

## ***Interactive comment on “Classification of clouds sampled at the puy de Dôme (France) from 10 yr monitoring: mean features of their physico-chemical properties” by L. Deguillaume et al.***

### **Anonymous Referee #1**

Received and published: 26 September 2013

The present manuscript describes 10 years of cloud chemistry observations at puy de Dome in France. It features an impressive dataset in terms of physical and chemical parameters measured and samples analyzed. Such large long term datasets of cloud chemistry are very rare and by themselves worthwhile additions to the scientific literature. The discussion of the results is very detailed but clear and organized in a logical way. Some results like the importance of hydroxyacetaldehyde are rather novel while other observations are more expected/traditional. Overall the study is an interesting contribution to atmospheric chemistry and I would support publication of the

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manuscript while encouraging the authors to consider the following comments.

The measurements span over 10 years, or other long term fog and cloud studies showed significant changes in chemistry over such timeframes (e.g. Aleksic et al., 2009). Now, sulfur reduction happened likely before in France but one might wonder if there are still changes ongoing and if those could impact observations. The authors might want to address somewhere the potential for a temporal change.

About the methodology, do the authors know if the technique used quantifies only free aldehydes or also aldehyde/sulfite adducts (e.g. HMSA for formaldehyde). This would be interesting to know as the aldehyde concentrations could be lower estimates if they do not include HMSA and other adducts.

The statistics methodology could benefit from some more details. May be the authors could specify if they really used straight PCA or may be some rotation? Or other transformation? Also it is not completely clear to this reviewer what a hierarchical classification is? Cluster analysis run by a statistics package? Finally it says that all data were included with 1 missing value maximum. How were the missing values treated?

One detail is also that while principal component analysis does not strictly require data normality, a very skewed data distribution can result in PCA artifacts. Some statement on how this is or is not an issue might be useful. For the least the term correlation in the context of PCA should be changed. I commend the authors though for their approach to other correlation tests because of the non normality.

Linking the differentiation between “marine” and “highly marine” to an elevated pH is a little debatable especially if one sees in the table that as the range of pH stretches higher in the marine category and one wonders if there is really a difference between the two in pH?

The comparisons to other studies are interesting. It should be noted though that some-

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times different timeframes are compared (cf also comment on evolution), e.g. a pH of 2.4 at Whiteface Mountain is cited but this was in the 1990s.. recent work shows pH >one unit higher at this site (Aleksic et al., 2009).. so the difference looks less substantial if similar timeframes are being compared. On the other hand one wonders why the study does not compare itself then against the old French studies in the Vosges Mountains (e.g. Lammel and Metzger in 1991 and Herckes et al 2002).

Details:

P22808 TOC discussion. "Remote" or background samples can also have high TOC because of local biogenic emissions e.g. Whistler in (Ervens et al., 2013). Hence may be mentioning of the season and/or lack of vegetation at pdD might be useful?

In the text citations: may be cite multiple references in chronological order not random

The paper could benefit from some language editing.

P22797L20 replace interactions with interaction.

P22803L5 "." Not in right spot?

P22804L27 and throughout the text, please use only significant digits not 50.41% for a PC...

P22807L25 "reducing" not "reducer"?

P22810L20: please provide a reference for "some studies"

P22813L21 "to our point of view" sounds odd. . .

P22816L9 replace "underlined" by "highlighted"?

P22818L13 replace "followed" by "investigated"?

P22818L16 may be replace "frame" with "framework"?

Table1 and 2: please use only significant digits, e.g. Na<sup>+</sup> concentration of 679 not

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678.6

Table 3. if possible, please provide a number <X rather than BDL

Figure 1: could you please put the fonts larger for the elements

Supplemental info:

Please also here use only significant digits in the tables. (e.g. 2193.00 looks odd)

Please check formatting: some boxes didn't seem to have lines. Some boxes were shaded while others not. The references were not uniform (e.g. some with doi some not)

References cited here:

N. Aleksic, K. Roy, G. Sistla, J. Dukett, N. Houck and P. Casson, Analysis of cloud and precipitation chemistry at Whiteface Mountain, NY, Atmospheric Environment 43 (2009) 2709–2716

P. Herckes, R. Wendling, N. Sauret, Ph. Mirabel and H. Wortham, Cloudwater studies at a high elevation site in the Vosges Mountains (France), Environmental Pollution, 117 (2002) 169-177

G. Lammel and G. Metzger, Multiphase chemistry of orographic clouds: Observations at subalpine mountain stations, Fresenius J. Anal. Chem. 340 (1991) 564 - 574

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