AUTHOR'S RESPONSE

REFEREE #1:

Overall

The study concentrates on the aerosol particle measurements done in the southern parts of the Pyrenees, and gives valuable information on the behavior and sources of the sub and super-micron aerosol concentrations in western Mediterranean region. The overall measurements seem to be well made, but I have several issues related to presented quantities and on the way some of the conclusions are drawn. This, connected with some more minor needed corrections and clarifications mean that I suggest that the authors will have to do major corrections for this article for it to be accepted for publication in the ACP. However, the article is nicely written, and the overall results are relatively interesting, especially as they can give new reference on the particle behavior in region not well characterized before.

The authors acknowledge the reviewer for the detailed suggestions made to the manuscript. The response to each of the comments and suggestions is written below in a point by point manner.

Major comments and suggestions

First of all, I think it is very important to increase the network of quality measurements to the Western Mediterranean region, and I think the addition of MSC station as a background station will increase the coverage, especially due to the relatively large anthropogenic influences on MSY. The inclusions of these data sets (hopefully available somewhere soon?) will increase our understanding of the region. Perhaps the most worrying parts of the paper, however, are the lack of consideration of two key issues in the discussion and conclusions: The variability of the concentrations, and the role of the BL versus FT air on the concentrations.

Variability and conclusions:

In many parts of the paper, the authors claim that concentrations from one period to next or source region to next are larger or smaller than on some reference case. However, no indication of which mean value (arithmetic?) is used is given, which is especially worrisome as most of the aerosol properties are often log-normally distributed, making e.g. medians or geometric means as more natural comparison points. Outside of figures 4 and 11, no indication of the role of the variability in included in the discussions. I am not necessarily suggesting use of statistical tests (as the data sets will be strongly auto-correlating, making most of them rather tricky to do correctly), but I would always consider at least the overall level of variation (AND the amount of data per e.g. month!) in consideration of differences between the seasons. Good news seems to be that e.g. N on the different years seems to follow similar patterns. This is especially difficult on the Fig 3 and associated discussion. If you can not get the comparison values for the other stations it is fine (altough if you got them from EBAS [is this the "ACTRIS data centre"?], you should get them). But include at least the variability which you have. One other key problem in this paper is the comparison of different cut-off instruments at the station, when they have not measured in the same time! There is a lot of interannual variability (even though in these two years it seems to be

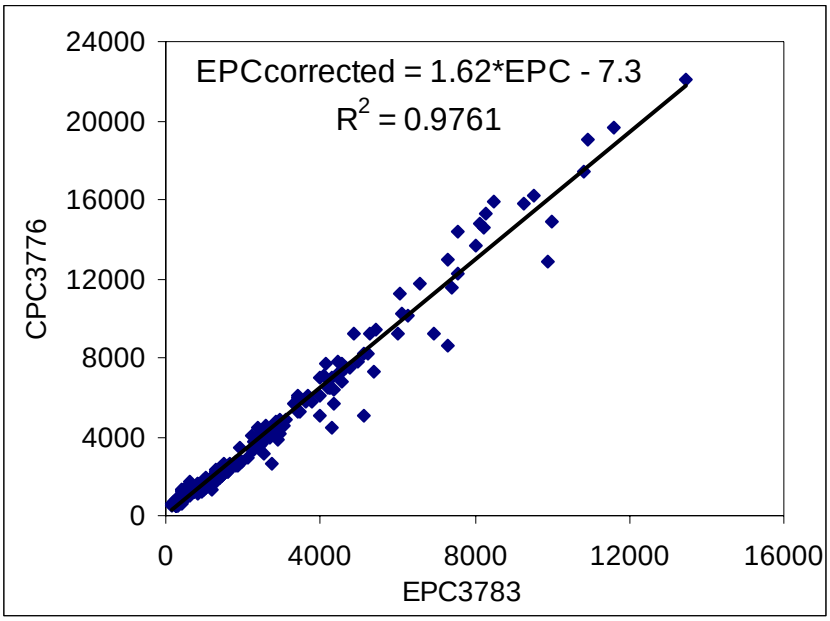
relatively stable), and thus you can not be sure that the differences are due different sized particles, instead of different annual situations.

The mean values shown in the whole paper are arithmetical averages unless otherwise specified (medians in the case of Fig. 4 and Fig. 10 of the new version for example). The text throughout the manuscript has been modified to better specified the type of average used. Moreover, the following sentence has been added in section 2.2 of the revised manuscript to clarify how the means have been calculated:

"The mean values shown in the whole paper are arithmetical averages unless otherwise specified".

Standard deviations have been included in the new version of the manuscript for the three-year averages of PM, BC and N concentrations measured at MSC and MSY. Moreover, arithmetic averages of weekly cycles have been changed for median averages and the percentiles 5, 25, 75 and 95 have been added (Fig. 11 and Fig. S9 of the new version).

Authors are aware of the limitations of comparing results from instruments with different size detection limits. However, the two instruments were measuring simultaneously during one month after the study period (April 2013), and the slope shows that the N_3 was higher than the N_7, which hence indicates that the differences between both instruments can be attributed to the particles in the range 3-7nm. The scatter plot and the correlation is shown in the plot below:



The amount of data per month suggested by the reviewer is shown in Fig. 9 of the new version for PMx, BC and N. Regarding the possible interannual variability, PM and BC seasonal patterns (Fig. 9) did not show a specific interannual variation from year to year in the three years study period. Thus, the authors think that the differences between the two sampling periods with different CPCs are more due to different sized particles than to different annual situations.

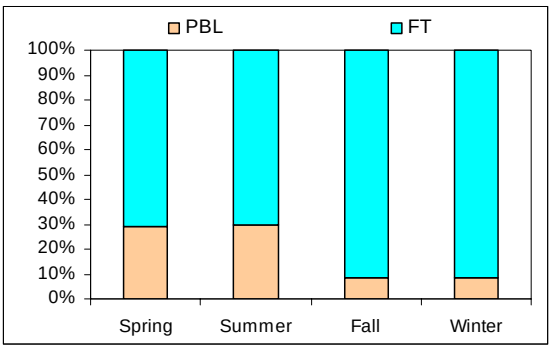
BL vs. FT

You indicate in figure S3 some idea of including the BLH variation on the analysis. This is also reflected on some parts of the text. However, the main results of this paper (concentrations) are strongly dependent on the BL/FT split, and thus the results can be indicative of mainly this. It is somewhat approached in the discussion, but for fairness I think that there should be some way (even roughly) to separate the concentrations between the two periods. For this reason, I strongly suggest that the authors include concentration histograms (e.g. in the supporting material) which could show e.g. two peaked distributions, indicative of BL/FT split. Accurate differentiation between the two will be very difficult, but even the rough estimates from modelled BLHs could be indicative. For this, e.g. re-analysis sets (with high time resolution) could be useful, and clearly already used by the authors. A difficulty could still arise from mountain winds, lifting air up (or down) hill based on large scale horizontal winds. This would not show easily on the modelled (rough resolution) BLH heights. This is a constant problem in all mountain measurements, and should be clearly stated and attacked in some form in the article. Some authors have used e.g. BC indicators, but I would not think this is a good idea in this paper, partly due to the shown BC long range transport issues. Perhaps add some discussion on this issue.
Overall: take the FT/BL split more into account in all analyses done in the paper.

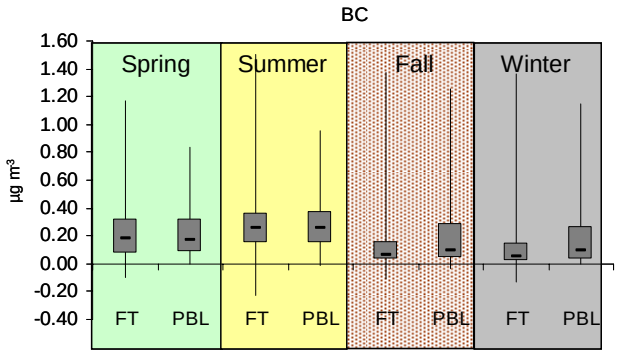
The authors are conscious of the importance of the PBL height in determining the aerosol concentrations variation in a mountain site such as MSC. For this reason, we included in the revised version of the supplementary material the variation of the aerosol parameters concentrations when MSC was within the PBL and when it was in the FT. This variation has been studied as a function of season, given that MSC is within the PBL during 30% of the time in spring and summer, whereas it only happens during 10% of the time in fall and winter (see figure below). As explained in the manuscript in section 2.3, PBL height was calculated using HYSPLIT model. This model uses a 50 km resolution grid. Since this horizontal resolution is not very good for an irregular terrain such as MSC (1000 m peak in just 20 kms, Fig.1 bottom), the estimate average terrain height from the model is around 580 m whereas the real altitude of MSC is 1570 m. Furthermore, the model gives only the PBL height every three hours and we assumed that the PBL height does not change during the next three hours after the datapoint. These limitations have to be considered when analyzing the data as a function of the PBL height.

The following figure shows the percentage of time that MSC is within or outside the PBL as a function of season and the concentrations of different parameters (BC, PM_{10}, PM_1 and N_3) as a function of the height of the PBL and the season. The figure has been added as supplementary material in the revised version of the manuscript.

