

Interactive comment on “New particle formation and ultrafine charged aerosol climatology at a high altitude site in the Alps (Jungfrauoch, 3580 m a.s.l., Switzerland)” by J. Boulon et al.

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General recommendation

The authors investigated new-particle formation events using specific instrument with extremely low detection limit also for the neutral particles. Therefore, more or less direct detection of newly formed particles was possible. What makes this study to stand out is that the high altitude site Jungfrauoch allows new-particle formation studies both in boundary layer and free troposphere conditions. I recommend publication in ACP. Nevertheless, there are some issues to be resolved, mainly related to some corrections and providing more details, which help understanding the methods and the results.

C4343

Specific comments

1. p. 11364, section 2.1: The instrument is called Neutraal cluster and Air ion Spectrometer (NAIS) according to current practice (e.g. Manninen et al. BER 2009, ACPD 2010; Mirme et al. ACP 2010; Paasonen et al. ACPD 2010; Wehner et al. ACP 2010). Also check p. 11378, line 23.
2. p. 11365, lines 1-4: Why did you choose to use Tammet's mass diameter instead of Millikan-Stokes mobility diameter as you mentioned in the text? Nowadays, with the ion spectrometer data Millikan mobility diameter is commonly used (see e.g. Mäkelä et al. JCP 1996). This can cause some difference in results when comparing nucleation parameters calculated for the same size range but in Millikan diameter. What were the mean local pressure and temperature?
3. p. 11365, line 7: The Ions are not filtered away with the prefilter in particle mode before the charging of the sample. When the NAIS measures in particle mode the prefilter and precharger are both turned off, whereas the main charger and the post-filter are active. Therefore, in particle mode both the neutral and the charged particles are measured. To avoid this misunderstanding I prefer to call the spectra measured in particle mode as total particle number size distribution. This should be kept in mind also when doing the data analysis (neutrals = (total particles - [neg ions + pos ions])). In alternative mode the precharger is used together with the main charger and the post-filter.
4. Section 2.1, p. 11365: Could you tell something about the calibration of the instrument and the data quality checks?
5. Section 2.2.2, p. 11366: The tree first sentences are unclear and needs rephrasing. Usually, the particle formation processes (like activation or nucleation) are followed by the growth.
6. Section 2.2.3, p. 11367: How was the coagulation sink calculated? Why did you

C4344

choose to use both the NAIS and the SMPS data? The SMPS data is much more reliable in the large particle sizes because the NAIS does not take into account multiple charging of the particles (see Manninen et al. BER 2009).

7. Section 3.2.2, p. 113673: How was the condensation sink calculated? The units are wrong ($\text{cm}^{-3} \text{s}^{-1}$ should be s^{-1}).

8. Section 3.3: How comparable are the different methods to calculate e.g. growth and formation rates? Could you say something about the error limits of your calculations? What could explain the difference in results between this study and study by Manninen et al. ACPD 2010?

9. Section 3.3.3, pages 11374-11375: Firstly, when you calculate the formation rates from the particle data measured with the NAIS particle mode, you'll get the total particle formation rates (total = neutral + neg + pos). Page 11374, line 25: Replace "neutral" with "total particle" or "particle". Secondly about the terminology, ion-mediated nucleation (IMN) is ion-induced nucleation (IIN) involving ion-ion recombination. Check how you should use these two terms in the text. The fraction of IIN out of the total particle formation can be calculated as

$$(J2(+) + J2(-)) / J2(\text{total}),$$

whereas the fraction of IMN is

$$(J2(+) + J2(-) + J2(\text{rec})) / J2(\text{total}),$$

where $J2(\text{rec})$ is the ion-ion recombination rate [$\text{cm}^{-3} \text{s}^{-1}$] (Manninen et al. ACP 2009).

$$J2(\text{rec}) = \varepsilon \times \alpha \times N(+) \times N(-)$$

is the recombination rate. If you divide it with coagulation sink [s^{-1}] (as in your Eq. 3), you'll get the maximum ($\varepsilon=1$) concentration of neutral recombination clusters or particles formed in ion-ion recombination [cm^{-3}]. The Eq. 3 in your ACPD paper is the fraction of neutral particles formed in ion-ion recombination out of the total particle

C4345

number concentration in that certain size range. Thus, it's not fraction of ion-mediated nucleation rate. In paper Manninen et al. ACPD 2010, only the fraction of IIN was calculated (ion-ion recombination was not yet included). Manninen et al. 2010 got IIN fraction of 27% for Jungfraujoch, whereas your IIN fraction would be $(0.19+0.28) \text{ cm}^{-3} \text{ s}^{-1} / 2.03 \text{ cm}^{-3} \text{ s}^{-1} = 23\%$ using median values reported in the text. Perhaps, you could also calculate this value more accurately as median fraction for all the event days. In my opinion the agreement is good. Anyway, we have used different number of days in our analysis.

