

This study details the composition of positive and negative ions detected at Jungfraujoch over a 9 month campaign. Negative ions were primarily sulfuric acid (and its clusters), MSA, malonic, nitric, SO_5^- , and halogenated species. Positive ions were amines and clusters with ammonium/amines. The authors show very interesting correlations of the SO_5^- and MSA, even during night time measurements. The halogenated compounds were tracked back to air masses from the ocean or sea.

This paper is written very clearly and presents new and important science about the composition atmospheric ions and the dynamics at Jungfraujoch. I recommend this paper be accepted after the authors address a few, mostly minor comments.

Major Comments:

Section 3.4, NPF events: The authors explain how the mass spectra differ between observed NPF events. These results, and using FLEXPART to relate NPF to CO, have already been explained in Bianchi et al. (2016). The section in this paper does not add any more information to the results shown in Bianchi et al. (2016) and does not relate to the title/focus of this manuscript. The authors should either add in new NPF information in this section that is not covered in Bianchi et al. (2016) or should just remove the entire section (and corresponding figures and text in abstract and conclusion).

Page 3, Line 2 and Page 11 line 24: The authors conclude that an increase in sulfuric acid signal during the night could be due to heating of snowflakes on the APi-ToF sample tube. 1) Can the authors specify the temperature at the inlet of the APi-ToF (right before the ions enter the vacuum chamber)? Is this temperature close to the outdoor temperature or the temperature of the room the instrument resides in? Noting the room temperature would also be useful. 2) Page 8, Line 3: Is it possible that I^- (or other ions) signals showing peaks at night is also due to evaporation from particles?

Minor comments:

Page 1, Line 20: “we also measured...” what do the authors mean by events?

Page 1, Line 14 and Page 3, line 5: There are two altitudes listed: 3450 and 3454 m a.s.l. Page 4, line 1 gives 3454. Page 2, line 29 says 3580 m a.s.l. Which one is it? Is the mountain growing (this is a serious question)?

Page 1, line 14: it would be useful to note the exact months and year the study was carried out instead of just saying 9 months.

Page 3, Line 10: “with nine months...” not a complete sentence after the semicolon

Page 5, Line 27-28: “we found mostly N-containing...” the phrase “that could not be undoubtedly” is confusing.

Page 7, line 5: “sulfuric acid concentrations” The authors only use signal intensities in this paper. To use the word concentration requires showing a calibration constant of some kind. Also, it should be mentioned how the APi-ToF sensitivity would affect observed signals, i.e. are HSO_4^- ions transmitted/detected more efficiently than MSA ions? Without this knowledge, the reader cannot compare the signals of different ions. This explanation (or statement of the assumption the authors are using) could be added in the instrumentation description.

Page 8, Line 24: change to “During these events other iodine species were also detected, as described below...”

Page 9, Line 6: should be coastal, not costal

Page 9, Line 17: “with ions of sulfuric acid” are the iodine compounds the ions? Or is the sulfuric acid? Also in line 18, $I_2O_2 \cdot CH_3SO_3$, which one is holding the charge?

Page 10, R.10: what is M?

Page 11, Line 16, typo in listing of figures.

Page 12, Lines 11-14: The first sentence suggests they are clusters but the authors conclude they are not. Please explain more carefully why they are not clusters.

Page 13, Line 4: “The latter...” using the word latter means there are only two phrases but there are three. Maybe change to “the last process”

Figure 1: The positive ions have higher signals than the negative ions (amine is roughly 4 times higher than sulfuric acid). The authors state that the amine concentrations must be quite low (page 14, line 22) as amines are not seen in ion clusters. The ion signals say otherwise. Can the authors comment on this? Discussing the efficiency of detection for the APi-ToF would be very useful here.

Figure 2, last line of caption: The comma (or is it a period?) after SO_5^- looks strange.

Figure 3: The colors for HSO_4^- and $CH_3SO_3^-$ look the same on a printed page. Also the units for signal are inconsistent with the other graphs (ions s^{-1} vs ions/s).

Figure 6: the top part of several lines are cut off (e.g., CSI and IO₃-)

Figure 7: What does τ mean on the bottom left map graph in B and D? Also the units need superscripts. What is R?

Figure 8: same comments as figure 7

Works cited in this review:

Bianchi, F., Junninen, H., Frege, C., Henne, S., Hoyle, C. R., Molteni, U., Herrmann, E., Adamov, A., Bukowiecki, N., Chen, X., Duplissy, J., Gysel, M., Hutterli, M., Kangasluoma, J., Kontkanen, J., Manninen, H. E., Munch, S., Perakyla, O., Rondo, L., Williamson, C., Weingartner, E., Curtius, J., Worsnop, D. R., Kulmala, M., Dommen, J. and Baltensperger, U.: New particle formation in the free troposphere: A question of chemistry and timing, *Science*, 352(6289), 1109, doi:10.1126/science.aad5456, 2016.