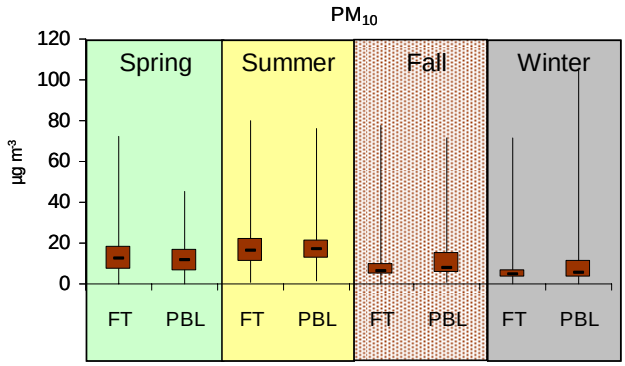
a)



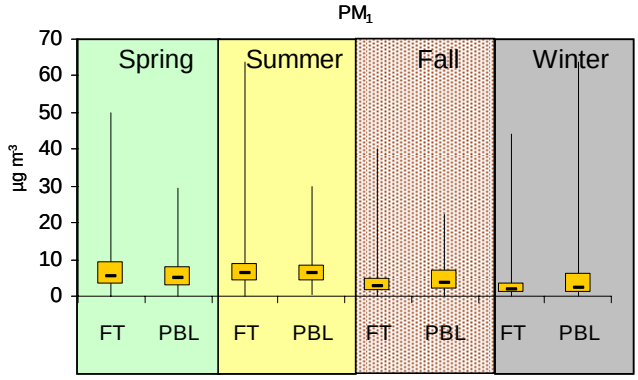
b)



c)



d)



e)

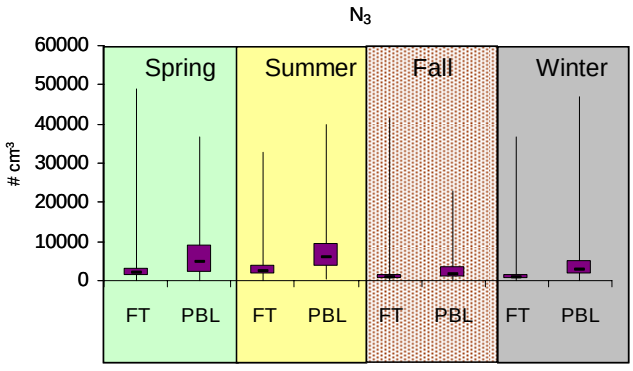


Fig. S5. (a) Percentage of time that MSC is within or outside the PBL as a function of season. FT indicates that MSC was in the free troposphere; PBL indicates that MSC was within the planetary boundary layer. (b-e) Median (black line within the boxes) and percentiles (5-25-75-95, boxes and whiskers) of BC, PM_{10}, PM_1 and N_3 concentrations during the study period as a function of the season and the PBL height.

Median average concentrations of BC, PM_{10} and PM_1 showed almost no differences between PBL and FT conditions in the warmer seasons, probably due to the regional and long-range transport of dust and pollutants at different altitudes, even in

the FT, and to the recirculation over the WMB, which cause the formation of reserve strata at a relatively high altitude (Millan et al., 1997), in these seasons. By contrast, during the colder seasons results showed some differences between PBL and FT conditions with lower concentrations of BC, PM_{10} and PM_1 under FT conditions, probably owing to the fact that during the colder seasons the long range transport is from the Atlantic for about 70% of the days (whereas this proportion reaches only 55% in the warmer seasons) and it is a transport of clean air masses. Moreover, the thermal inversions are very frequent in the colder seasons and these situations prevent the transport of pollutants from the most populated areas towards high altitudes. Median average concentrations of N showed more differences between PBL and FT conditions throughout the year, with lower concentrations under FT conditions. This could be due to the fact that N concentrations at MSC are more associated with a local origin than with a long-range transport.

This figure has been referenced in the revised manuscript in section 3.2.2, when explaining the higher summer concentrations with respect to winter, and pointing as one of the reasons the differences in the PBL height. The text reads as follows:

"The summer maximum is caused by a variety of factors: […] 6) the increase in the PBL height, which favors the mixing of atmospheric pollutants at regional scale (Fig. S4 and S5)".

In section 3.2.4 of the revised manuscript Fig. S5 has been also referenced to explain the strongest diurnal variation of PM and BC concentrations. The text reads as follows:

"By contrast, in the colder seasons PM and BC concentrations showed strongest diurnal variation […] probably because MSC is located most of the day within the FT in the colder seasons, whereas PBL air mass is usually only advected to the site during the central hours of the day (Fig. S4 and S5)."

Moreover, this figure has been also used to explain the daily amplitude variation of N concentrations along the year. The text reads as follows:

"This seasonal variation could be associated with the fact that MSC is frequently in the FT in the colder seasons whereas in the warmer seasons it is more affected by the PBL air mass (Fig. S4, S5a and S5e), which increases N concentrations because biogenic emissions and photochemistry are enhanced by high temperature and high solar radiation intensity in the warmer seasons, favoring biogenic condensation and NPF processes, although other processes can not be discarded."

<u>Minor comments and suggestions</u>

Please find some other way to present the different N size ranges. The current method of having N>7nm is really meaningless, you can not compare N (in units of density) with a particle size (in units of length). Perhaps, putting the comparison value in subscript (as sometimes used in the literature) might be a better choice.

As suggested, the presentation of the different N size range (N>3nm, N>7nm and N>10nm) has been replaced for the lower size value in subscript (N_3, N_7 and N_{10}).

I would cut out the weekday variation part: the statistical test done by Barmet et al (2009) is poorly suited for weekday variation studies. See e.g. doi:10.1029/2012JD017574 . Overall, if the variation would be added to fig 12, I

think the differences would be too small to detect. Maybe a short mention could be enough; I do not think you need a section on this.

The authors acknowledge the limitations of the *Kruskal-Wallis* test, but as this is not a specific weekly cycle paper and the aim is not to identify a weekly pattern itself, the *Kruskal-Wallis* test is used only to look for statistically significant differences between two groups: Tuesday-Saturday and Sunday-Monday for MSC (Monday-Friday and Saturday-Sunday for MSY). Thus, the authors think that *Kruskal-Wallis* test is enough. Moreover, the authors would like to keep this section, because they found very interesting the differences found between MSC and MSY. Whereas the reduced human activity during the weekend is reflected on Sat and Sun at MSY, the effect is seen with one day of delay at MSC (so lower concentrations Sun and Mon), which confirms that MSC is located at a sufficient distance from anthropogenic emissions. BC is a clear anthropogenic component and showed a statistically significant difference between Tuesday-Saturday and Sunday-Monday groups at MSC.

Nevertheless, Fig. 11 in the new version of the manuscript has been changed to show only BC concentrations, and it has been modified to include the mean, and percentiles 5, 25, 75 and 95. The weekly cycles of PM_{1-10}, PM_1 and N concentrations with percentiles 5, 25, 75 and 95 have moved to supplementary material (Fig. S9).

The new figures are shown below:

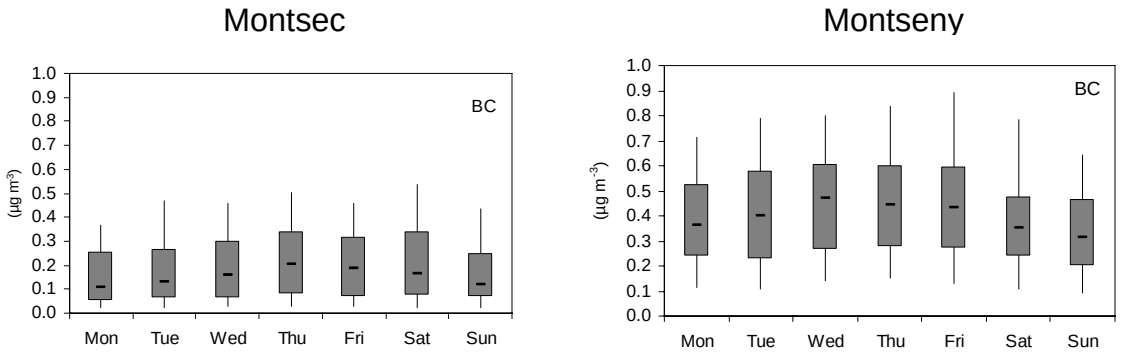


Fig. 11. Daily median (black line within the boxes) and percentiles (5-25-75-95, boxes and whiskers) of BC concentration during the study period at Montsec and Montseny.

