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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Long-term observations of positive cluster ion concentration, sources and sinks at the high altitude site of the Puy de Dôme" *by* C. Rose et al.

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We thank referee N°3 for his comments and remarks which contributed to improve and clarify the present paper. Our answers to the suggestions are listed below.

Comment 1: I agree that both polarities should be added to the study, and when the ion spectrometer is fully operating there is no scientific reason to neglect the other polarity. I understand that here the reasoning was that they wanted to focus on just positive polarity, as in the negative cluster ion concentrations they didn't observe a clear seasonal behaviors especially during the NPF events. This is not totally true as

Fig. 2 shows a diurnal cycle for the negative cluster ions.

Reply 1: It is true that when considering the annual variation of the negative cluster ions concentration we do see a variation, which is not exactly the same as the one observed for positive ions. However, the main purpose of this paper was to investigate the differences between event and non-event days. Since the most significant differences were seen for the positive polarity, we chose to focus exclusively on the positive ions behavior, which seemed to be more interesting regarding the formation of new particles at the Puy de Dôme.

Comment 2: In Fig. 3, at winter the positive ions had a minimum during noon in cluster ion concentrations. Was this seen also in the negative polarity? Could it be coupled with rain or snow episodes? Does it rain at PDD site? During rain and snowing the negative intermediate ion concentration increases and usually a small decrease is seen in the cluster sizes. See the paper by Tammet, Hõrrak and Kulmala (ACP 2009).

Reply 2: It is true that, even if no clear diurnal variation is observed, it is eventually possible to distinguish a minimum in the median positive cluster ion concentration around noon on non-event days in winter. For negative small ions, the diurnal variation is even less pronounced but still, minimum values are again obtained around noon. Rain episodes are rarely observed at the Puy de Dôme, and they are not recorded, which does not ease an investigation between rain episodes and cluster ion concentration. However, we do believe that rain events are associated with cloudy conditions, which should have been removed in the analysis. Moreover, there is no reason for the rain to have an impact on ion concentration exclusively around noon. Concerning snow episodes, they are of course numerous in winter, but once again they are often coupled with in cloud conditions at the station and they can occur at any time during the day. Moreover, it is likely that the role of snow still remain unclear since cluster and intermediate ion concentrations were found to be increased during snow storms (Vana et al., 2006; Virkkula et al., 2007).

Comment 3: Atmospheric (di)electrode effect is not mentioned in the paper. The effect causes the depletion of negative ions near the ground. Thus, higher positive ion concentrations should be measured close to earth (effects works up to few meters from earth's surface) as the ground works as negative electrode attracting more positive ions.

Reply 3: The electrode effect of the atmosphere was not mentioned in the manuscript but is of course very interesting to discuss since measurements at the Puy de Dôme contrast with the expected observation. Thus, a more complete discussion on the observed concentrations as well as on the electrode effect was included in Section 3.2: "At the Junfraujoch station, lower negative cluster ion concentrations were also reported by Vana et al. (2006). According to the authors, lower pressure conditions at high altitude increased the mobility of cluster ions, which could eventually exclude the smallest (especially negative) from the measurement range of the instrument. The fact that at the Puy de Dôme negative ions are, with the exception of winter, slightly more numerous than positive ones contrasts with what would be expected from the atmospheric electrode effect. Indeed, this effect predicts that in calm air, higher positive cluster ion concentrations should be detected near the ground and up to a few meters because of the negative charging of the Earth (Hoppel et al., 1986). At the Puy de Dôme, horizontal and vertical winds from the valley are often observed. This turbulence mixes the air, which may hide or suppress the effect of electric field, and thus partly explain why positive ions are not found to be predominant."

Comment 4: In the introduction section, there should be added few words about the importance of the ions for the atmosphere and the climate. See review by Hirsikko et al. (ACP 2011).

Reply 4: A few words were added on the global role of atmospheric ions: "Air ions are carriers of electrical current in the atmosphere. The air ion population is commonly divided into small or cluster, intermediate and large ions". Moreover, potential connection between GCR, aerosol and clouds was also discussed: "A connection between

GCR, aerosols and clouds through the nucleation process was proposed by Dickinson (1975): sulphate aerosol particles forming from ions produced by GCR might grow to CCN sizes and form cloud droplets. In that case, the variation of GCR ionization over the solar cycle would have a direct impact on cloud properties (lifetime, albedo) and thus on the radiative forcing of the Earth. However, more recent studies did not find significant support for a correlation between the variation in GCR ionization and low cloud cover Sloan and Wolfendale (2008)". Also the relevant paper by Hirsikko et al. (2011) was mentioned.

Comment 5: Add more clear definitions to the text, starting with instrument measured and when, and whether the measurement set-up and locations was exactly the same during the whole time. Also more technical issues like how to convert the mobility mobility equivalent Millikan diameter, and what kind of corrections was done to take into account the high altitude measurement site. It's important to add more information to figure and table captions: when the data what collected and which size range is under inspection.

