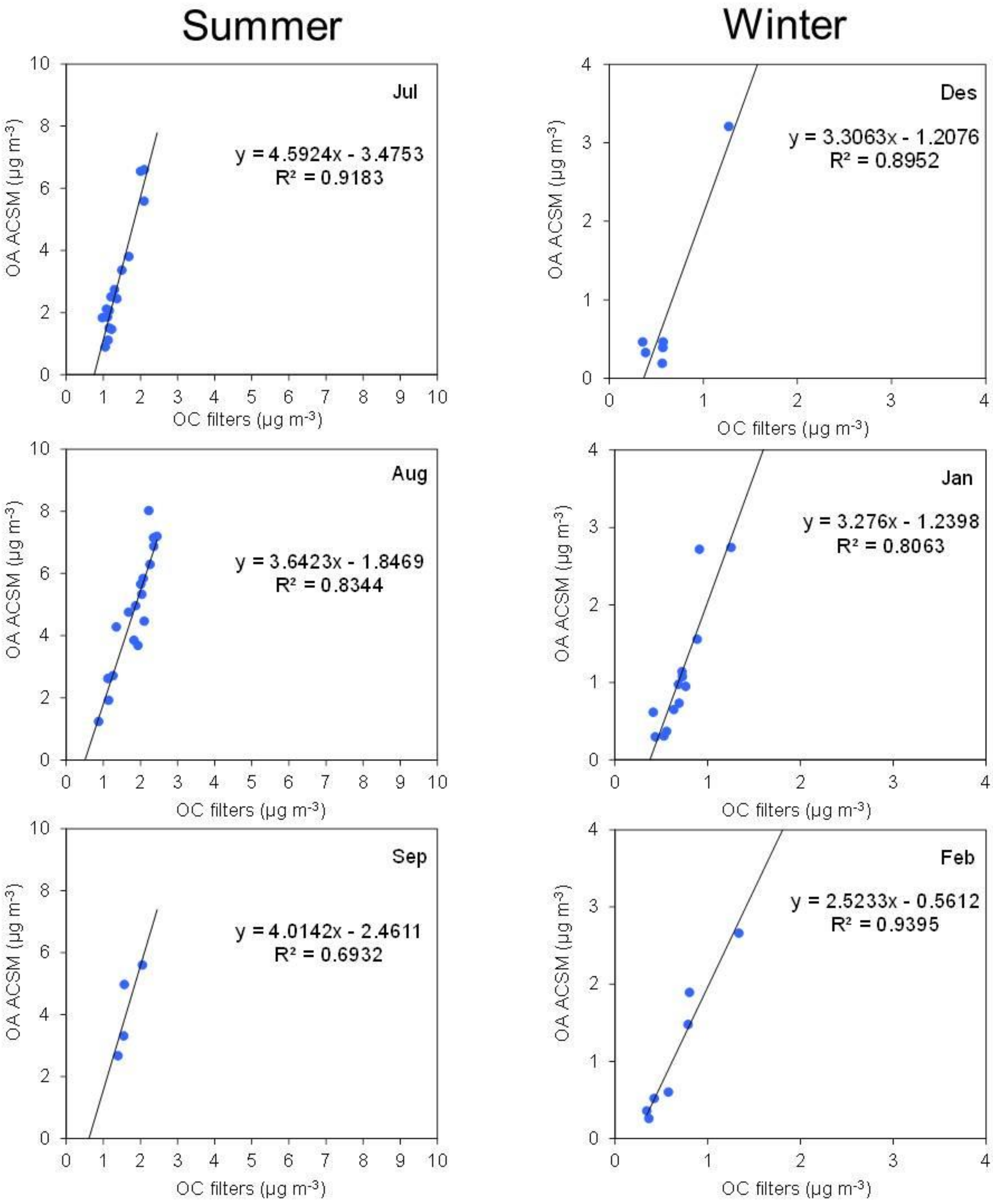
- In summer the f60 values were mainly below the background and hence no BBOA was expected in summer.
- We decided to take the entire dataset for the source apportionment to have a wider range of concentrations (usually low in winter) and hence the inclusion of a BBOA factor did not make much sense. We tried constraining a BBOA factor for the whole dataset but the solution did not have physical meaning.
- The 2 factors profiles from the 2 factors solution (with unconstrained PMF) in summer were similar to the 2 factors profiles in winter (with unconstrained PMF). Hence, a single source apportionment with the entire dataset was more robust and therefore this solution was chosen.

Based on the reviewer comment, we re-considered the presence of BBOA in the OA aerosol at Montsec in winter and hence tested more possible solutions by constraining some factors applying ME-2. We again divided the dataset into summer and winter to be able to constrain the BBOA factor. Therefore, we reached a different solution by applying ME-2, including 3 factors for winter (HOA, BBOA and OOA) and 3 factors for summer (HOA, LV-OOA and SV-OOA). Still, the majority of the OA is composed of OOA (91% in summer and 67% in winter), so the new solution does not differ largely from the one in the ACPD manuscript, but we think it explains better what the OA at Montsec is.

The authors refer the reviewer to the new version of the manuscript to see the details of the new chosen solution.

Page 18, Line 10: are you really so certain that it is an overestimation by the ACSM and not negative filter artifacts? Provide some more discussion, why one reason has been chosen over another?

The authors are not 100% sure that it is an overestimation by the ACSM but we think that it is the most likely possibility. If the problem was the negative filter artifact, we would expect some seasonal variation, but the slope is quite constant along the year as it can be seen in the following figure. It should be noted that the high volume samplers are kept inside containers at controlled temperature, so the samples never reach temperatures above 25ºC. Furthermore, similar discrepancy has been found for a study in Atlanta and they did not use quartz filters.



Additional comment from authors to the editor:

The authors list has been updated. It now includes Yuliya Sosedova, Francesco Canonaco and Andre Prévôt, from PSI, for their contribution in the OA source apportionment analysis of Montsec data by applying ME-2 with the SoFi tool developed in PSI.

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