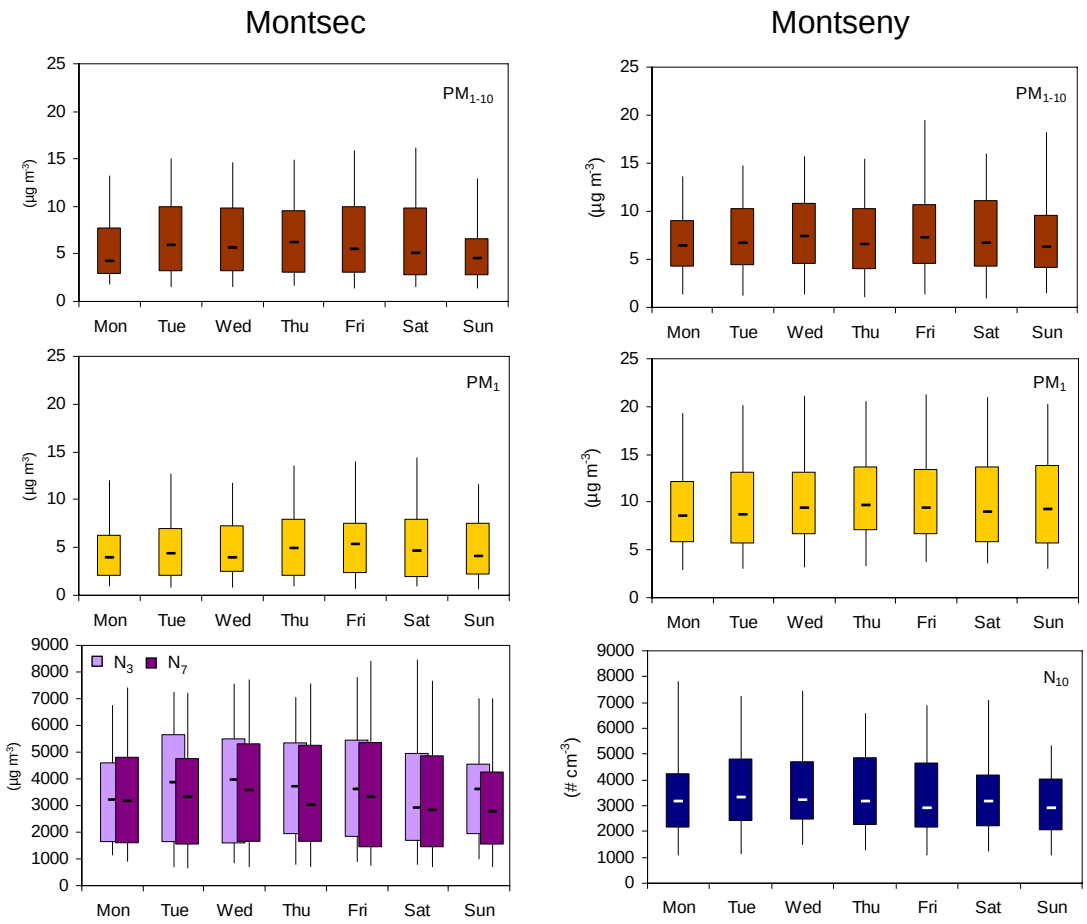
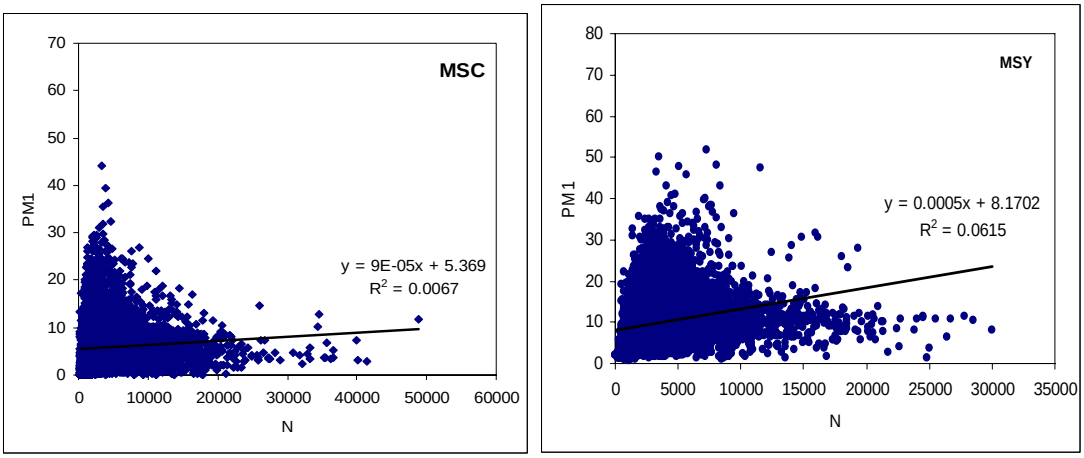


Fig. S9. Daily median (black line within the boxes) and percentiles (5-25-75-95, boxes and whiskers) of PM_{1-10}, PM_1 and N concentration during the study period at Montsec and Montseny.

The role of NPF is very weirdly attributed on this paper. Every increase of N is attributed to NPF, and specifically solar radiation. True, radiation plays a role, but not necessarily dominant part. I would be more interested to see the anticorrelation (or lack of..) between larger particles (e.g. PM1) and N, as this could be more indicative of another explanation of the behaviour: decrease in of sink term could start nucleation overall. Do you have SO2 measurements on-site? Overall think that role of radiation as the key element is not "confirmed" (27210, ln 2), is a possible explanation. Overall, please refrain of using very clear concluding remarks on issues which are not very clear, especially on these NPF issues, as you do not have even size segragation onsite.

The different time variation of PM and BC concentrations compared with that of N suggests that these aerosol parameters were governed by diverse factors. N concentrations depended more on local meteorological variables than on the air mass origin (transport). Moreover, N concentrations showed a marked diurnal cycle throughout the year with a peak at midday whereas PM and BC did not. For these reasons, the increase of N concentrations was more attributed to nearby origins and NPF processes than to a transport from the surrounding area.

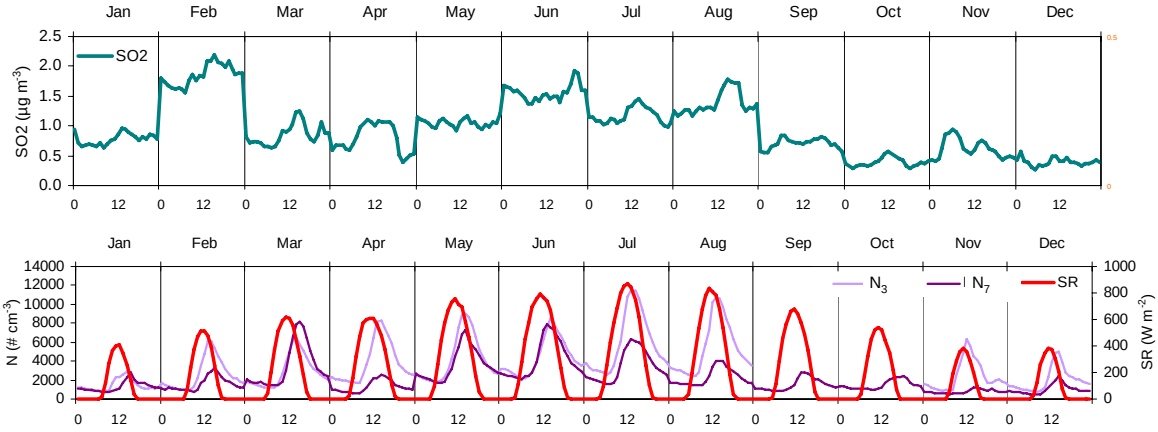
In addition, no correlation between N and PM_1 concentrations was observed, as it is showed in the following figures:



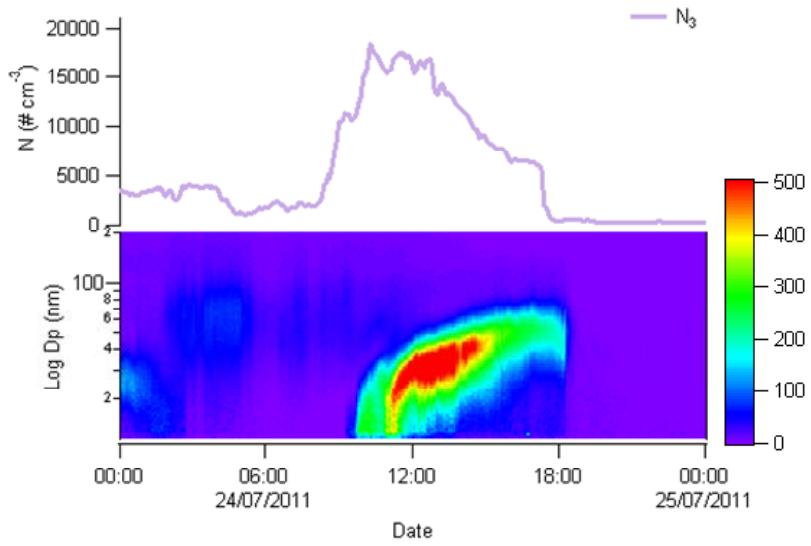
This information has been included in the text as follows:

"The lack of correlation between PM_1 and N concentrations observed at both sites (R^2=0.0067 and R^2=0.0615 at MSC and MSY, respectively) points to NPF processes occurring in-situ, rather than to the transport of very fine particles together with PM_1 mass."

Regarding SO_2 measurements, they are available but the results did not show a clear relation between N and SO_2, as shown in the following figures:



Size segregation data are available for a short period of time, since a SMPS was deployed at MSC for an intensive measurement campaign. These SMPS data confirmed that the increase of N at midday is attributed to the NPF processes, which is followed by growing of these new-formed particles to larger sizes, as shown in the following figure with data from 24th July 2011. Basically these results are in accordance with the findings by Cusack et al., 2013 at MSY. The most pronounce difference in the case of MSC is the recurrence and intensity of such NPF processes, which is probably linked, as the referee suggest, by the lower PM1 concentrations at MSC associated to breeze regimes than those at MSY under the same meteorological phenomenon. Nevertheless, these data are not shown in the present manuscript since the authors consider that they are out of the main scope of the paper and they will be part of future publications, where they will be studied in detail.



