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Atmospheric Chemistry and Physics Discussions

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

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We thank all three reviewers for their conscientious work on this paper; we followed most suggestions and answered all comments; Most answers are now included in the text. We hope we have satisfactorily ameliorated the manuscript to meet the reviewers expectations

1 General comments

Referee #2 : *This manuscript deals with NAIS measurements in Jungraujoch GAW-station at Alps in Switzerland during extensive EUCAARI measurement period. Data of 309 days is analysed and discussed. The theme is suitable for this to be published in ACP. However, the analysis are quite thin and does not use any supporting data from station that has quite extensive measurement program, this leaves the analysis and conclusions quite shallow and speculative, which I think is the weakness of this paper. Some of the results have also been presented in Manninen et al. 2010 leaving this paper only little more new results. Also English is some points quite hard to understand, I also recommend checking it carefully.*

Authors : Referees 1 and 2 both argue that our analysis is too simplistic, that new results are needed compared to the paper of Maninnen et al. 2010, and that some of our conclusions are only speculative.
First, we agree that some of our conclusions were reached solely on speculation. We now affine our analysis to either withdraw some conclusions, or strengthen others. Ancillary data are indeed numerous at JFJ, but very few can be related to nucleation and NPF (no biogenic VOC, no H_2SO_4, ...). However, we now examine the relationship between NPF events and H_2SO_4 calculated from SO_2 and UV radiation, and with the CS calculated form the SMPS data. We found that H_2SO_4 seems to have only a minor contribution to NPF events and that other condensing species are probably involved. NPF event occurrence is enhanced when the CS is high, suggesting that in such a low CS environment, the presence of condensing vapours is a determining parameter. Conclusions are far more convincing than in the previous version of the paper and we greatly thank the reviewers for suggesting helpful ameliorations.

J2 nor the initial steps of cluster growth do not show a seasonal dependency but rather an air mass type dependency. Hence, we also investigate the relationship between NPF events and the condensational sink in different air mass types, in order to strengthen our conclusions. We also now examine the formation of new clusters in these different air masses. The new findings are that NPF processes differ according to the air mass type: in most air masses new clusters are created (i.e. nucleation occurs) while in Eastern European air masses, which bear the highest probability of NPF events, the growth of preexisting clusters is rather occuring. In fact, in eastern European air masses, NPF are not often class 1 events, compared to NPF in Atlantic air masses.
We believe that the investigation on the role of clouds on nucleation and NPF event is necessary but complex, and that cloudy conditions should be filtered out to be able to understand what other factors are influencing these processes at high altitude. Conclusions are far more convincing than in the previous version of the paper and we greatly thank the reviewers for suggesting helpful ameliorations. English language was checked.

2 Response to anonymous referee #2

1. R: *Aerosol nucleation, new particle formation etc are used, I would stick just one term.*

   A: The different terms used throughout the manuscript do not refer to the same processes. Aerosol nucleation refers to the first steps of new particle formation, which can be observed only using an instrumentation detecting nanometer-size particles. When the instrumentation detection limit is higher than the nanometer scale, the term "nucleation" can not be used anymore and it has to be replaced by "new particle formation".

2. R: *Ions are classified into 3 different size classes, there are different kind of limits through- out the paper, why is this, why not use one widely accepted size ranges*

   A: We use different size classes for two reasons. A first type of size ranges is use for computing growth factors. We chose those ranges for being homogeneous with the EUCAARI project community, in which GR are computed from NAIS and AIS data for the size classes [1.3:3], [3:7] and [7:20]. The scientific reasons for choosing these size ranges, as explained to referee #1, is that they are representative of different steps of cluster growth to the aerosol size. A second range of size classes are used for calculating 1- the cluster concentrations (considered as condensational supports during new particle formation events), 2- intermediate ions/particle concentrations (which concentrations increase indicate that a NPF event is taking place) and 3- the rest of the ion/aerosol distribution up to 45 nm.

3. R: *Page 11362: Line 5, also theoretical approaches are used as well experiments in nucleation chambers*

   A: We added "theoretical approaches are used as well experiments in smog chambers".

4. R: *Page 11363: I don't understand sentence from line 1 to 5.*

   A: "The formation of those secondary aerosols have been studied by many re-searchers but if the general mechanism is established (gas - particle conversion), predicting where and when the new particle formation will take place remains difficult in natural conditions." is substituted by "The formation of secondary aerosols have now been studied for several decades, but the degree of our knowledge on the theoretical mechanism (gas - particle conversion), is not good enough for allowing to predict where, when and with which intensity new particle formation events will take place in the real atmosphere ".

