# Supporting Information for "Contribution of new particle formation to the total aerosol concentration at the high altitude site Jungfraujoch (3'580 m a.s.l., Switzerland)"

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## 1 Comparison of several instruments stationed at the Sphinx observatory

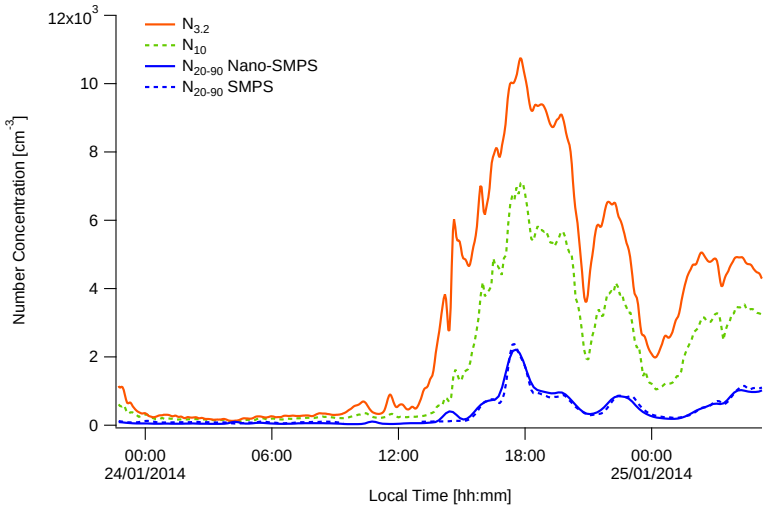


Figure S1: Comparison of total number concentration during nucleation event measured with nano-SMPS $N_{20-90}$, SMPS $N_{20-90}$, CPC 3772 ($N_{10}$) and CPC 3776 ($N_{3.2}$). After diffusion loss, charging efficiency and CPC detection efficiency correction there is excellent agreement between SMPS and nano-SMPS.

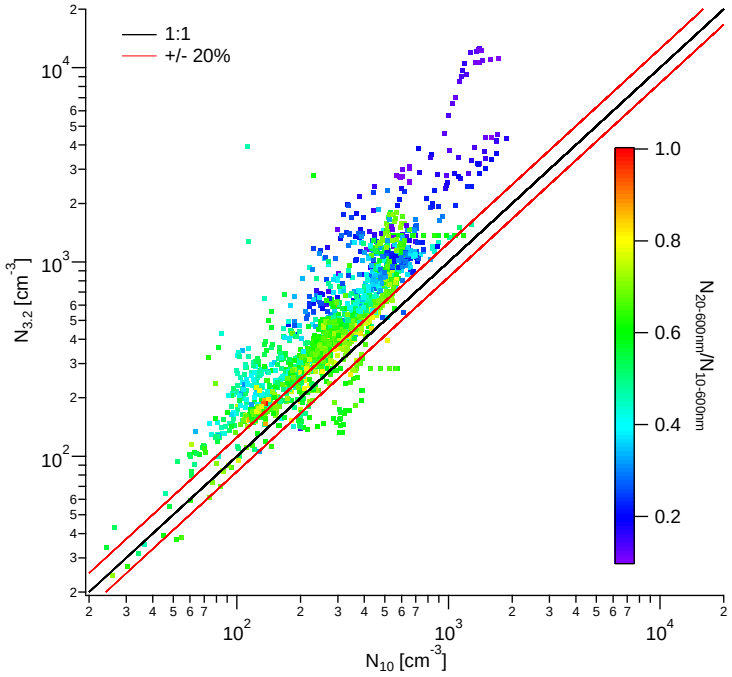


Figure S2: Detailed comparison of CPC 3772 ($N_{10}$) and CPC 3776 ($N_{3.2}$). Due to the different cut-off efficiencies, we expect that $N_{3.2}$ is always larger than $N_{10}$, as the aerosol number size distribution is generally dominated by particles in the Aitken and nucleation mode (see Figure S3). At times when the cut-off difference is of minor relevance, the two CPCs are within $\pm 20\%$.