This information has been only included in section 3.2.4, when explaining the relevant role of NPF processes at midday at MSC. The text reads as follows:

"The higher N_3 concentrations at midday with respect to the N_7 concentrations and the agreement of N_3 concentrations with the number size distribution data from an intensive campaign, which showed the typical banana profile (ongoing studies), further confirmed the relevant role of nucleation episodes and particles growth processes at MSC."

Specific line points (first page, then line)

27202

5 sites do not register. Instruments do. Maybe "At MSC, the PM10 (..) and particle number concentrations for larger than 7 nm particles (N7) (..) were higher.." or something like that

6 which concentrations? Annual arithmetic means?

The text has been modified as follows:

"At MSC, PM_{10} (12±8 μg m^{-3}) and N_7 (2140±1542 # cm^{-3}) three-year arithmetic average concentrations were higher than those measured at other high altitude sites in central Europe for the same period (PM_{10}: 3-9 μg m^{-3} and N: 634-2070 # cm^{-3})"

15 air outbreaks

This has been included in the text as follows:

"PM and BC concentrations showed marked differences for different meteorological scenarios, with enhanced concentrations under North African air outbreaks"

17 sentence starting with "Because.." is quite awkward, rephrase

The sentence has been rephrased as follows:

"PM and BC concentrations increased in summer, with a secondary maximum in early spring, and were at their lowest in winter, due to the contrasting origin of the air masses in the warmer seasons (spring and summer) and in the colder seasons (autumn and winter)."

27203
first para: Again, all concentrations should indicate what they are, means? Annual?

The text has been modified as follows:
"Therefore, arithmetic averages as a function of meteorological episodes showed the highest concentrations of N during summer regional episodes (N_3: 4461 # cm^{-3} and N_7: 3021 # cm^{-3}) and the lowest concentrations during winter regional scenarios (N_3: 2496 # cm^{-3} and N_7: 1073 # cm^{-3})."

11 I am not sure that you can be sure of lower emissions (although they are very likely explanation. Use more careful way to say this

The sentence has been rephrased as follows:
"Our results highlight the importance of the NPF processes in southern Europe, underline the high contribution of long-range dust transport with respect to Central Europe and its prevalence in elevated layers, and reveal that MSC is much less affected by anthropogenic emissions than other high altitude sites in Central Europe."

27204
14 Inter alia should be before first on the list

This has been changed in the text as follows:
"Earlier studies at high altitude sites in Europe have been carried out inter alia in Switzerland (Jungfraujoch, 3578 m), France (Puy de Dôme, 1465 m), and Italy (Mt Cimone, 2165 m)."

22 Here you mention elevated emissions, but on the abstract you speak of low emissions compared to C.Eur.

In the abstract the low emissions referred to the low contribution of the anthropogenic emissions on BC concentrations at MSC compared to other high altitude sites in Central Europe (now slightly re-phrased as shown above), and here the elevated emissions refer to a WMB characteristic. The fact that the WMB is characterized by elevated anthropogenic emissions does not imply that MSC have to be more influenced by these emissions than how influenced are other high altitude sites in Central Europe by their surrounding anthropogenic emissions.

27205
21 Some fine resolution modellers might want more accurate location information

23 "Axial Pyrenees" ? This could be a correct term, I just have not hear d it before Here in general: Add information on the very local situation of the station: Is it next to a cliff? Are there wind obstructions?

Some more information about the local situation of the station is now given in the revised version as follows:

"This observatory is situated at the highest part of the Montsec d'Ares mountain, at an altitude of 1570 m.a.s.l. (42º3'N, 0º44'E), in a plain near to the edge of a 1000 m cliff to the S, with no wind obstructions present around."

27207
5 what was the MAC for your station?

For the period 2010-2012 the average MAC at MSC site was 9.1 m^2 g^{-1}, as shown in the Fig. 7 of the new version, but we preferred to use the instrument's default MAC because it is more used by the scientific community and this allow us to better compare our measurements with other BC measurements. This has been included in the text as follows:

"This comparison enabled us to determine the specific MAC for this site (9.1 m^2 g^{-1})."

Further information on the MAC at MSC is given by Pandolfi et al., 2014.

(Pandolfi, M., Ripoll, A., Querol, X., and Alastuey, A.: Climatology of aerosol optical properties and black carbon mass absorption cross section at a remote high altitude site in the Western Mediterranean Basin, Atmos. Chem. Phys. Discuss., 14, 3777-3814, doi:10.5194/acpd-14-3777-2014, 2014.)

8 the upper limit of 3772 is not so strictly 1 um. Perhaps just give the smaller end, as you will not see any of the larger particles over the small particle variation anyhow

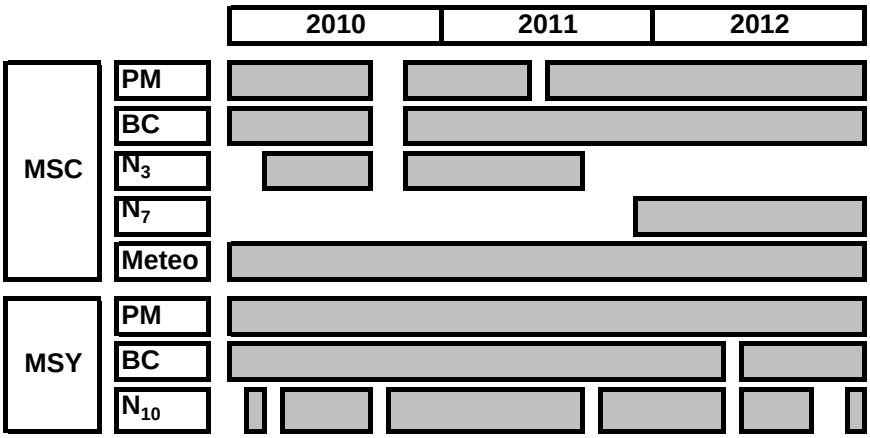
This has been changed in the text as follows:

"Measurements of particle number (N) concentrations were carried out using a low-size detection (detects particles with aerodynamic diameter (Dp) higher than 3 nm) condensation particle counter (TSI, CPC 3776) from March 2010 to August 2011, and an environmental (Dp>7 nm) particle counter (TSI, EPC 3783) from December 2011 to December 2012 (Fig. S1).

At MSY, concentrations of PM, BC and N were also measured from January 2010 to December 2012 (Fig. S1) using a GRIMM (model 180), a MAAP (model 5012) and a CPC (model 3772, Dp>10 nm), respectively."

9 add to S1 the times these were changed

Fig. S1 has been changed as follows:



24 This is important: Please indicate HOW this was actually done. By hand? It is completely ok, but then we would need a map (preferably in the main document) explaining the sectors, and which were the criteria (or any) for selecting one region from another

The classification of meteorological episodes was done based on the interpretation of backward trajectories, as described in the methods section, together with the information provided by the different tools explained in the methods section. It was not done in an automatic or programmed way, but it was done after visually examining the aforementioned information. This has been included in the text as follows:

"120-hours backward trajectories (for 12 a.m. modeling vertical velocity and for 3 different heights, 750, 1500 and 2500 m.a.g.l) were computed on each day of measurements, and interpreted and classified visually according to their predominant transport direction in: 1) Atlantic North (AN), 2) Atlantic North West (ANW), 3) Atlantic South West (ASW), 4) North Africa (NAF), 5) Mediterranean (MED), 6) Europe (EU), 7) Winter Regional (WREG, from November to April), and 8) Summer Regional (SREG, from May to October) (Fig. Sxx)."

Furthermore, a map has been included as supplementary material, Fig. S3:

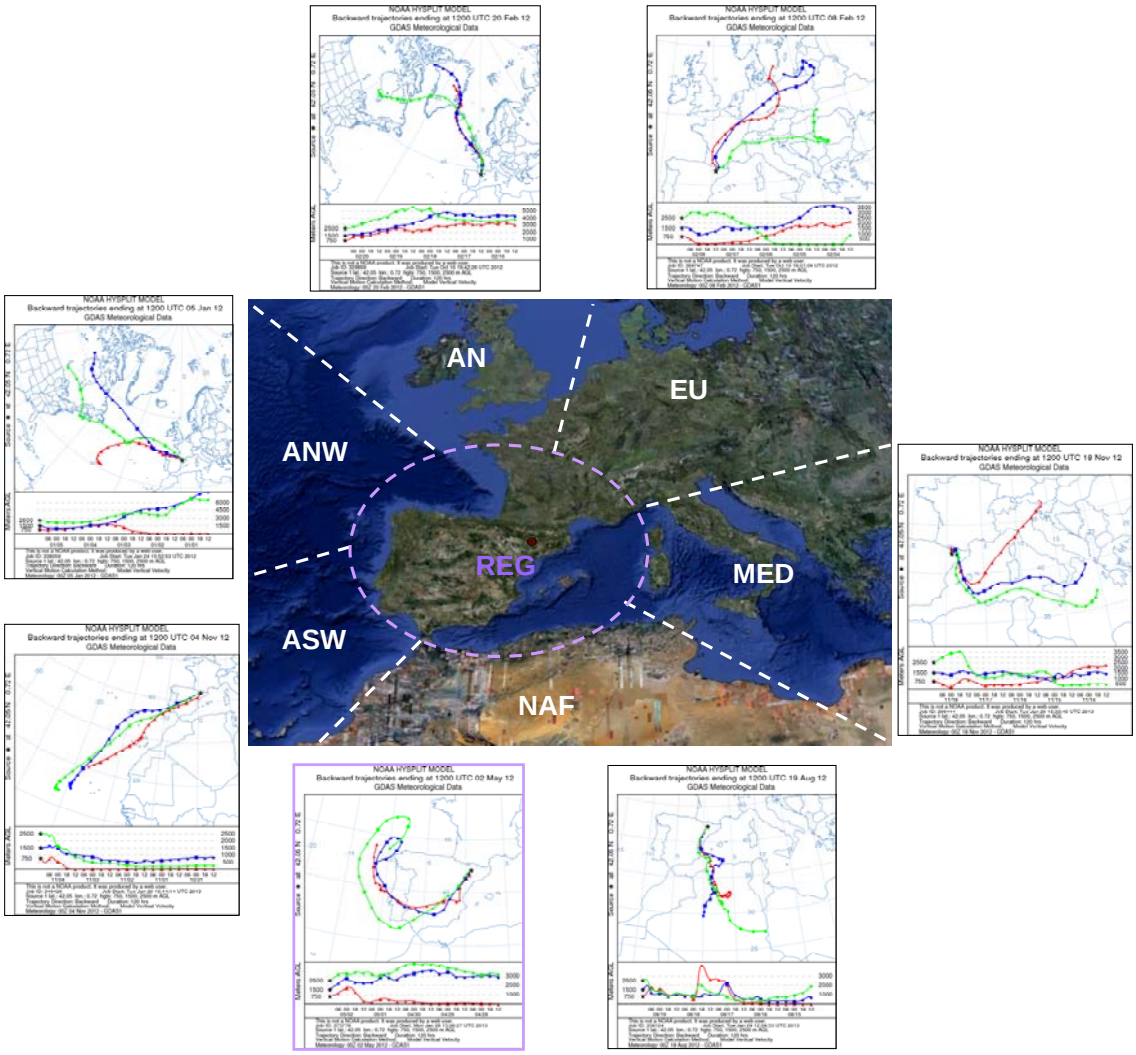


Fig. 3. Air mass origin sectors map and examples of backward trajectories for each sector according to their predominant transport direction.

27208

This long list of mean values (again, which means?) should be really moved to a table

As suggested, the list of averages has been moved to a table in the supplementary material, as follows:

Table S4. Three-year (2010-2012) arithmetic average concentrations of PM, BC and N at different high altitude and rural stations in Europe.

		PM_{10} ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)	PM_1 ($\mu g\ m^{-3}$)	BC ($\mu g\ m^{-3}$)	N_{10} (# cm^{-3})	N_7 (# cm^{-3})	N_3 (# cm^{-3})
Switzerland	Jungfraujoch (3578 m)	2.9	-	-	0.06*	634	-	-
	Rigi (1030 m)	8.0	7.5	5.8	-	-	-	-
	Chaumont (1137 m)	8.6	-	-	-	-	-	-
Italy	Mt. Cimone (2165 m)	8.8	-	-	0.33*	1847	-	-
Austria	Vorhegg (1020 m)	9.3	-	-	-	-	-	-
Germany	Schauinsland (1205 m)	9.3	7.3	-	0.38	-	-	-
	Schneefernerhaus (2650 m)	-	-	-	0.20	-	-	-
France	Puy de Dôme (1465 m)	-	-	-	0.22	2070	-	-
Spain	Campisábalos (1360 m)	10.3	5.1	-	-	-	-	-
	Risco Llano (1241 m)	11.5	5.8	-	-	-	-	-
	Montsec (1570 m)	11.9	8.2	5.3	0.19	-	2140	3716
	Zarra (885 m)	12.6	5.8	-	-	-	-	-
	Els Toms (470 m)	13.5	7.6	-	-	-	-	-
	Víznar (1265 m)	16.6	9.2	-	-	-	-	-
	Izaña (2373 m)	16.6	-	-	0.13	-	-	1467
	Cap de Creus (23 m)	16.8	7.9	-	-	-	-	-
	Montseny (720 m)	18.0	12.7	10.3	0.41	3475	-	-

Data from the ACTRIS Data Center web site.
* Jungfraujoch and Mt.Cimone BC concentrations averaged from 2007 to 2009.

27209

11 Here is an example of the "clear" comparison: please also indicate the variability of the two. Just comparing means could be biased, if there are outliers (quite common with optical measurements)

The standard deviation is now included in the revised version.

27211

2 "scenarios"? what is meant by this?

In this case “scenarios” is used as synonymous of “episode”.

19 Are you sure that these are the only two reasons? First you present the hypotheses of dust connected to BC (due refineries and BB etc), but then suggest more completely that the BC might just be an artifact. Which do you think is more reliable? Would the artifact effect also affect the fig 7?

The authors think that these are the two most likely and reliable reasons. Nevertheless, the sentence has been slightly changed as follows:

“The high BC concentrations observed during NAF episodes could be due to two reasons.”

Fig. 7 of the old version, shows dust and smoke surface concentrations from a model which is not affected by this artifact.

27212

5 What is ICP-AES? Are these data reserved for continuation paper? The measurement should anyhow be indentified somehow!

The chemical composition of PM will be the focus of forthcoming publications. The mineral matter determination is now explained in the Methodology section as follows:

"Mineral matter (MM) determination was done for the same filters than EC determination using an inductively coupled plasma atomic emission spectroscopy (IPC-AES), in order to better identify NAF episodes."

8 avoid "fine"

This has been changed in the text as follows:
"A good example was recorded on 28-29 June 2012 (Fig. 6)"

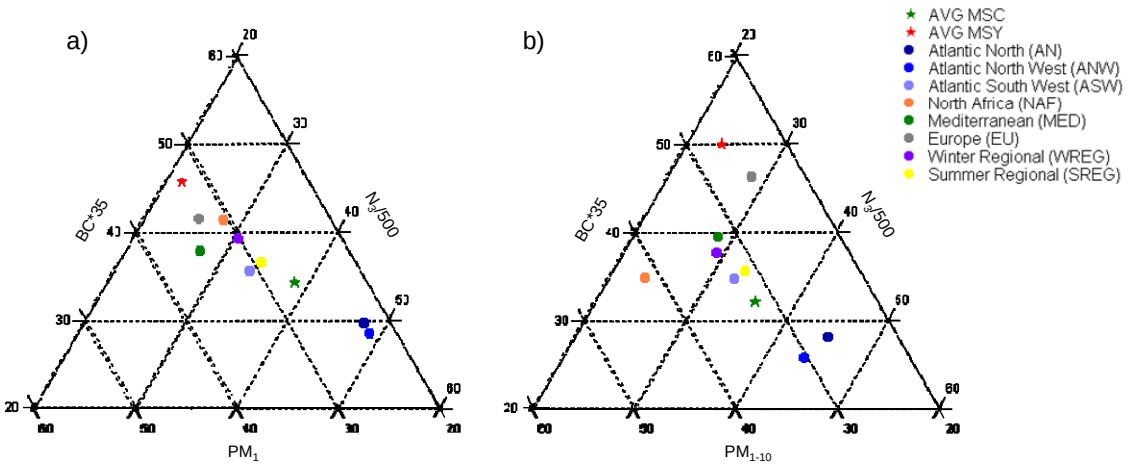
10- again, is this an artifact or not? Discussion!

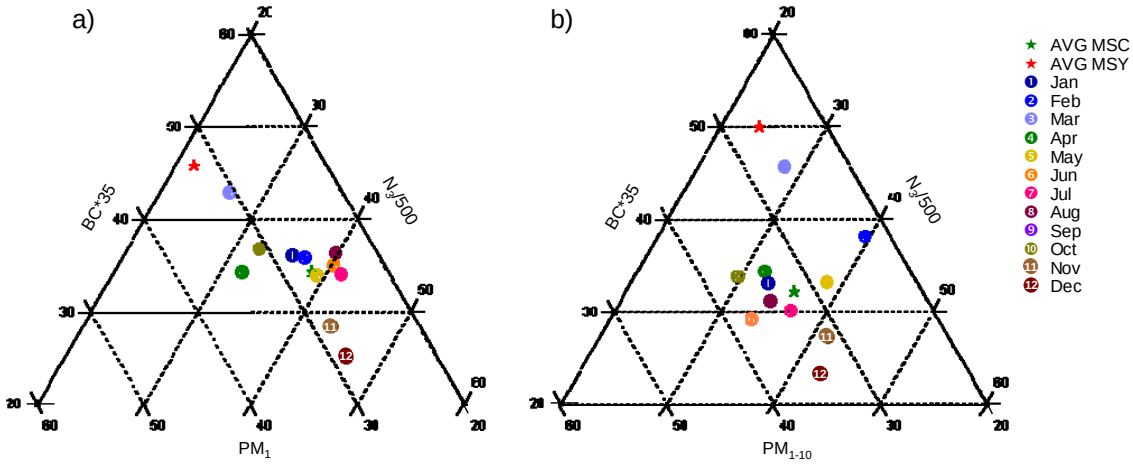
This is an artifact, but it is only evident when there is a pure Saharan dust episode, under mixed Saharan dust episodes it is hard to see it.

27213

Top paragraphs: Note my comment on Fig 9. The figures have merit, but need better scaling, and hopefully some indication of variability.