5. R: *Page 11365: line 13, can you be more precise why limit is 2 nm, I have understood that the reason is slightly different.*

   A: "Previous study defined the limit of the neutral particle detection down to 2 nm. Below this size, particles measurement are not relevant since the post-filtering process affects the sampled newly charged particles (Asmi et al., 2009a)" is substituted by "Previous study defined the limit of the neutral particle detection down to 2 nm. Below this size, particles measurement are not relevant since the charger ions from the corona charger might artificially increase the measurement signal (Asmi et al., 2009a)."

6. R: *Figure 1: I don't find this figure necessary.*

   A: We thougth that this figure could help to understand how we computed GR. We've removed it.

7. R: *Section 2.2.4: What is the pressure level the trajectories are calculated, is there difference in height path of trajectories arriving at 0000 and 1200 hours.*

   A: We used 3580 m a.s.l. as a starting point and isobaric trajectory. About the variation of the altitude and path of the air masses, we add a plot of air mass trajectory density according to altitude but reader have to keep in mind that Hysplit model use 1 degree resolution meteorological data as input so we assume that the output is not relevant to describe local air mass motion such as topographical effects or convection. Assuming that and in our case, altitudes are probably over-estimated since Hysplit cannot explain the air mass ending at 3600 m a.s.l. by topographical effects (see figure 1).

8. R: *Section 3.1: To me diurnal variation is not strong, it exist.*

   A: OK, we've corrected this sentence.

9. R: *Page 11369: paragraph starting from line 19. Is there some support for this conclusion on nighttime FT and daytime advection, daytime advection should be seen also other parameters measured at Junfraujoch.*

   A: Yes, you're right. We add the plot of the diurnal variation of CO which, in first apporximation, could be considered as a PBL tracer. On this graphical view, we can see the diurnal variation of the CO concentration with an increase from the early morning to the early afternoon. This pattern is linked to advection process from the valley to the measurement site.

10. R: *Page 11370: 2nd paragraph. This would be quite easily demonstrated by calculations and analysis, it would make it much more convincing.*

    A: We add a graphical view of the diurnal variation of the CS at JFJ ($\bar{CS} = 2.39 \pm 1.56 \times 10^{-4} s^{-1}$).See figure 2.

11. R: *Section 3.2: Is the cloudiness condition only analysed based on RH, this is not enough, again there is quite extensive measurement program.*

    A: Unfortunatly no LWC data are available for the campaign duration.

12. R: *Page 11371: 2nd paragraph: In Lihavainen et al. 2007 intermediate are lower in cloud that clear sky conditions on the contrary explained here.*

    A: You're right, I made a mistake for intermediate ions behaviour in Lihavainen et al. (2007) and also in Venzac et al. (2007). In both papers, clusters decrease in a presence of a cloud, but intermediate ions concentrations show different behaviours: they decrease in Lihavainen et al. (PBL site), remain unchanged at altitude site (Venzac et al.) and increase in case of high altitude site (this study). Further studies are necessary to understand this phenomenon.

13. R: *Page 11371: 3rd paragraph: This should be rewritten, and needs more analysis, actually I did not get the leading thought in here.*

    A: Here we discuss the impact of cloudy conditions on charged aerosol concentration for different size classes. The difference between median value and mean value indicates that production or growth events occur in the cloud or in the vicinity of the cloud since mean values are always higher thant median values.

14. R: *Page 11373: in GR analysis only 3 cases are in 1a. This is by no means enough to rep- resent statistical meaning, I would combine 1a and 1b cases. The GRs for presented here differ clearly from analysis in Manninen et al, 2010.*

    What is the reason for this?

    A: We combine Ia and Ib event classes. Mean growth rate are [5.1$\pm$1.7, 5.3$\pm$3.5, 5.7$\pm$2.2] for 1.3$-$3, 3$-$7 and 7$-$20nm size classes. Manninen et al. only present GR for the 1 - 3 nm size class. Differences remain in the numerical values but they are still not significant according to the standard deviation. GRs values are computed with an homemade program using size class temporal evolution which in general are very noisy. To fit the gaussian curve on the local maximum of each size class, we need to choose arbitrary bounds on time to avoid bad fitting procedure (pollution peak in the morning could lead to bad fitting for example). According to that and to the accuracy of measurements, it's normal to have slight difference between two operator even if the fitting procedure is the same. I'm not sure Manninen et al. use the same mathematical procedure as us, so difference can occur.

