

The authors are very grateful for the positive feedback and the comments from the referees concerning this manuscript. Here are our answers to the minor remarks from both referees (original remarks in italics, answers in red). The manuscript was carefully revised taking them into consideration.

### Comments from referee 1:

*Additionally, although the paper is certainly very understandable, it would benefit in a few places from some minor editing to improve the English.*

The language was revised by a native speaker.

*P26629. Line 21: It is hard to unambiguously assign new particle formation events to natural or anthropogenic sources. I suggest removing the word "Natural".*

Suggestion taken.

*P26635, Line 10-11. "Correspond well": in what sense? It would be good if you could be more quantitative here.*

We meant that the concentrations measured with the two instruments agree and have a similar time series. This is now stated more clearly in the paper.

*P26635, Line 14-15. How is the clean sector defined? Please give details.*

The details of the air mass analysis are given in chapter 2.3. We added a reference to O'Dowd et al. (2002), in which a figure of the sectors is presented.

*Table 1. The ranges quoted in the paper are very broad. Are these the range of minimum to maximum concentrations that were observed? Please define more clearly. Also I suggest including campaign mean and median concentrations at both sites.*

The ranges corresponded roughly to 5 – 95 percentiles of the data sets. This is now stated more clearly, and the values are given with 2 significant digits. The mean and median was added to the table.

*P26637, Line 7. Is this the only explanation for the difference in diurnal cycle and charged fraction? I think it is difficult to unambiguously use this to conclude that the sources and chemical composition of the clusters at the 2 sites are different? Indeed, the next sentence of your conclusion states that the composition is hard to measure and is not known.*

It is true that for example the meteorological conditions and background aerosol population (sinks) also affect the concentrations and diurnal variation of the nano-CN. So maybe we jumped to a conclusion here, thanks for pointing that out.

However, we did show that the CoagS is not enough to explain the difference in concentrations, and that a bigger fraction of the nano-CN is explained by ions in Mace Head. Thus it does seem that the sources differ at least in magnitude. We revised the first chapter

in section 4 to make the statement more precise and abstained from mentioning the composition.

*Page 26637, Line 12. A relationship between solar radiation and nano-CN has not been well shown. If the authors want to make this conclusion then I would like to see this analysis extended somewhat to demonstrate this more clearly.*

We added the calculated mean solar elevation in Mace Head during the campaign period to figure 4 (new version in the end of this document). Now it shows clearly, that as long as we don't have sunlight, the nano-CN concentration is stable and low, and the concentration starts to increase soon after sunrise, reaching the maximum at the same time as the sun is at its highest elevation.

*P26632, Line 4. Define GAW.  
Page 26637, Line. Change "form" to "from".*

Done.

#### **Comments from referee 2:**

##### *Remarks/Questions:*

- p. 26630, l 3: . . . to incorporate. . .*
- p. 26630, l 7: . . . nucleation on 1.2 nm clusters. .*
- p. 26633, l 19: . . . calculated with the HYSPLIT. . .*
- p. 26635, l 21: . . . On a few days nighttime maxima. . .*

Corrected.

*p. 26632, l 28: Isn't this somewhat in between an optical and a mobility diameter which is determined by the PH-CPC? Sipilä et al found the inversion of PH-CPC data to depend on the particle composition. Could this dependence account for a significant part of the missing nano-CN's at Mace Head? I admit however, that this seems unlikely and therefore may have no severe implications for the conclusions drawn in this study (and you shortly point to this on p. 26637, l 24 ff).*

The pulse height analysis relies on detecting the optical size of the final droplet (the particle that has grown in the CPC by condensation of butanol), that's true. However, the relationship between the final droplet size and the initial particle size was experimentally defined based on mobility diameters. The PH-CPC was calibrated against charged particles classified with a high-resolution DMA. Thus for insoluble singly charged particles, the inversion gives out a mobility diameter. A small error is made when extending the calibration results to neutral particles, since the charge affects the activation probability (Winkler et al. 2008).

The activation probability of very small particles in the CPC depends also on composition. Again, the calibrations were done for insoluble particles, so unless the particles would repulse butanol, we shouldn't seriously underestimate the concentration in Mace Head due to composition. On the contrary, we might overestimate the size and/or the concentration, if the nano-CN are lipophilic attracting butanol (Kulmala et al. 2007).