10. I apologize, that in Manninen et al. ACPD 2010 paper we have classified Jungfraujoch events inaccurately for May and June 2008. Bad data days seem to be somehow shifted to NPF event day class. We'll correct it. Nevertheless, this won't change our results for the NPF event parameters like growth and formation rates. We calculated those parameters only for the most representative events.

Technical corrections

1. Check the language.

2. p. 11362, lines 6-7: The expression 'secondary charged aerosols' is misleading and should be improved.

3. p. 11362, line 10: "We found that the total ion concentration, which is dominated by cluster ions, shows a strong diurnal pattern..." Here, the suggestions are marked with italicization. In the ACPD paper you introduce only ion number size distributions. Therefore the original sentence was misleading.

4. p. 11363, line 5: The word "Long term" should be changed to "Long-term".

5. p. 11363, line 17: replace ":" with ";;".

6. p. 11363, lines 24-25: Add following details: "...during the EUCAARI (European Integrated project on Aerosol Cloud Climate and Air Quality interactions) intensive observation year 2008-2009 (Kulmala et al. 2009; Manninen et al., 2010)."

C4346

7. Perhaps you could have following structure in section 2: 2.Methods, 2.1 Measurement site, 2.2 Neutral cluster and Air Ion Spectrometer (NAIS), 2.3 Data analysis 2.3.1...
8. page 11364, lines 18-19: The word “from” and “detected” should be changed to “with” and “classified”, respectively, and the word “selected” should be removed.
9. page 11364, lines 25-27: rephrase unclear sentence e.g. “...selection of 21 different mobility ranges of atmospheric ions with two parallel differential mobility analyzer – one of each polarity – and their...”
10. page 11365, lines 10-11: “Previous study by Asmi et al. (2009) defined the limit of the neutral particle detection down to ~2 nm. Below this size, particles measurements are not relevant since the post-filtering process cuts also the sampled newly formed particles.
11. page 11366, line 7: Replace “0.4 nm” with “0.5 nm” because you did not even measure at 0.4 nm.
12. page 11368, line 4: Replace Manninen et al. 2009 reference to Tammet and Kulmala, 2005 (this is the original paper referred also in Manninen et al. 2009). Tammet, H. and Kulmala, M.: Simulation tool for atmospheric aerosol nucleation bursts, *J. Aerosol Sci.*, 36, 173–196, 2005.
13. page 11368, line 20: “During the night, the total concentration of ions...”
14. page 11369, line 10: Did you mean NO_x?
15. page 11369, line 10: “...some gases (like CO)..”
16. page 11369, lines 15-16: “...Considering those previous works and the diurnal variation observed at Jungfraujoch within this study, the data set has been segregated into two sub data sets composed of...”
17. page 11370, line 8: remove abbreviation “(GCR)”. It’s not used later in the text.

C4347

18. page 11370, line 26: Replace “were” with “are”.
19. page 11371, lines 2-3: Unclear sentence: “The limit value of RH used to distinguish clear sky and cloud conditions was validated on Puy de D  me data (unpublished data).”
20. page 11371, line 6: Move Table 1 to Appendix A (as Table A1). Reference to Appendix A is missing in the text.
21. page 11371, lines 8, 16: Fig 3 is missing a- and b-subplot labels. Add to Figure 3 labels a) and b).
22. page 11371, line 17: Replace “average” with “median” and add “average” before words “clear sky concentrations”.
23. page 11372, lines 14, 16: Add reference also to Manninen et al. ACPD 2010.
24. page 11373, line 6: Add the closing parenthesis.
25. page 11373, line 8: “the mean growth rates for Ia and Ib events for size classes....”
26. page 11373, lines 15-17: “Concentration of condensable vapor and source rate were calculated from GRs values according to Dal Maso et al. (2002), detailed results are reported Table 3.” The Table 3 does not include these values!
27. page 11374, line 17: Replace “:” with “and”.
28. page 11374, line 25: Replace “neutral” with “particle”.
29. page 11375, line 15: Replace “2009” with “2010”.
30. Section 3.4: Check the usage of brackets.
31. page 11378, line 24: Don’t start sentence with a number.
32. page 11380, line 24: “high growth rates”

C4348

33. page 11380, line 27: Remove "origin".
34. page 11396, captions: Add "on daytime and nighttime" .
35. page 11394, Fig 1: Why did you want to include size range 0.6-1.3 in to the figure?
In my opinion it could be removed.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/10/C4343/2010/acpd-10-C4343-2010-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 11361, 2010.