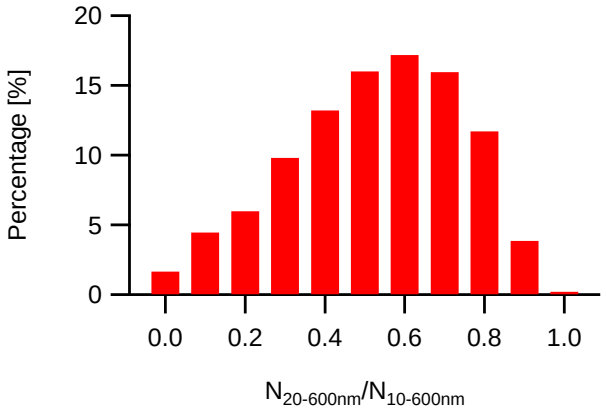


Figure S3: Histogram of the ratio $N_{20-600}/N_{10-600}$ during the NUCLACE LOP. $N_{20-600}$ is measured with the SMPS which is corrected for diffusion losses, single charging efficiency and multiple charges. $N_{10-600}$ is measured with the CPC 3772 (TSI). The flow in the sampling line is high so that diffusion losses are minimized. A difference in the ratio $N_{20-600}/N_{10-600}$ thus mainly reflects the differences in the cut-off of the instruments. We can therefore conclude that Aitken and nucleation mode particles dominate the aerosol number size distribution at the JFJ.

## 2 Determination of formation rates

We tested different nucleation rate calculations consistent with Kulmala et al. (2013) , where we consider a smaller bin (3.2 - 5 nm and 3.2 - 10 nm) with the corresponding growth correction to account for particles that grow out of the size bin. The results were similar but not identical, as expected, as we consider different sizes with these approaches. We decided to use the method introduced in the manuscript methods, as it allows to estimate the nucleation rate of particles with the lowest possible diameter. For all methods (even for the method with reduced size bin), there could be transport of particles in the considered smaller size bins, but it is not possible to determine the value exactly, so that for all tested methods an increased uncertainty due to transport remains. The fraction of transported particles is more significant in summer; however the number concentration due to nucleation increases so dramatically that this increase in larger particles becomes often negligible.

To roughly estimate the contribution of net transport to the apparent nucleation rate (which can be positive or negative), we calculate the formation rate of 10 nm particles ($J_{10}$). $J_{10}$ is corrected

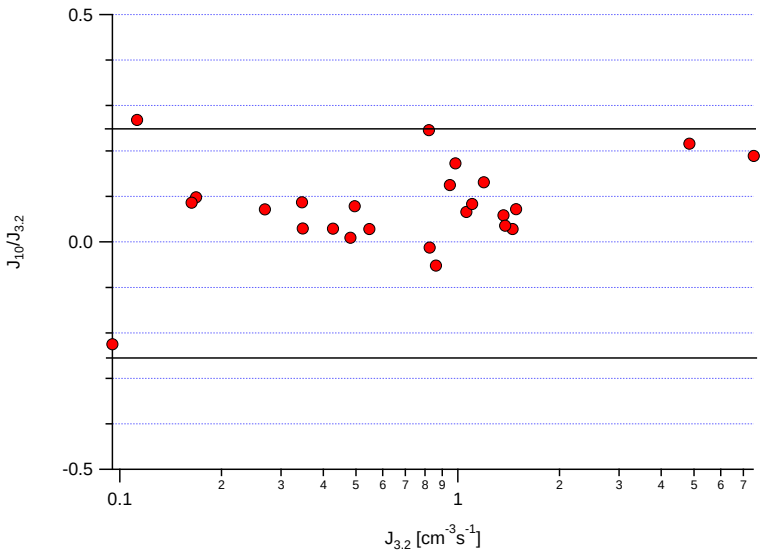


Figure S4: Comparison of $J_{3.2}$ and $J_{10}$ to estimate a possible bias due to transport. The ratio $J_{10}/J_{3.2}$ is at least a factor of four larger, often orders of magnitudes.

