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Comment

## ***Interactive comment on “Three years of aerosol mass, black carbon and particle number concentrations at Montsec (southern Pyrenees, 1570 m a.s.l.)” by A. Ripoll et al.***

### **Anonymous Referee #1**

Received and published: 7 January 2014

Referee Comments Ripoli et al., ACPD, 13, 27201-

Please correct the title (without a tilde!)

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, con-

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nected 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.

### 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, is the lack of consideration of two key issues in the discussion and conclusions: The variability of the concentrations, and the 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 3 and 11, no indication of the role of the variability is 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 seem to be that e.g. N on the different years seem to follow similar patterns. This is especially difficult on the Fig 3 and associated discussion. If you can not get the comparison values

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for the other stations it is fine (although 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 inter-annual 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.

### 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.

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## Minor comments and suggestions

Please find some other way to present the different N size ranges. The current method of having  $N > 7\text{nm}$  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.

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 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 SO<sub>2</sub> measurements on-site? Overall think that role of radiation as the key element is not "confirmed" (27210, In 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 segregation on-site.

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?

15 air outbreaks

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

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

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

27204

14 Inter alia should be before first on the list

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

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 heard 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?

27207

5 what was the MAC for your station?

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

9 add to S1 the times these were changed

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

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

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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)

27211

2 "scenarios"? what is meant by this?

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?

27212

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

8 avoid "fine"

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

27213

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

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?

C10708

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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.

27215

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

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.

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

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.

Specific points on tables and figures

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

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

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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)

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

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

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

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

Fig 9 and S4. I really like the idea of these plots. However, the presentation needs some adjustments: 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 thre reader do not need to check the text for the abreviations. Or then, include a table for these.

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

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

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.

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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

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

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 27201, 2013.

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