15. R: *Page 11374: line 13-14, this comment is speculative without more analysis.*

    A: We removed the comment.

16. R: *Page 11375: These values for J+/- are different that presented in paper by Manninen et al. 2010. What is the reason for this ?*

    A: Formation rate calculation use GR values and coagulation sink values. To compute those values Manninen et al. and us use different mathematical procedure and program. As we explained earlier for the GR calculation, differences can occur if procedures are different and if operators have to define arbitrary parameters to run the program. This also mean that too much precision on those calculation are useless since each research team use his own procedure. Both for the GR and J calculations, we now compare our results to the ones of Manninen et al., explain from where differences can arise, and conclude that the differences give the uncertainties on such measurements.

17. R: *Page 11376: A figure would greatly help imaging the air mass origin.*

    A: We add a graphical view of air mass origin and path.

18. R: *Page 11376: throughout the manuscript Junfraujoch is mentioned as low CS environment, there should be clear comparison to other sites.*

    A: Yes, $\bar{CS} = 2.39 \pm 1.56 \times 10^{-4}s^{-1}$ at Jungfraujoch (nucleation event: $\bar{CS} = 2.90 \pm 1.12 \times 10^{-4}s^{-1}$, non-event: $\bar{CS} = 1.67 \pm 0.14 \times 10^{-4}s^{-1}$ in out-of-cloud conditions). This value is lower than the one computed by Venzac et al. (2007) for the altitude station puy de Dôme station ($\bar{CS} = 58 \times 10^{-4}s^{-1}$) and for Everest station (Venzac et al., 2009; $\bar{CS} = 15.6 \pm 3.6 \times 10^{-4}s^{-1}$). Compared to other EUCAARI sites (Manninen et al., 2010), JFJ is the station where the condensational sink is the lower.

19. R: *Figure 5 is not clear, which is a and which b, what is the difference between a and b...*

    A: Correction done. a. is the first one and it represents the size distribution of positively charged aerosols according to the hour of the day and the air mass origin for non event days. b. is for event days.

20. R: *Page 11377: line 8: again conclusion are drawn without supporting data, what about wind directions etc., the updraft might be the reason but without any other evidence than earlier studies (measured times ago) it is still speculations.*

    A: As we showed previously with diurnal variation of CO and of the condensational sink, change in size class concentration as well as in size distribution are due to updraft of air parcells from the valley. This is confirm by the diurnal wind pattern which is tyical of valley breeze.

21. R: *Page 11377: line 11: The intermediate ions have different population depending air mass origin, what could be reason for this.*

    A: This is a good remark. Higher intermediate ions concentrations are usually linked to new particle formation events, because under these circumstances, the growth rate of cluster ions is higher than the recombination rate. However there might be other reasons for the intermediate particles to be charged. For instance, when the CS (and hence ion sink) is lower, ions might be more efficiently attaching to intermediate particles. Because we still miss stastistically reliable data (we do not have any CS nor GR (no class I event) for the Nordic air masses which show the highest intermediate ion concentrations), we can not have explanations for these differences which would not be speculation.

22. R: *Page 11378, line 12; It is concluded "that nucleation occurs when condensable vapor concentration are high enough to activate cluster growth". Again, why supporting data from the station is not used. Condensation sinks etc.*

    A: Mean value of condensational sink is higher for event days ($\bar{CS} =$

$2.90 \pm 1.12 \times 10^{-4}s^{-1}$) than for non-event days ($\bar{CS} = 2.54 \pm 1.52 \times 10^{-4}s^{-1}$). Since there is no significant difference in cluster concentration between event days and non-event days, we conclude that nucleation is not triggered by new cluster formation but by the growth of pre-existing clusters. To allow the growth of those clusters condensable vapors are necessary. The fact that the condensational sink is higher during event days could indicates that more "polluted" air parcels are updraft to the site. Those more "polluted" air parcels allowed the cluster growth that's why we think they bring condensable vapors which could activate the cluster growth. We analyzed few anthropogenic VOCs which are measured at the station (Benzene, Toluene, Ethane, Propane and N-butane) but no significant difference where found between event days and non-event days concentration (event days concentration are slightly lower than non-event days). This result show that other compound than those VOCs are implicate in the activation and growth of pre-existing clusters.

23. R: *Page 11379: 1st line, on what bases the diurnal variation is related to updraft, there is no evidence, no supporting data etc. This might be the reason but the conclusion are drawn quite lightly.*

    A: I think we answered to this point along this discussion.