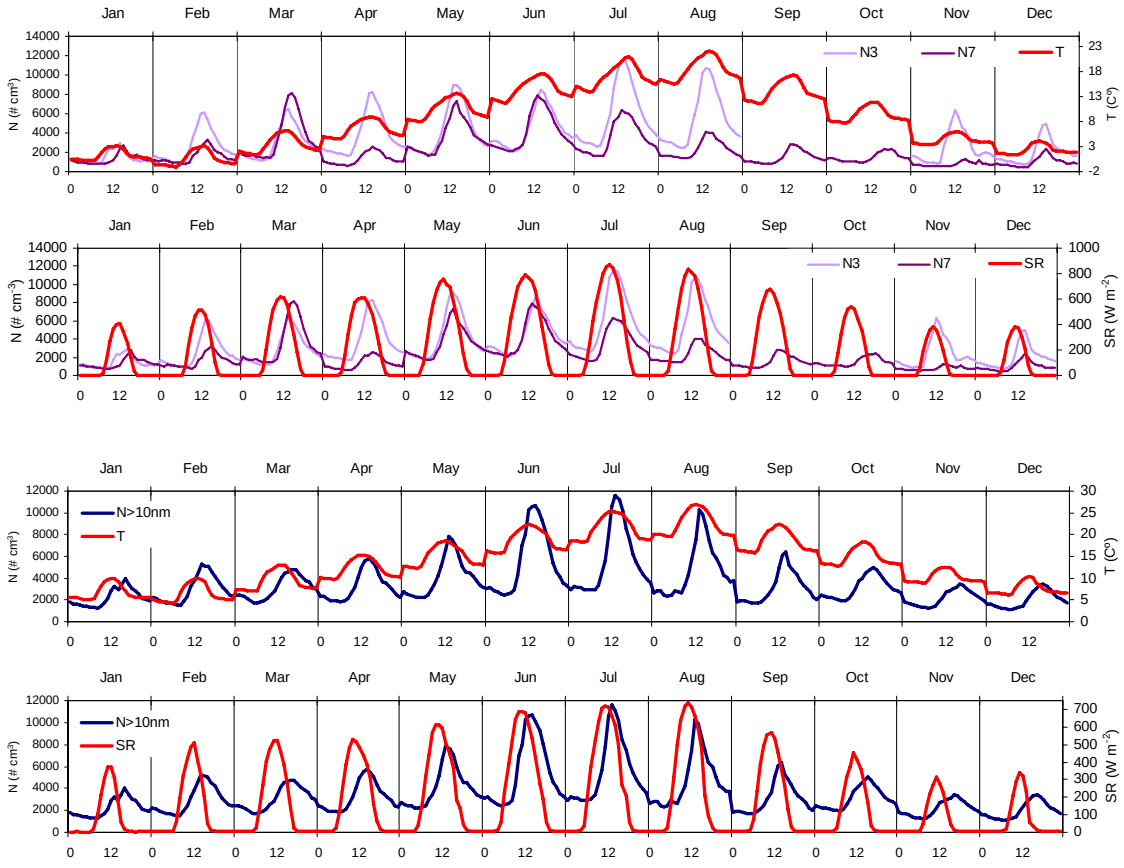
Fig. 8 and S8 of the new version of the manuscript have been changed with better scaling. However, indications of variability could not be done.





26 Higher solar radiation can affect isoprene emissions, but often aerosol mass properties are connected to monoterpenes, and thus temperature (see e.g. DOI: 10.1038/NGEO1800 and Guenther et al, JGR 100, D5, 8873-8892, 1995). Would the temperature be better explanatory variable than radiation?

Initially, solar radiation was chosen because of the more pronounced diurnal cycle compared to that of temperature. Nevertheless, after re-examining the data, it has been observed that N peak shows a higher correlation with temperature than with solar radiation. So, Fig. 12 and Fig. S10 have been changed to include the T variation as follows:



Moreover, the text has been modified to discuss these variations as follows:

"N concentrations had a markedly daily pattern throughout the year (Fig. 12c and d), with the highest concentrations between 12:00 and 16:00 UTC and the lowest at night and in the early morning. Although this increase in N concentrations takes place simultaneously with moderate southwesterlies (usually lower than 5 m s^{-1}) (Fig. 5d and 5e), it is not associated to transport but to local/regional processes governing N variation (happening during 12:00 to 16:00 mainly), since the daily pattern was independent of the synoptic conditions. This daily pattern is similar to that observed at other mountain sites such as Puy de Dôme (Venzac et al., 2009), where it was attributed to the height development of the PBL and to the higher frequency of new particle events at midday. In the present study, the diurnal cycle of N concentrations showed a very similar pattern than that of temperature and solar radiation intensity (Fig. 12c and d) since these parameters peak almost at the same time. Previous studies in the WMB found a dependency of monoterpenes biogenic emissions with temperature (Seco et al., 2011). Hence, the good correlation between temperature and N concentrations could be due to the condensation of the oxidation products of the biogenic volatile organic compounds (BVOCs), for example monoterpenes, onto small particles not detected by the CPC leading to grown particles above the lower size detection of the CPC. Furthermore, the diurnal patterns of temperature, solar radiation and N concentrations seem to indicate a better correlation between temperature and N than between solar radiation and N. This could point to a higher importance of the BVOCs condensation processes than of the NPF processes from photochemical oxidation (solar radiation) in N variation, which has been reported in recent studies (e.g. Paasonen et al., 2013)."

general: It could be good to discuss the relationship between PM and N. Do they show anticorrelation, no correlation or correlation? This could indicate the role of nucleation processes in N, and could give light how the processes affect the concentrations on different seasons.

It has been discussed in the present document of responses in the minor comments and suggestions section.

27215

7- Please avoid "confirm". I am not quite convinced yet, especially due the uncertainties and many assumptions of your hypotheses pathway.

This has been changed in the text as follows:

"The lack of correlation between PM_1 and N concentrations observed at both sites (R^2=0.0067 and R^2=0.0615 at MSC and MSY, respectively) points to NPF processes occurring in-situ, rather than to the transport of very fine particles together with PM_1 mass."

27218

1-3 Again: "Clearly related" is not very clear to me. I would say that there is a correlation, and one possible causative pathway could be this. There are many other properties which could be affecting the issue, and one correclation, especially as you do not even consider the time scales of the properties, is very dangerous to generalize as a relation.

This has been changed in the text as follows:

"Moreover, the diurnal cycle of N concentrations showed a very similar pattern than that of temperature and solar radiation intensity (Fig. 12c and d) since these parameters peak almost at the same time."

15-20 again: how about the temperature relation, instead of radiation? Or combination of both?

This has been included in the text as follows:

"This seasonal variation could be associated with the fact that MSC is frequently in the FT in the colder seasons whereas in the warmer seasons it is more affected by the PBL air mass (Fig. S4, S5a and S5e), which increases N concentrations because biogenic emissions and photochemistry are enhanced by high temperature and high solar radiation intensity in the warmer seasons, favoring biogenic condensation and NPF processes, although other processes can not be discard."

Conclusion part

I will not go over in detail: Just make sure that your concluding remarks are on the level which your data sets can be interpreted. Too many steps, and too many assumptions will not lead to clear indications.

Some changes have been done in the conclusion part of the revised version in order to better explain the concluding remarks.

Specific points on tables and figures

Fig 1. CI assumes that the profile is given according to the line on the map? Why that line?

The line is given to better understand the topography of the study area and to see the distance to Barcelona city. The epigraph in Fig. 1 has been changed to:

"Fig. 1. Top: location of the two monitoring stations (Montsec and Montseny). Bottom: topography of the Montsec area following the red line."

Fig 2. Please see my comments on the air mass origins in general (above). I would add here

The authors have included a map with air mass origin sectors in the supplementary material (Fig S3).

Fig 3. What is the order of the stations? It seems that they are ordered according to the concentrations. This is a little repetitive. Perhaps more useful order might be according to the height (a.s.l. or even better, from the surrounding land, if possible)

They are ordered according to the concentrations to help the reader. The height of each site is included in the x-axis label.

Fig 4. Add x-axis labels on all subfigures.

X-axis labels have been added to all subfigures.

Fig 6. the x-axis should be DATE. Please indicate the start of the Saharan airmasses coming.

X-axis label has been changed and start and end of Saharan dust episodes have been identified.

Fig 7. As this is not a modelling paper, and the results are not so much used, move this to supplementatry

Fig. 7 has been moved to supplementary material and now is Fig. S7.

Fig 8. Text does not indicate how the mineral matter concentration was acquired. Was the linear relation got with all the datapoints?

In the revised version of the manuscript the mineral matter determination method has been included as explained above. The points of the plot correspond to daily values. Absorption daily averages were calculated from the hourly values. EC and mineral matter concentrations were determined in daily filters, and hence the raw concentrations were daily concentrations. The legend of Figure 7 (old Fig. 8) has been modified as follows:

"Fig. 7. Measured absorption versus PM_{10} elemental carbon (EC) concentration as a function of PM_{10} mineral matter concentration during the study period at Montsec. Data points correspond to daily values."

Fig 9 and S4. I really like the idea of these plots. However, the presentation needs some adjustements: 1) scaling should be much improved. Now we have only very weirdly attributed acises on N and BC, and all of the values are in the middle as a small patch. It is very difficult to separate each. I suggest re-scaling, and considering using log-axis for N. 2) It is clear that this comes from some ready-made plotting tool, so this might be a little harder to do, but I suggest that you indicate somehow the overall variability on each direction. It might make, even better scaled, image hard to read though. Anyhow, please indicate FULL names of the regions in the figure, so that the reader does not need to check the text for the abbreviations. Or then, include a table for these.