in the same way as $J_{3.2}$, this means transport processes were not included in the analysis. We averaged $J_{10}$ for the same time interval, where $J_{3.2}$ was estimated. During the time when $J_{3.2}$ is calculated, the particles hardly have reached the size of 10 nm so that the true formation rate of 10 nm particles is virtually zero. Thus, $J_{10}$ should represent interference from transport processes only and the ratio of $J_{10}$ and $J_{3.2}$ is an indicator of the interference from transport. $J_{10}$ is always at least a factor of 4 smaller, often a factor of 10, indicating that transport interference is always smaller than $\sim$25% of the measured $J_{3.2}$ and often smaller than $\sim$10%. To be conservative, we assume a maximum additional uncertainty of $\pm$25% due to the net transport. Using this approach one limitation remains, as we cannot differentiate between off-site nucleation events that add particles below 10 nm and real nucleated particles that were formed in-situ. However, the concentration of particles resulting from off-site nucleation events is rather low compared to the concentrations during in-situ NPF events, which can be seen by comparing Figure 2 and Figure 10. However, it needs to be noted that it is impossible to precisely quantify the contribution of transport processes to the nucleation rate.

# 3 NPF during summer

In summer we find two different situations (see Figure S5).

- Nucleation in the afternoon:

  Mountain ranges favor the formation of the so-called injection layer (Henne et al., 2005a). Especially in summer, this injection layer can reach up to 4000 m a.s.l. (in particular during the early afternoon), so that the JFJ sits within this injection layer (Nyeki et al., 2000; Henne et al., 2005a; Nyeki et al., 2002). This injection layer is a large scale phenomenon in the Alps which allows for spatial homogeneity. This layer consist of 20-30% of air that originated from the PBL with the rest being attributed to FT (Henne et al., 2005b). As we do see the onset of nucleation, the nucleation must occur either in the FT or in this injection layer. It

is not possible to differentiate experimentally between those two layers, as it would require e.g. LIDAR measurements, which were not available during NUCLACE.

- Nucleation before noon:

  Here the precursors might have been entrained the previous days or by up-slope wind, so that photo-oxidation can already start before noon, with the first mechanism similar to that suggested by Bianchi et al. (2016) and supported by FLEXPART dispersion modelling.

In winter we only experience the second case, as the large scale venting is not as pronounced at the altitude of the Jungfraujoch anymore. We find that nucleation was always triggered by the injection of PBL air, as indicated by the CO/NOy ratio.