The good agreement with the NAIS, which is detecting and classifying the particles electrically, and the PH-CPC in Mace Head (Fig.3) gives confidence that our method is not too much affected by the composition.

*p. 26633, l 25ff: A brief motivation for the application (ion- vs. neutral particle activation) of eq. 1 would facilitate to keep track at this point.*

We added a few sentences motivating the use of eq. 1 to the beginning of the second chapter in section 2.3.

*p. 26635, l 25: Are there estimates for the relative contributions of radon-induced and cosmic ray-induced ion concentrations. Can the latter at some point dominate?*

Cosmic rays have a significant contribution to the total ionization rates, producing about 2 ion pairs  $\text{s}^{-1} \text{cm}^{-3}$  in the boundary layer (Carslaw et al, 2002). The ionization by radon ( $^{222}\text{Rn}$ ), and therefore the relative contribution of these two sources, varies a lot depending on the geographical location, soil type and water content etc. In Hyytiälä, the contribution of radon to total ion production rate is on average 10% (Hirsikko et al., 2007). The emission of radon from oceans is negligible compared to land areas, so the cosmic rays will certainly dominate the ion production in marine air masses. In fact, the radon concentrations have been used as a tracer for continental air masses in Mace Head (e.g. Biraud et al, 2002).

The ion concentrations, however, are also affected by the ion sinks, which may also vary between the marine and continental air masses.

*p. 26636: l 17ff: Can it be excluded that vapour depletion leads to significant underestimation or even miss of extreme particle bursts?*

*Fig 6: If you can't explain the sharp drop in particle concentration around 13:00 LT, it might be better to exclude or mask these data in the plot.*

In general, it cannot be excluded that the vapor depletion leads to underestimation of the nano-CN concentration when background particle concentration is very high or changes rapidly. We wouldn't miss a particle burst completely, but the size distribution would be distorted. This is one reason for always using multiple instruments at a site simultaneously.

In the example case, as can be seen from total concentration data in the original figure 6 and also from the NAIS&SMPS data, the particles appear simultaneously in the whole size range ~2-20 nm. The background concentration stays very stable until the particle burst occurs. Thus the vapor depletion is not an issue BEFORE the burst, and we do not miss any changes in the nano-CN concentration due to it. Only DURING the burst, when the concentrations are already high, we are likely to underestimate the concentration of sub-2 nm particles, which is explaining the dip around 13:00 in the PH-CPC data.

Thus we can still rule out, that the particle formation would have started from nano-CN sizes at the measurement location, but it seems more likely that the nucleation happens on the coastline and the particles have time to grow before reaching the measurements. Ion measurements support this conclusion.

We chose to include the NAIS data to figure 6 to give confidence to the results and show the size distribution until 40 nm. The new version of Fig 6 is in the end of this document. A shortened version of the explanation above is included in the text in chapter 3.2.

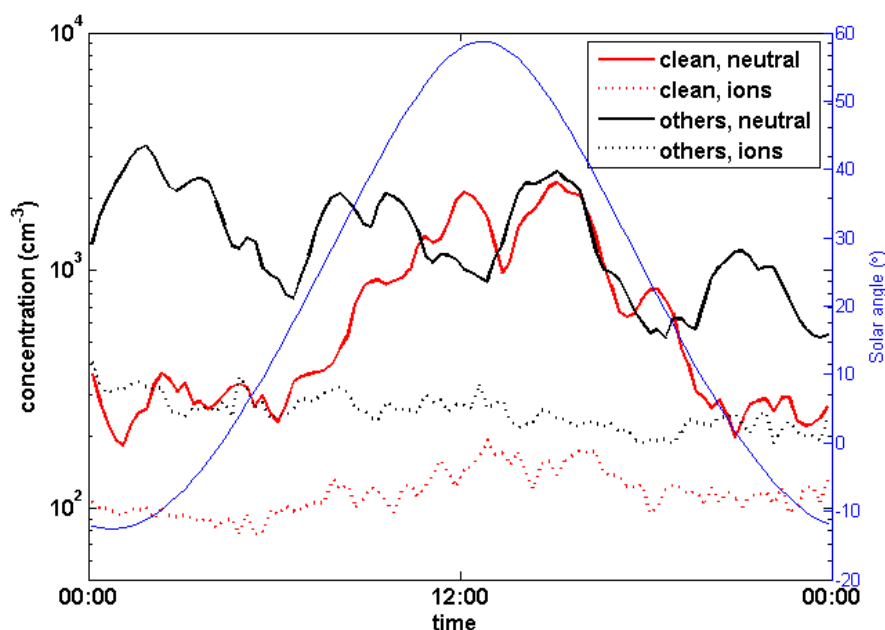
Figures:

Fig. 3: Reduce the symbol size for PH-CPC

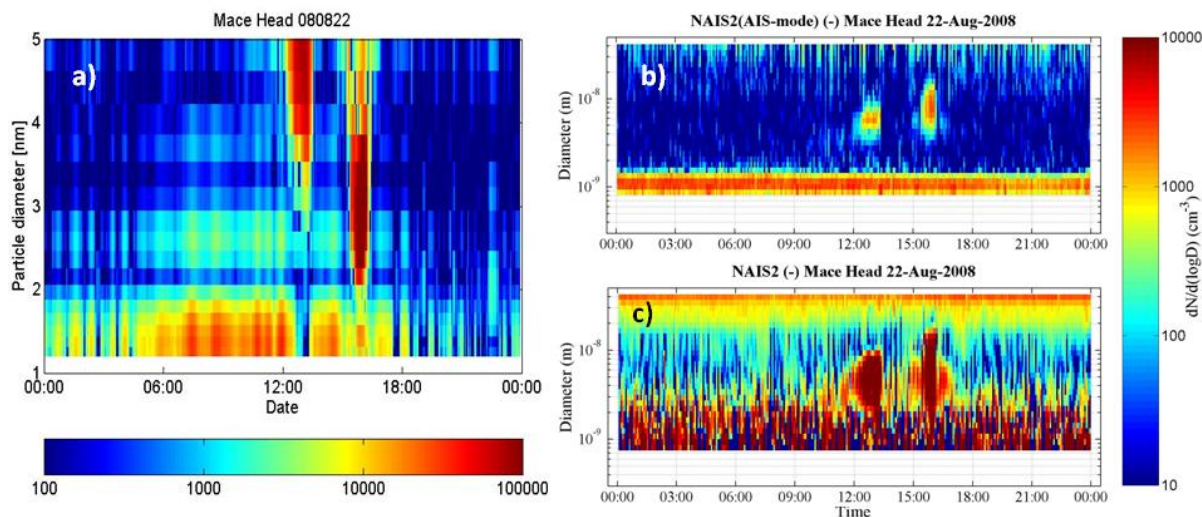
Done.

Fig 1+4: In both figures there occurs an obvious peak in the nano-CN diurnal variation around mid-night. Can this be an averaging artefact?

This was tested by choosing different averaging windows for the data. It seems that the midnight-peak in Fig.4 indeed is (at least partly) an artifact, since with the original averaging time step, we had a lot of NaNs as the first value, and the median of the remaining values happened to be quite large. When changing the time step to 15min, the problem of the first (and last) values was fixed. This did not change any other general features of the graph (the new figure is attached below). Figure 1 already had the averaging window as 15min, and since we cannot really see any strange behavior at midnight (nor did it change when testing different window sizes), the figure was left as is. It also makes more sense to have the same averaging period in both figures. Thank you for noticing this!



**Figure 4.** Diurnal variation of nano-CN and 1.5-3 nm ions (pos+neg) in Mace Head. The data is divided into two fractions depending on whether the air mass trajectories came directly from the ocean (clean, red lines), or if they had passed over land areas (others, black lines). The blue line represents the solar elevation angle in Mace Head during summer.



**Figure 6.** Size distribution of 1.3-5 nm particles measured with the PH-CPC (a), 0.8-40 nm negative ions (b) and total particle distribution (c) measured with the NAIS in Mace Head 22 August 2008. Low tide occurred around 3:30 and 16:00.

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