24. R: *H. E. Manninen et al., EUCAARI ion spectrometer measurements at 12 European sites, ACPD, 10, 11251-11313, 2010.*

    A: Correction done.

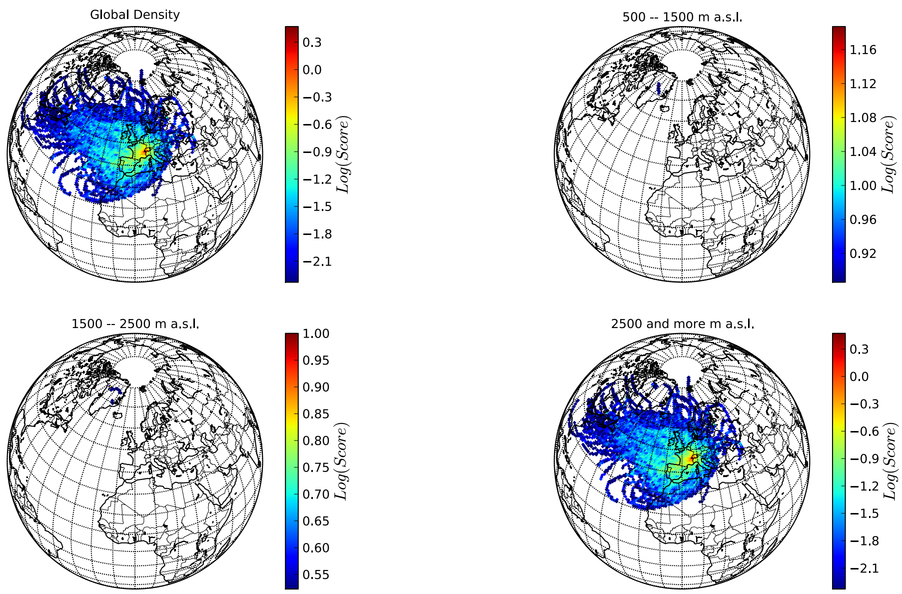

Fig. 1. Hysplit 3-days backtrajectories calculations.

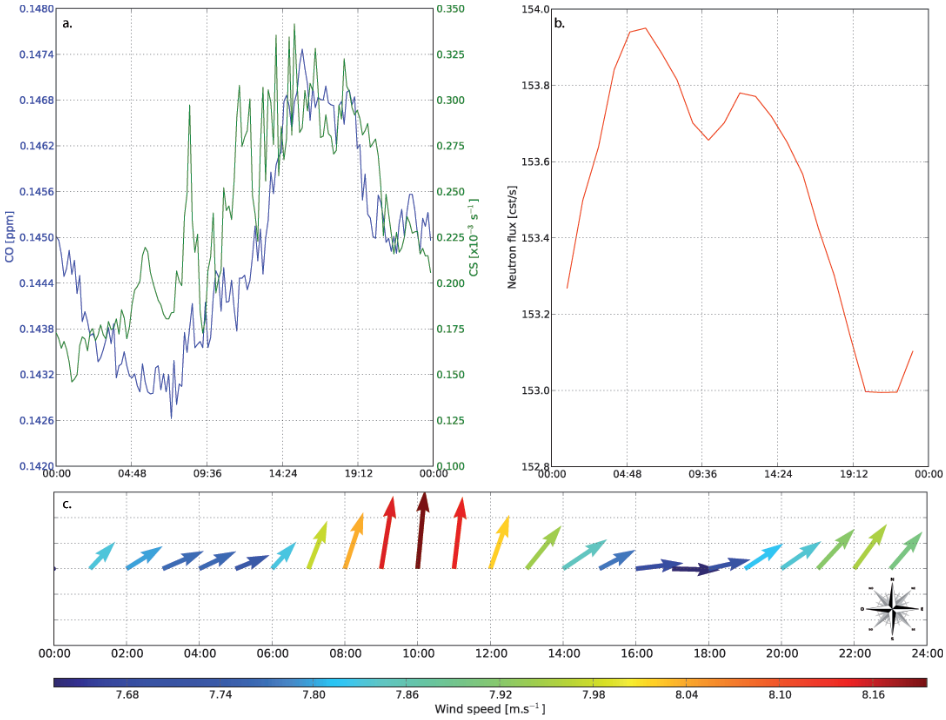

Fig. 2. Dirunal variation of the CS and CO (upper left panel), the neutron flux (upper right panel) and of the wind direction and speed (lower panel).