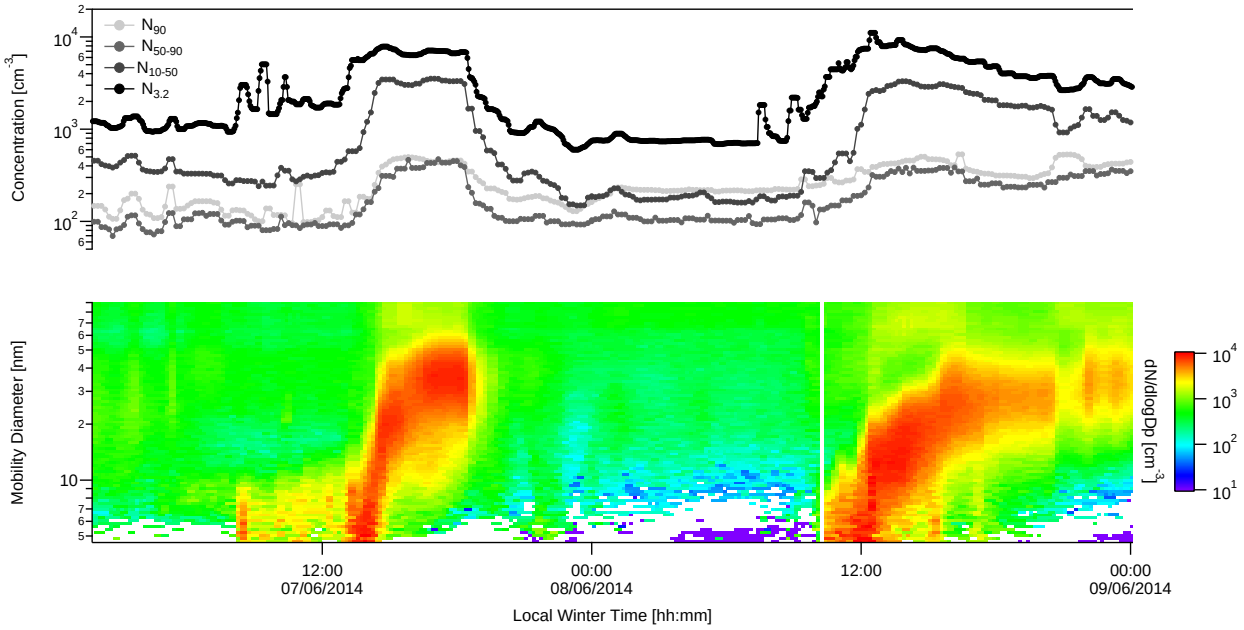


Figure S5: The first event starts in the afternoon and is most likely caused by local venting of polluted boundary layer air and thus aerosol precursor into the injection layer. The second event starts before noon. The precursors could have been entrained in the approaching air masses some hours before nucleation, as suggested by Bianchi et al. (2016) and supported by FLEXPART dispersion modelling, or they could have been entrained via local up-slope wind.

# 4 $SO_2$ vs CO

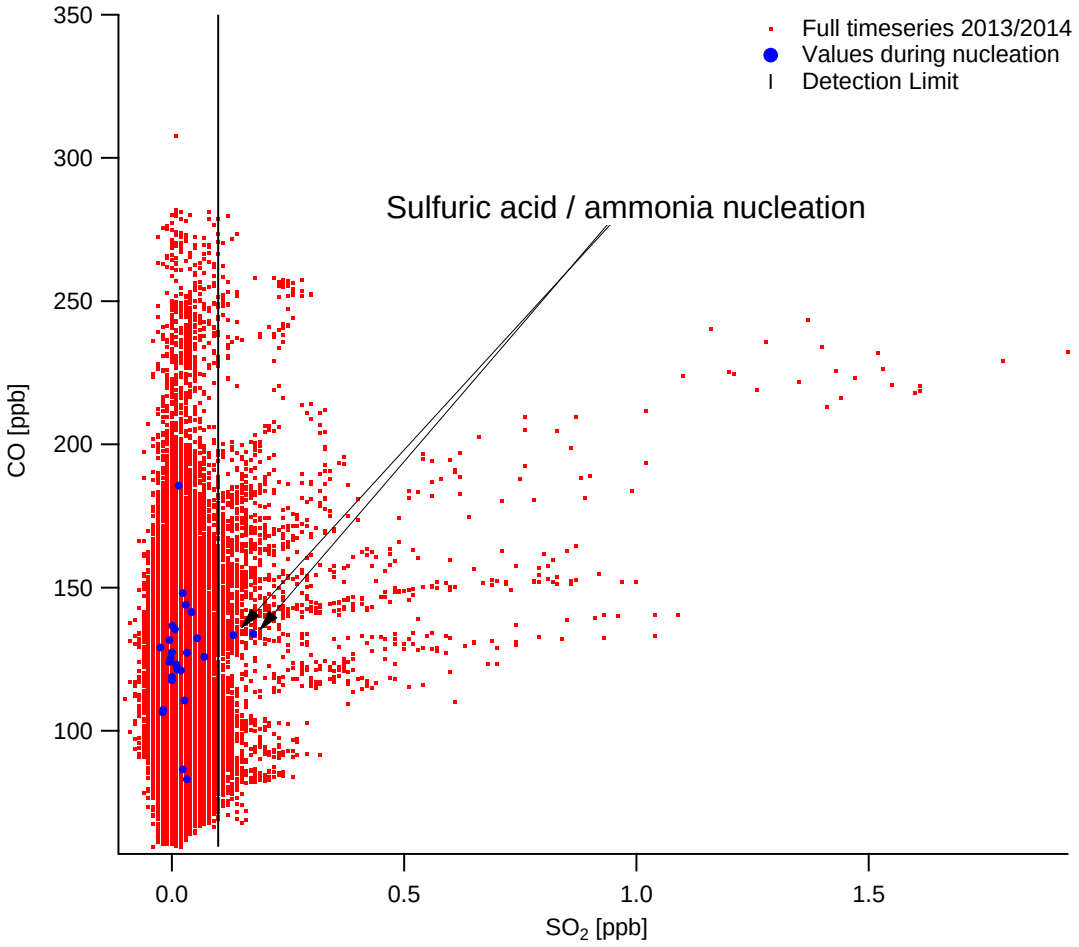


Figure S6: CO vs $SO_2$ for the period 2013/2014 (red dots) and during nucleation events (blue dots). Only two clear sulfuric acid and ammonia driven nucleation events (based on APi-TOF data), are above the detection threshold.

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