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Atmospheric Chemistry and Physics Discussions

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 #2

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The authors present the results of 10 years of measurements of chemical concentrations (including inorganic and some organic species) and supporting physical quantities made at puy de Dôme. The data are valuable and should be published. Detailed comments follow.

1. Between the abstract and conclusions, the paper reads like a good scientific publication. However, the conclusions and the abstract are devoid of the many results discussed in the paper. Instead there is some speculative discussion and opinion. I

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don't understand what the authors intend here, but the abstract and conclusions need to be made consistent with the rest of the paper.

2. Abstract – line 12 – I can't find a discussion of 'standard chemical scenarios' anywhere in the text. What do you mean by this, and how do you justify such a term in the abstract without any discussion elsewhere in the paper?

3. Page 4, line 17 – why only "degradation" and not production?

4. Page 5, line 5 and lines 15-16 – "natural background site" implies a site that measures air free of local and regional anthropogenic influences. You demonstrate that is not the case for pdD. The same reasoning applies to the reference to of using these measurements as a benchmark for "natural free tropospheric conditions". How do you derive such conditions from your dataset?

5. Page 6, lines 8-13 – how is the liquid or ice transferred from the impaction plate?

6. Page 6, lines 20-24 – how were the LWC and the TOC calibrated?

7. Page 9, lines 19-22 – How are these criteria used to "further" classify cloud events, when they are derived from the PCA analysis already used to classify the events? Also, I don't understand the context of the mention of the 57 samples. Presumably the values in Table 1 are from the 138, and so what is the purpose of the discussion of the other 57?

8. Page 13, lines 5-6 – add N2O5 to the bracketed trace gases. Also "concentration" on line 6 should be concentrations.

9. Page 13, lines 8-9 – I assume that you mean something to the effect that "The relatively high correlations suggest that the contributions to the ammonium, nitrate and sulfate ion concentrations are larger from nucleation scavenging than from gas scavenging." Perhaps, but what about the relative contributions seen in the dry aerosol at pdD over the years; e.g. does it show that much more NH4NO3 in polluted air during the daytime? Nitrate is frequently and in many places evident in higher concentrations

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in cloud water samples relative to sulphate than in dry aerosol samples. That argues for a predominantly gas-phase source (HNO3 in the daytime; N2O5 in the night-time), and HNO3 can also be correlated to sulphate.

10. Page 14, lines 1-3 – The statement "for most of the values reported elsewhere in the world" is incorrect. See Leaitch et al. (JGR, 1986 and JGR, 1988), and there are other more recent observations that also show increased nitrate in cloud water relative to sulphate. Lines 10-14 – Also, see Leaitch et al. JGR 1986 for another discussion of calcium in cloud water samples.

11. Page 15, lines 12-14 – replace "On the opposite. . ." with "In contrast, the polluted air masses exhibit much higher concentrations of nitrate in the cloud water and the cloud water samples in highly marine air are characterized by relatively high levels of chloride and sodium." (assuming that is what you mean.)

12. Page 15, line 22 – in the context of air mass, what do you mean by origins?

13. Page 15, lines 23-24 – how do you know they are acids and not salts?

14. Page 16, lines 7-9 – A reference for this statement: Sorooshian, A., et al. (2006), Oxalic acid in clear and cloudy atmospheres: Analysis of data from International Consortium for Atmospheric Research on Transport and Transformation 2004, J. Geophys. Res., 111, D23S45, doi:10.1029/2005JD006880.

15. Page 18, lines 13-15 – Li S.-M., A. M. Macdonald, A. Leithead, W. R. Leaitch, W. Gong, K. G. Anlauf, D. Toom-Sauntry, K. Hayden, J. Bottenheim, D. Wang (2008), Investigation of carbonyls in cloudwater during ICARTT, J. Geophys. Res., 113, D17206, doi:10.1029/2007JD009364

16. Since you are characterizing bulk samples of cloud water, there should be some discussion of the potential differences of cloud water chemistry for different sizes of droplets within a cloud. For example, your continental and marine clouds may contain more sulphate in smaller droplets and more sodium in larger droplets, whereas nitrate

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may cover a broader range of droplet sizes if it comes from the gas phase. You reference work by Moore et al for example, which is one source you could draw on for this.

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