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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 #1 : *This paper investigates the process of new particle formation at an alpine site based on one year of data from a neutral aerosol and atmospheric ion spectrometer (NAIS) (although I believe other data were available). In my view the paper needs more work and should not be published in its present form. Some of the analysis is overly simplistic, it seems to mainly just repeat what has been done with this instrument at other sites, no ancillary data is presented to support the NAIS data (the instrument description is minimal), and major conclusions are reached based solely on speculation.*

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 Detailed response to anonymous referee # 1

R: *My understanding is that there is still some conflicting views on the importance of ion or neutral species on NPF as a function of location based on both model simulations and measurements. A summary of current findings/views on this would help place the results presented in this paper in a better context.*

A: I add this paragraph in the introduction section :
The formation of those secondary aerosols have been studied by many researchers 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. The role of ions or neutral species remain also unclear. Ion-mediated nucleation (IMN) involves the condensation of vapors on positive or negative ions.

The attractive potential between ions and the dipole moment (induced or not) of the condensable vapor reduces the thermodynamic barrier for nucleation and by the way, enhances the condensational growth (Lovejoy et al., 2004, Nadykto et al., 2004). Conflicting views about the relative importance of IMN subsist between modeling results (see for example Harrison-03, Yu-08) which assess that IMN should be an important source of atmospheric particles and field measurements (see for example Eisele et al., 2006, Manninen et al., 2009). Those later tend to minimize the contribution of IMN to new particle formation in BL environment. A recent paper by Yu (Yu et al., 2008) suggest that IMN can lead to significant new particle production in troposphere. Long term aerosol measurements in different types of environments is still a valuable approach to better understand which conditions promote the new particle production in the atmosphere...

R: *What is special about the air mass location 3 days prior, why not 5 or 10 days prior. In the analysis of data by air mass classification in later sections there is no discussion on the path or variation in altitude of the air mass to the measurement site, can't this affect the air mass characteristics that are important to NPF? The air mass analysis and interpretation of data seems overly simplistic. Maybe a map showing trajectories for the various air masses would clarify this issue.*

A: Because the turnover time of aerosol particles has been evaluated to be from 1.6-1.7 days for nuclei size ranges, to 2.4 days for 200 nm particles (Tunved et al., 2005), we arbitrarily limit the back trajectories calculations to 72 h. 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). We now mention this in the paper.

R: *Pg 11370 lines 19 and 20, updrifted, replace with drifted up, and typo, aroud*

A: We've replaced "updrifted" with "drifted up" and have corrected "aroud" (i.e. "around")

R: *Pg 11372, line 10, what about January - why does it not follow the trend?*

A: Monthly mean values of RH indicates that Januray 2009 was characterized by a very low value of RH (55%) compared to other monthes (71.8 $\pm$ 2.3%). Furthermore, for winter monthes the cloud frequency was extremly low (2.0 $\pm$ 1.3%). This observation is mainly due to the classification method of cloud events (RH > 96%) which could only detect non-freezed clouds but it remains the only way to identify potential cloud events since no LWC measurements are available during all this filed campagn. Explanations still hard to find with data we have. A longer study is needed to better charcterized this seasonal variation.

R: *Pg 11372, on the role of clouds and NPF events. If my understanding is correct, in this paper clouds mean the measurement site is within a cloud (ie the RH is above 0.96), but this classification says nothing about clouds in the vicinity or if air masses reaching the site passed through clouds. This needs clarification and more discussion. I do not find it that surprising that few NPF events are observed in clouds due to scavenging - was the particle surface area measured (was there opc data, or at least look at the SMPS data), if it was I think the authors would reword "cloud*

droplets which COULD remove clusters or/and condensable vapours". (is there any question about this?) I am not sure why the authors make a big deal about this, am I missing something, is there any references to in-cloud NPF? There are references to nucleation in the vicinity of clouds.

A: If NPF in the vicinity of clouds have been observed and are documented (see Shaw, 1989; Hegg et al., 1990), a lack of references remain concerning nucleation and subsequently growth in clouds or in the vicinity of clouds. What we observed is that 1- the nucleation frequency is very low in the presence of a cloud and 2- cloud has an effect on the aerosol size distribution: the main effect is the decreasing of clusters concentration. We find important to emphasis the fact that clouds impact NPF and cluster concentrations, since they are not taken into account in more global/statististical analysis at the European scale (see Maninnen et al. 2010, Spracklen et al. 2010). We now mention this aspect in the conclusion. However, the effect of clouds on the cluster concentrations and NPF occurrence only takes a small section of the paper, it's goal is to justify to segregate between clear sky and cloudy conditions for the seasonality of NPF frequency analysis and air mass dependency analysis. This is alos now better expressed in the text.