Reply 5: It is true that we should have been more precise on the instrumental setup which has changed during the measurement period. Indeed, three different instruments were used and the location of the instrument also changed one time, and only the second location was mentioned in the article. However, it is important to note that no discontinuities in the measurement where detected when changing the instrument or when moving it from one place to the other. The general description of the instrumental setup was finally changed to:" During the whole measurement period, three different instruments were used, being AIS 7, NAIS 3 and NAIS 13. From February 2007 to the end of 2010, the instrument was operating in a shelter, with a short inlet (length 30cm, inner diameter 3 cm) sampling approximately 2 m high from the ground. At the end of 2010, the instrument moved on the roof of a new shelter located closed to the old one, sampling 11m high from the ground with the same individual non-heated short inlet. For the two different measurement setups, one should note that measured ion size distributions were directly influenced by the presence of a cloud". When converting the measured mobility to the particle diameter, we refer to mobility diameter, i.e. Millikan diameter. The corresponding reference, Mäkelä et al. (1996) is now mentioned in the introduction as well as in Section 2.2. Beside the fact that pressure (around 860hPa) is considered when converting mobilities into diameters, no special correction was done to take into account the altitude of the measurement site. We found no evidence in the literature for systematic additional corrections that should be done. Especially, the work by Manninen et al. (2010) reports NAIS measurements from high altitude stations, sometimes higher than the Puy de Dôme (Jungfraujoch) but no kind of correction is mentioned. The papers by Boulon et al. (2010, 2011) also report measurements from high altitude sites, without correcting the data for altitude. Efforts have been put to be more precise in the table headings as well as in the figure captions.

Comment 6: One more interesting question is that how did the size distributions look like for the positive and negative ions? In the paper (p. 14941, line 26) Rose et al. says "Indeed the measured small ion concentrations could increase on event days, not because the concentration of the cluster ion mode is changing but only because the fraction detected by the instrument is larger since ions are getting bigger." Does this mean that the reason why you observe smaller number for the negative cluster ions is that negative ions are more mobile (also smaller in mobility diameter) are thus likely lower range of the cluster band out of the scope of the instrument?

Reply 6: If not to take into account the values of the concentrations, the shapes of the positive and negative cluster ions size distributions which are now showed in Fig 5 are representative of what is on average observed at the Puy de Dôme. Negative ions are smaller than positive ones but the maximum of the mode is always visible, being far enough from the lower size detection limit of the instrument to ensure that most of the negative clusters are detected. Moreover, since negative cluster ion concentrations are on average higher (and not lower, see Table 2 and 3) than positive ones at the Puy de Dôme, we do believe that negative clusters are measured efficiently. But, in fact, the

smaller size of negative cluster ions could have been a good explanation in case of lower concentrations, as suggested by Vana et al. (2006).

References

Boulon, J., Sellegri, K., Venzac, H., Picard, D., Weingartner, E., Wehrle, G., Collaud Coen, M., Bütikofer, R., Flückiger, E., Baltensperger, U. and Laj, P.: New particle formation and ultrafine charged aerosol climatology at a high altitude site in the Alps (Jungfraujoch, 3580 m a.s.l., Switzerland), Atmos. Chem. Phys., 10, 9333–9349, doi:10.5194/acp-10-9333-2010, 2010.

Boulon, J., Sellegri, K., Hervo, M., Picard, D., Pichon, J.-M., Fréville, P. and Laj, P.: Investigation of nucleation events vertical extent: a long term study at two different altitude sites, Atmos. Chem. Phys., 11(12), 5625–5639, doi:10.5194/acp-11-5625-2011, 2011. Dickinson, R. E.: Solar variability and the lower atmosphere, Bulletin of the American Meteorological Society, 56(12), 1240–1248, 1975.

Hirsikko, A., Nieminen, T., Gagné, S., Lehtipalo, K., Manninen, H. E., Ehn, M., Hõrrak, U., Kerminen, V.-M., Laakso, L., McMurry, P. H., Mirme, A., Mirme, S., Petäjä, T., Tammet, H., Vakkari, V., Vana, M., and Kulmala, M.: Atmospheric ions and nucleation: a review of observations, Atmos. Chem. Phys., 11(2), 767–798, doi:10.5194/acp-11-767-2011, 2011.

Hoppel, W. A., Anderson, R. V. and Willett, J. C.: Atmospheric electricity in the planetary boundary layer, The Earth's Electrical Environment, 149–165, 1986. Mäkelä, J. M., Riihelä, M., Ukkonen, A., Jokinen, V. and Keskinen, J.: Comparison of mobility equivalent diameter with KelvinâĂŘThomson diameter using ion mobility data, J. Chem. Phys., 105, 1562–1571, doi:10.1063/1.472017, 1996.

Manninen, H. E., Nieminen, T., Asmi, E., Gagné, S., Häkkinen, S., Lehtipalo, K., Aalto, P., Vana, M., Mirme, A., Mirme, S., Hõrrak, U., et al.: EUCAARI ion spectrometer measurements at 12 European sites – analysis of new particle formation events, Atmos.

Chem. Phys., 10(16), 7907–7927, doi:10.5194/acp-10-7907-2010, 2010.

Sloan, T. and Wolfendale, A. W.: Testing the proposed causal link between cosmic rays and cloud cover, Environmental Research Letters, 3(2), 024001, 2008.

Vana, M., Hirsikko, A., Tammet, E., Aalto, P., Kulmala, M., Verheggen, B., Cozic, J., Weingartner, E. and Baltensperger, U.: Characteristics of air ions and aerosol articlesat the high alpine research station Jungfraujoch, Proceedings of 7-th International Aerosol Conference, the American Association for Aerosol Research (AAAR), ISBN 978-0-9788735-0-9, 1427, 2006.

Virkkula, A., Hirsikko, A., Vana, M., Aalto, P. P., Hillamo, R. and Kulmala, M.: Charged particle size distributions and analysis of particle formation events at the Finnish Antarctic research station Aboa, Boreal Environ. Res., 12(3), 397–408, 2007.

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