

**From ACP editor on February 20th (Roya Bahreini):**

*Dear All,*

*Given the unique set of measurements provided here, I would like to consider this manuscript for publication as a Research article (and not a Measurement Report) if you can provide more context on atmospheric implications of the observed trends. Please let me know if you have any questions.*

*We thank the editor for considering our article as a Research Article.*

*As suggested also by the two reviewers, we added more context on the atmospheric implication in the conclusion. The added paragraph is written in green font:*

*From the conclusion:*

*A new tailored flying API-TOF mass spectrometer was deployed in a Cessna 172 aircraft to measure the vertical profiles of atmospheric ions above the boreal forest at the SMEAR II station, covering the stable boundary layer (SBL), residual layer (RL), mixed layer (ML) and free troposphere (FT). The measurements showed that the ion composition within the BL and FT is distinctly different, and that the ion chemistry of ML, SBL, RL, and FT is highly dependent on the time of day and air mass origin.*

*We detected HOM, originating from the forest VOC emissions, within the whole BL during daytime. The measured spectra in the ML were identical to a co-located reference measurement, which was performed on top of a 35-meter tower. Due to ongoing photo-oxidation during the daytime, most of the detected negative ions were the bisulfate ion and its clusters with sulfuric acid, forming sulfuric acid dimer and trimer, and bisulfate-HOM and nitric acid-HOM clusters within the whole ML. The nocturnal RL was clearly distinguishable from the other layers by its differing ion composition and revealed its own ion chemistry with the highest day-to-day variability compared to the other layers. Depending on the air mass origin and possible decoupling of the SBL, the measured air in the RL could contain halogen compounds, MSA and carboxylic acids, and HOM. The FT showed a distinctly different ion composition than the ML, SBL or RL. In the FT, a high abundance of carboxylic acids and some unknown compounds with masses in the range 200–400 Th were observed, probably clustered with carboxylic acids. Furthermore, we detected some very specific ion patterns on two days of measurements within the FT, which likely contained halogens (most likely chlorine), due to their apparent isotopic pattern. The daytime measurements in the FT mainly showed sulfuric acid, MSA, nitric acid and malonic acid, however we did not detect HOM.*

*As we summarised in the introduction, ion-induced nucleation is the main NPF pathway in several environments, especially under conditions when abundances of nucleating precursor gases are relatively low. At high enough precursor concentrations, ion-induced nucleation becomes less relevant for initiating atmospheric NPF. The compounds we observed in the various layers favour NPF, and even though our measurements focused on ions rather than neutral vapours, the ion chemistry sheds light on the available precursor gases. From our observations we could conclude that in the FT above the boreal forest, NPF could be initiated via ion-induced nucleation of inorganic compounds, such as sulfuric acid and iodic acid, also under the aspect that lower temperatures favour the nucleation (e.g. He et al., 2021; Duplissy et al., 2016). Within the boundary layer,*

however, it is more likely that a mixture of various compounds initiates the nucleation, especially as HOM have been observed within the whole ML. Furthermore, NPF has been observed at Jungfraujoch (Bianchi et al., 2016) and the Himalayan mountains (Bianchi et al., 2021) when organic compounds were transported upwards and mixed into the free troposphere. Also Rose et al. (2015) observed NPF at Puy de Dôme, at the interface between the boundary layer and FT, albeit nucleation there was driven by neutral compounds rather than via ion-induced pathways. Still, this could indicate that a possible mixing of the organics in the mixing layer with the inorganics in the FT would provide good conditions for nucleation, e.g. during roll vortex events (Lampilahti et al., 2020). Thus, it could be speculated that the multi-component nucleation as proposed by Lehtipalo et al. (2018) is driving the nucleation. Moreover, above we discussed the nucleation mechanisms and nucleating precursor vapours, while it should not be forgotten that for NPF as a whole, also vapours that further grow small clusters into larger sizes are highly relevant.

*Our observations represent only a snapshot of the ion composition in the BL and FT during springtime above the boreal forest. More observations, especially in the RL and FT, during other times of the year would be necessary for getting a better understanding of the chemistry and behaviour of ions in those layers and, ultimately to quantify the role of ions in NPF in different parts of the global troposphere.*