R: *Pg 11373 lines 7-13. Can the authors give a physical explanation why or how it is possible that growth rates vary over differing ranges of nanometer sized particles. Given the uncertainty with these calculations, is there a real difference between say 5.3 and 7.8 nm/h? (ie maybe include error estimates for each mean growth rate size range and event category.*

A: Standard deviation to GR values were added. The mean growth rate for Ia and Ib for size class $1.3-3$, $3-7$ and $7-20$ nm are respectively [5.3$\pm$0.8, 7.8$\pm$3.3, 5.4$\pm$0.7]

and [5.0±1.9, 4.1±2.9, 5.9±2.7]. Since no significant difference exists between the mean growth rate value of the two event classes, we decided to only provide the mean growth rate values for each size class for all Ix events. Mean growth rate are [5.1±1.7, 5.3±3.5, 5.7±2.2] for $1.3-3$, $3-7$ and $7-20$ nm size classes. GR can vary over different size ranges of nanometer sized particles because (i) the condensable surface do not grow linearly with size and (ii) the condensable gazes concentrations vary with time (with the intensity of photochemistry for instance). These reasons usually lead the analysis of nanoparticle growth rates to be split in several size ranges. In fact, we found that given the uncertainty on the calculated GR, they do not vary significantly from one size range to the other, but decided to still split our results in usual size ranges for GR calculations.

R: *Pg 11374 line 26, reword J2 was less than four time bigger than J2+-. Maybe give the actual difference, eg, change to: on average J2 was (give the exact number) times bigger than J2+-.*

A: We change to "on average J2 was 8.8 times bigger than J2+-". We also corrected a type.

R: *Pg 11375 lines 20-23. If nucleation is occurring at sizes significantly smaller than 3nm what does the comparison of two measurement techniques for measuring 3nm particles say about nucleation rates? It only provides info on the formation rate of 3 nm particles.*

A: The comparison of the two intrtuments only provides informations on the formation rate of 3 nm particles but NAIS data provides infromation on sub-3 nm particles too. But we choose to use only data for diameter bigger than 2 nm since lower diameters

data are very noisy.

R: *Pg 11378, line 19, is the Metzger et al paper published?*

A: Yes, it is already publihed. The reference is at the end of the paper.

R: *Page 11379, line 1 states ... was related to the updraft of surface layer air parcels rich in preexisting particles and ion sources such as radon from the valley during the day. As far as I can tell there is no data in this paper to support that statement, it seems to me that all data on which this is based is circumstantial. There is really also no support for line 3, that radon was found to be the main cluster ion source at the station. Radon was never measured.*

A: You're right, radon was not measured during this field campagn. But previous study show that there is a strong diurnal variation of radon especially during summer time which is correlated with the increase of surface area (Lugauer-00). Radon is a tracer of PBL injection at the site. Furthermore, in order to argue this hypothesis we've added a new graphic showing the diurnal variation of a PBL tracers (CO,CS) and wind diurnal variation. It clearly shows that PBL injection occurs at the measurement site. The known sources of ions at the measurement site are either radioactive species such as radon or cosmic rays (CR). Due to the local cosmic rays anisotropy, regular daily variations of the CR flux is observed estimated to be on average 1% (Usoskin and Kovaltsov, 2008). Other variations of CR fluxes could be more important but their timescale are too long to explain the observed diurnal variation at the measurement site. Since the observed diurnal variation of charged aerosol concentration is higher than 1% (22.5% in average). To complete and proof this hypothesis, we've added the neutron flux diurnal variation and now it's clearly evident that the diurnal variation of

charged aerosol concentration is not linked to CR. According to that, we assumed that cosmic rays are not the major ions source at the measurement site and that radiocative species such as radon could be responsible of the ion production at low altitude and then, those ions could be transported to the measurements site. This is now better explained in the text (see figure 2).

R: *Pg 11380 line 4-6 (last line of conclusions). Again a conclusion is reached based on zero data. The authors have little justification to make the statement that "this result confirms that the nucleation process at Jungfraujoch depends on the presence of condensable vapours which allowed clusters to grow rather on nucleation of new cluster (should it be: rather then nucleate a new cluster?).*

A: Our conclusion is based on the recent study made Lanz et al., 2009. Authors have characterized hotspot emissions of VOC around JFJ and they conclude that eastern air masses are rich in VOC. We showed that the nucleation frequency of eastern air masses is higer than in other air masses. The first conclusion is a correlation between those two studies. To investigate this hypothesis we analyzed the VOC composition of each air mass origin (Toluene, Benzene, Propane, N-Butane, Ethene) and no significant difference was found between the VOC composition of air masses. According to these results we conclude that those VOC are not implicated in the nucleation/growth mechanism but other compounds or derivated species since compounds like Toluene, Benzene or Ethene are knew to be reactive species in atmospheric conditions. However, we agree with the reviewer that our conclusions were too definitive while based only on indirect evidences. We now are more cautious in our interpretation.