This has been changed as indicated in the response to page 27213 top paragraphs.

Fig 12. As with my comments on the WKD effect, I would either remove this altogether or move it to supplementary.

As described above, PM and N weekly cycles have been moved to the supplementary material.

Fig 13. I do not support arithmetic averaging of N, and I am not sure of the distribution of solar radiation.

The authors think that using arithmetic average is a good option.

Figure S1. Please indicate more carefully the time periods. Also, the difference on the MSC N instrument could be directly indicated as well in the figure.

Fig. S1 has been changed as described above.

Tables S1, S2 and S3. Please indicate the type of mean used. This is especially interesting for WD. Also, the text has no indication to my browsing on the local topography. As WD measurements might be very dependent on local

In the revised version of the supplementary material the type of means has been included as follows:

Table S1. Arithmetic annual average of meteorological parameters at Montsec

Montsec d'Ares	T (ºC)	Tmax (ºC)	Tmin (ºC)	RH (%)	TotalPP* (mm)	WS** (m s^{-1})	WD** (degrees)	P (hPa)	SR (W m^{-2})
2007	8.6	28.3	-8.9	62	506	4.7	-	-	-
2008	7.9	27.2	-10.1	70	1186	4.3	-	-	-
2009	9	27.6	-9.8	66	639	5.8	297	843	-
2010	7.4	28.5	-12.4	69	755	4.4	293	846	189
2011	9.4	29.7	-9.9	65	597	4.3	247	852	198
2012	8.9	30.4	-13.5	59	640	4.9	312	851	203

*Annual accumulated precipitation
**Vector annual average

Table S2. Arithmetic seasonal average of meteorological parameters at Montsec during the study

Montsec d'Ares	T (ºC)	Tmax (ºC)	Tmin (ºC)	RH (%)	TotalPP* (mm)	WS** (m s^{-1})	WD** (degrees)	P (hPa)	SR (W m^{-2})
Spring	9.0	24.2	-3.7	69	215	4.4	218	850	242
Summer	17.2	30.4	3.5	57	57	3.8	203	854	288
Autumn	6.1	22.9	-8.6	72	119	4.9	319	848	121
Winter	1.7	15.4	-13.5	61	108	5.3	359	848	129

*Seasonal accumulated precipitation
**Vector seasonal average

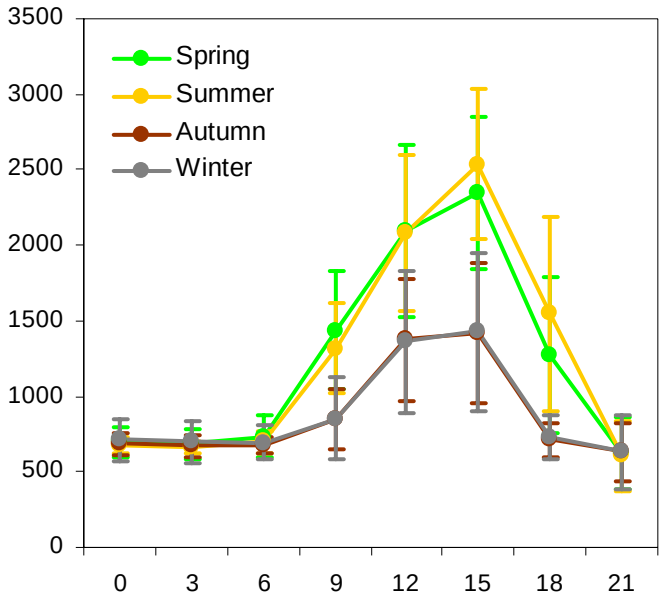
Table S3. Arithmetic average of meteorological parameters at Montsec as a function of air mass origin.

Montsec d'Ares	T (ºC)	Tmax (ºC)	Tmin (ºC)	RH (%)	TotalPP* (mm)	WS** (m s^{-1})	WD** (degrees)	P (hPa)	SR (W m^{-2})
AN	5.4	25.5	-11.1	57	31	4.7	16	850	190
ANW	9.1	28.1	-8.4	66	68	4.7	291	850	208
ASW	8.0	24.3	-5.0	77	90	4.6	233	847	133
NAF	14.4	30.4	-1.9	63	92	4.6	178	852	232
MED	7.4	20.3	-3.4	71	35	4.0	134	851	161
EU	3.7	22.1	-13.5	58	9	4.7	30	849	194
WREG	4.3	20.2	-5.8	78	24	4.3	295	848	111
SREG	14.9	27.4	0.4	66	66	4.0	189	852	249

*Accumulated precipitation
**Vector average

Figure S3. Again: What is the variability of the BLH on during the seasons?

The variability of the PBL height for each season is shown in the following figure. However, the authors preferred to show the diurnal variation for each month (Fig. S4).



REFEREE #2:

The authors present a rich dataset of aerosol observations at two elevated stations in northern Spain though, as stressed in the title, emphasis is put on a specific one. Data are presented and discussed in the light of main meteorological features and comparison with other high altitude station in Europe is proposed.

Overall my opinion agrees with Referee 1's to some extent and partly still refrains what observed in my first examination of this paper, confirming that the it is rather well written and organized, presents many elaborations but interpretation and some statements still looks at least a bit simplistic. Following the suggestions previously given, many technical details have been formally implemented in the present form though they are still treated very (too much?) synthetically. This is particularly true for basic statistics referring not only to correlation analysis with meteorological parameters as previously suggested (but elaborated in a non conclusive way as communicated in the authors' reply); statistical distribution (as pointed out also by Referee 1) of each parameter is missing either on the overall dataset or on a seasonal basis which would help to improve interpretation.

The authors acknowledge the reviewer for the detailed suggestions made to the manuscript. The response to each of the comments and suggestions is written below in a point by point manner. As for referee#1, we have done our best to include his/her suggestions in the revised manuscript.

Standard deviations of the three-year arithmetic averages have been included in the new version of the manuscript. Moreover, statistical distribution is already done with the analysis of air mass origin (Fig. 4) and seasonal variation (Fig.11) where the median and percentiles 5, 25, 75, 95 are shown.

The split of meteorological conditions according to air mass provenance makes sense only in part because seasonality is not esplicited, while it overlaps to or triggers inherent differences in aerosol behaviour.

The authors are aware of the limitations of the air mass origin analysis and for this reason the seasonality is partially included since regional episodes are differentiated for winter and summer. The occurrence of specific meteorological situations is inherent with seasonal variations. This is the case of winter anticyclonic situations and regional recirculation of air masses, characteristic of cold and warm seasons respectively. Likewise, at remote environments such as MSC, located in between Atlantic and Mediterranean influences, the origin of air masses is key to understand and interpret certain processes and aerosol concentration variability's. Whilst in the NW sector (Atlantic influence) there are no major sources of PM and BC, but important forested areas; E and SE are densely populated and industrialized and moderately forested; and the NE sector only account for European air mass origins. The interpretation of back-trajectories of air masses is enough to discriminate these provenances.

Going to some details in the paper analysis, I believe that the comparison described in 27209-lines 22-30 is quite hazardous, due to several reasons: time

interval for averaging, size range, elevation of the stations. Nucleation processes in fact might too be smoothed out either for the timing chosen (1 hour) or for the size limits of the available instrumentation especially in consideration of the nucleation mode and its tendency to develop below 10 nm. The instruments used in this paper have so different lowest size limits exactly in the range critical for the nucleation mode which might rise abruptly leading to extremely high levels of number densities potentially affecting in a significant way. Nucleation of BioVOC's is only a partial explanation of ultrafine particle formation, since other precursors (both organic and inorganic) are likely responsible for gas-to-particle conversion; within a thermal convective framework like that one projected for the Iberia peninsula affected by long lasting summer highs, significant pollutant uplift with "in transit" transformation (affected by trace gas concentration and RH%) is also highly expected.

As it is explained in the minor comments and suggestions section from Referee#1, the increase of N concentrations is attributed to nucleation processes due to its strong correlation with temperature and solar radiation rather than with the other parameters (PM and BC). Of course we cannot investigate in detail about the drivers of these NPF processes but, without doubts, biogenic emissions are playing an important role as demonstrated recently in different research studies. Now we modified the text to be less precise, as follows:

"This seasonal variation could be associated with the fact that MSC is frequently in the FT in the colder seasons whereas in the warmer seasons it is more affected by the PBL air mass (Fig. S4, S5a and S5e), which increases N concentrations because biogenic emissions and photochemistry are enhanced by high temperature and high solar radiation intensity in the warmer seasons, favoring biogenic condensation and NPF processes, although other processes can not be discard."

As concerns Saharan dust incursions and their effect on mass load and number density which are tentatively compared with other stations, each one with its unique characteristics, it must be recalled that the whole Mediterranean basin is affected by this natural source of aerosol; therefore at least for the southernmost stations elevation may not be a discriminant especially in the warm seasons when thermal convection efficiently ridistribute mineral dust 3Dimensionally (note the simultaneous cover of mineral dust on one of the four days NAAPS elaboration reported by the authors across Spain and northern Italy), while the effect north of Pirenees and Alps usually depends on intense events of mineral dust transport capable to cross the mountain barriers. Similarities and differences should be therefore managed with more sensitivity, so for the vertical distribution of mineral dust, as it is not demonstrated that most events mostly travel high. In fact as widely described in climatological investigations mineral dust source areas fluctuates in longitude and height of the lift up changing on an event and on a seasonal basis. Moreover the influence of long lasting pressure highs extending from Africa northward, a potential sign of climate change over the Mediterranean in terms of duration and areal extension, are likely responsible of intense convection (scarcely acknowledged in the text) with consequent mixing of dust upward.