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

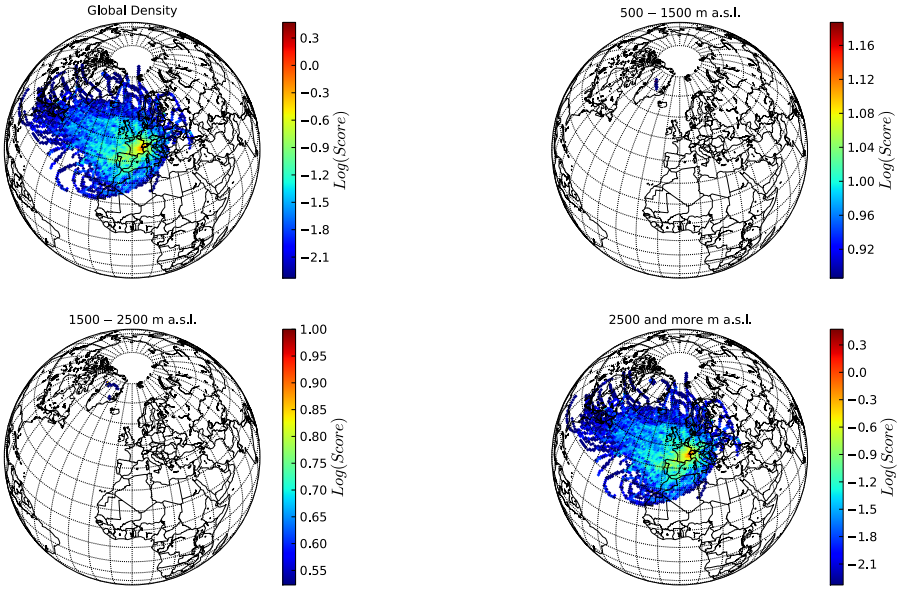

Fig. 1. Hysplit 3-days backtrajectories calculations.

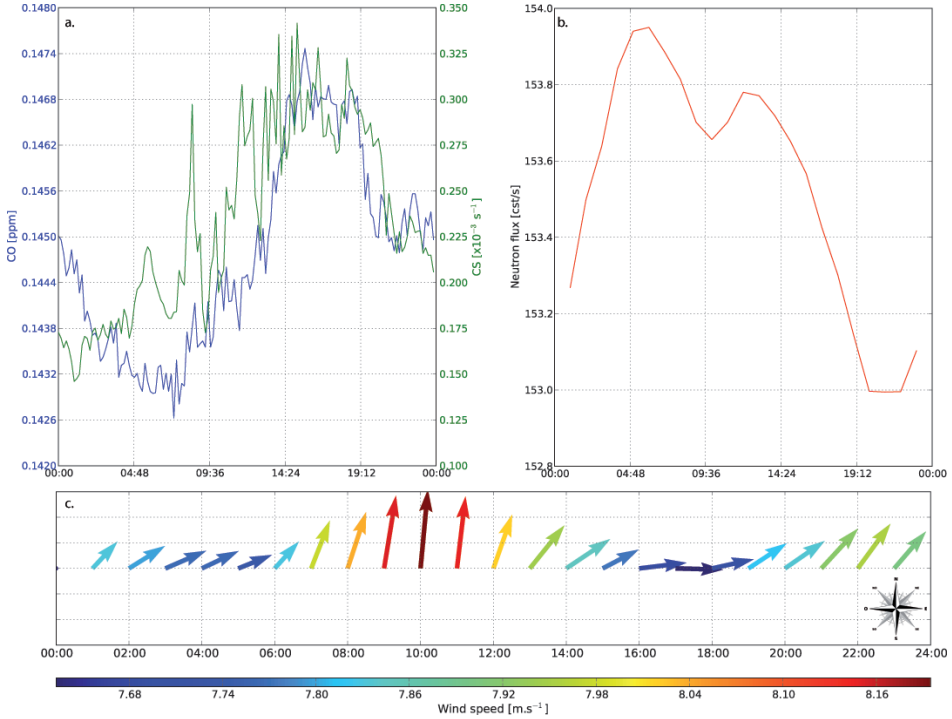

Fig. 2. Dirunal variation of the CS and CO (upper left panel), the neutron flux (upper right panel) and of the wind direction and speed (lower panel).