There are various studies on dust transport that found a prevalence of long-range dust transport at higher altitude layers (e.g. VanCuren, 2002). In our study, the

calculation of mass load from PM_{10} attributed to African dust at MSC and MSY demonstrated that in the WMB Saharan dust plumes travel at various altitudes but they present higher dust concentrations at upper layers, as it is shown in the following figure:

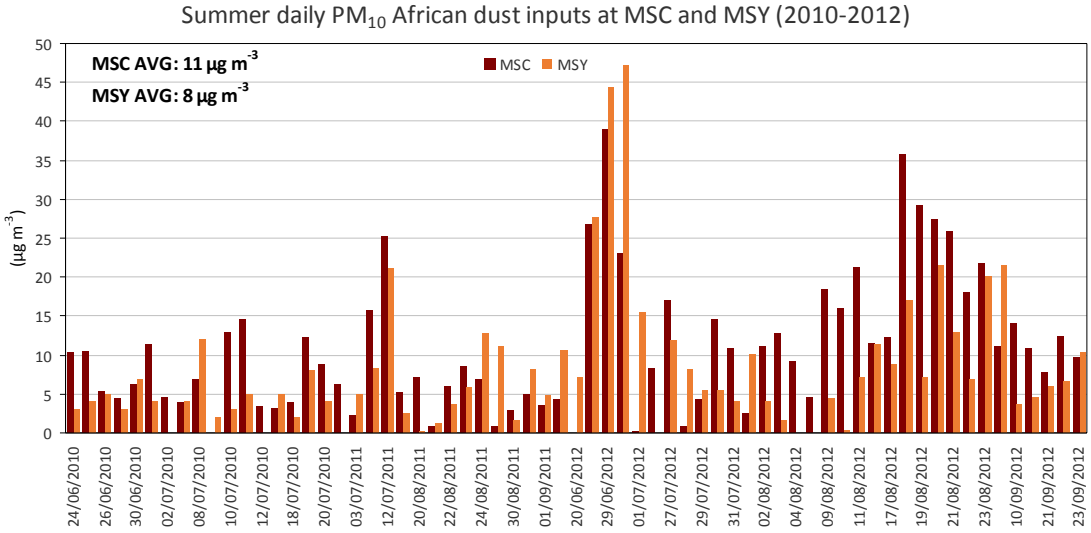
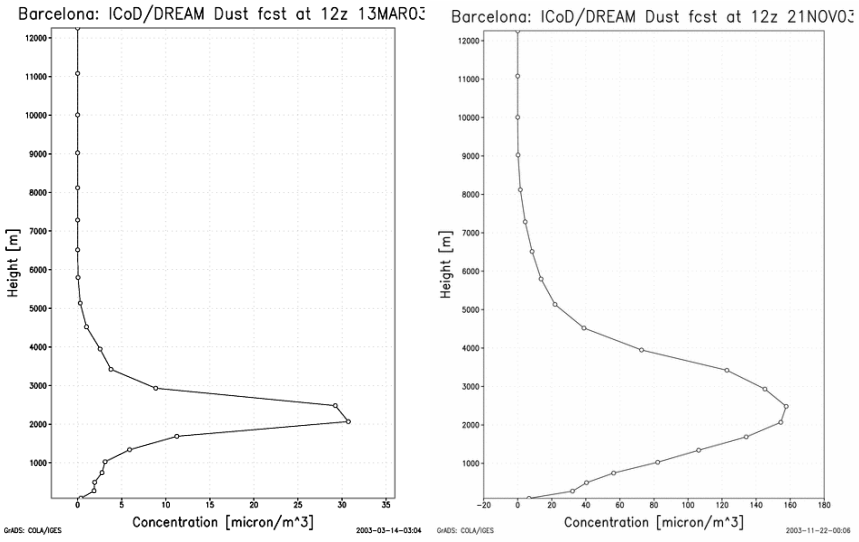


Fig. S6. Mass load from PM_{10} attributed to African dust in the warmer seasons and the three-year average at Montsec and Montseny.

This Fig. S6 has been added as supplementary material and has been referenced in the revised manuscript in section 3.2.1, when explaining the effect of meteorological episodes. The text reads as follows:

"The highest coarse PM mean concentration was observed at MSC under NAF influence (13 µg m^{-3}) which is in agreement with what was measured at MSY (Fig. 4b) (also 13 µg m^{-3}). Despite this similar average, calculation of mass load from PM_{10} attributed to African dust (Pey et al., 2013) at MSC and MSY in the warmer seasons (Fig. S6) showed that African dust plumes travel at various altitudes but they present higher dust concentrations at upper layers."

Moreover, DREAM-ICOD model during early 2000s gave vertical profiles showing the increase in dust concentrations with height, and therefore there could be a difference between MSY (720 m.a.s.l) and MSC (1570 m.a.s.l.). Examples shown below:



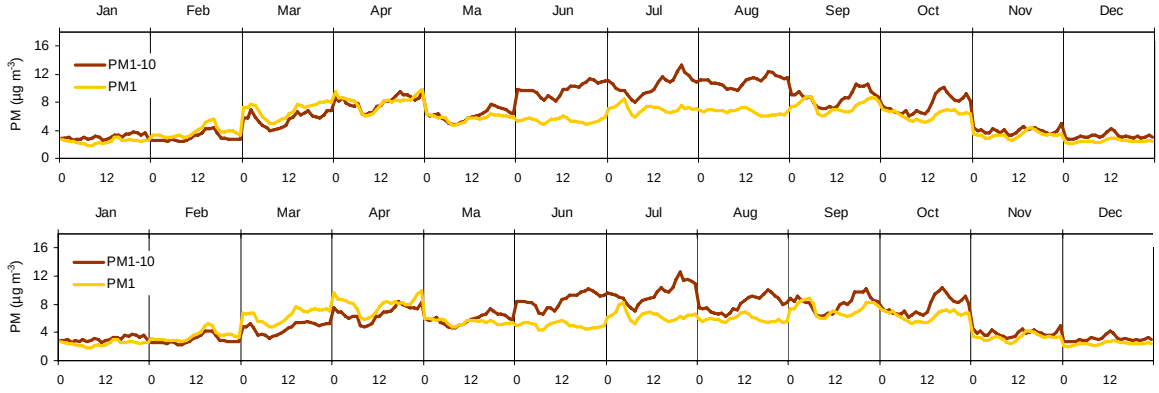
Daily variations: diagrams reveal to experts that in general the mountain valley breeze regime is active all the year round at least for some parameters, namely PM coarse and fine (possibly because more representative of aged aerosol constituents), but the behaviour of this daily periodicity in terms of the typical timing of maxima and minima is not esplicited . Usually maximum is found in the advanced afternoon. Please specify and give references.

This type of daily variations have been observed in other high-altitude sites. The explanation in the text reads as follows:

"By contrast, in the colder seasons PM and BC concentrations showed strongest diurnal variation, with a minimum at night and a maximum around 14:00-16:00 UTC […] Similar phenomena have been observed at Jungfraujoch (Baltensperger et al., 1997), Mt. Cimone (Marinoni et al., 2008), Himalayas (Marinoni et al., 2010), and Puy-de-Dôme (Freney et al., 2011)."

In addition "typical/average day" elaborations would preferably exclude saharan dust episodes from the averages; in fact they have been estimated to represent about 13% of all the air masses provenances, at least at Monsec (how about Monseni?), but they affect heavily the distribution of coarse particles in respect with fine ones, therefore it would be interesting to select/split data eliminating saharan dust.

The plots have been re-elaborated excluding the Saharan dust episodes. The figure below shows the average diurnal patterns including all the days (top) and excluding the Saharan dust days (bottom). The difference is soft in general, although the PM_{1-10} concentrations are slightly lower in March, April and August, when the majority of Saharan dust outbreaks occur. Nevertheless, for the sake of brevity, no more plots have been added to the main manuscript.



The position/timing (not always coincident for the several parameters) and shape (wide and noisy -possibly double- for the two PM parameters in the summer months, sharper and possibly anticipated for BC and N) of the maxima deserve some extra attention and at least a re-evaluation after data selection as suggested above.

The daily cycles for BC and N after excluding the Saharan dust episodes are similar to those elaborated including all the data. This was already expected since BC and N variations are not driven by the Saharan dust intrusions. Regarding the PM variations, as shown in the previous plots, the daily patterns do not change dramatically with or without Saharan dust intrusions. This is because the lack of a defined daily pattern at MSC is due to the combination of different phenomena, i.e. 1) the highest frequency of Saharan dust intrusions from North Africa (Fig. 2) associated with high concentrations of PM_{1-10} and BC during the day; 2) the widespread occurrence of wildfires around the WMB; 3) the relatively high frequency of European polluted episodes in spring (Fig. 2), both scenarios being linked to high concentrations of PM_1 and BC during the day and at night; and 4) the summer recirculation over the WMB, which creates a continuous increase in the background concentrations of PM and BC.