The manuscript by Brean et al. describes measurements from Beijing during spring/summer 2017. The main instrument deployed was a nitrate chemical ionization atmospheric pressure interface time of flight mass spectrometer (CI-APi-TOF) for the measurement of sulfuric acid and highly-oxygenated organic molecules (HOM). Other measured parameters include the particle size distribution, cluster and nanoparticle concentrations, meteorological conditions and mixing ratios of certain trace gases (SO_2, NO_x, O_3). A proton transfer reaction mass spectrometer (PTR-MS) measured the mixing ratios of isoprene, monoterpenes and C_2-benzenes. The main focus of the paper lies on the description of the observed HOM signals and relating these to observed new particle formation events. One conclusion is that the occurrence of nucleating clusters correlates with peak sulfuric acid concentrations, whereas the peak HOM concentrations occur at a later time and are thus rather related to particle growth than to nucleation. The authors further speculate that dimethylamine (DMA) together with sulfuric acid (SA) could be responsible for nucleation due to the observation of some SA-DMA-containing species. The present study describes the HOM signals in a mega-city environment where both anthropogenic and biogenic emissions are relevant. It is therefore an important contribution because previous studies mainly focused on environments where biogenic emissions dominate (e.g., Hyytiälä Finland or the Southeastern United States). The manuscript is very well-written and structured. It is also very much appreciated that a full list of identified signals from the CI-APi-TOF mass spectra is provided in the supplementary information. One flaw of the present study is that no information on HO_x and RO_2 is provided although the authors state that these compounds have been measured. Furthermore, I think that some more information can possibly be retrieved regarding the relevant nucleation mechanism. Suggestions for further data evaluation in this direction are provided below. Several further specific suggestions for improvements are listed in the following. These should be implemented before publication in ACP.

L27: please define the used acronyms (VOC, BVOC)

L27: It would be good to mention already in the abstract when the data were taken (month and year).

L37: "O_3 is lower on the days with higher HOM concentrations": This sounds as if O_3 inhibits the HOM formation. Can this just be coincidence as there are relatively few days of measurements?

L135: 3 sccm of carrier (sheath?) gas flow for N_2 is very low as this flow is typically on the order of 20 to 30 slm in CI-APi-TOF instruments, please check. In addition, only one unit for the flows should be used (currently Lpm, sccm and SLM are used).

L145: Usually the nitric acid trimer (m/z 188, i.e., $(HNO_3)_2NO_3^-$) yields a rather high signal in nitrate CI-APi-TOF spectra, too. If this signal is not observed it points to rather strong fragmentation of cluster ions. Is the trimer signal missing completely? Furthermore, it is mentioned here that all signals are normalized with the primary ion count rates; however, in the figures this normalization seems to be missing. The statement here also contradicts the statement in L149/150 ("… all values are reported in signal intensity, ions/s.").

L149/150: Rather than reporting signal intensity (ions/s) I highly recommend to report normalized signals in all figures, i.e., the data should be normalized by the sum of all primary ions (m/z 62, 80, 125 and 188, if present). It would also be good to mention that the conversion constant (from normalized counts to concentrations) is typically between a few 10^9 and 1×10^{10} molecule cm^{-3} (see e.g., Kürten et al., 2012). In this way the reader can get an idea of the rough HOM and sulfuric acid concentrations. One further suggestions relates to the fact, that concentrations of SO_2 and OH were measured along with the condensation sink. From these data the H_2SO_4 concentration can be estimated (using a simple steady-state assumption for the main source and the sink of H_2SO_4). In this way, an estimate for the calibration constant can be derived.

L150: It would be good to mention typical values for the mass resolving power and mass accuracy.

L165/166: Please swap the order of the reported size ranges as the LongSMPS is mentioned before the NanoSMPS.

L168 and L170: The term "saturator pressures" is used here; however, in the PSM the saturator flow rates are varied in order to achieve different diethylene glycol supersaturations; this should be clarified.

L172: It is not clear what is meant by "similar behavior of the upper and two lower size cuts". Do the authors mean that the concentrations for the lower and upper two size channels typically correlate very well?

L187: It is mentioned that OH, RO_2 and HO_2 concentrations were measured, yet, none of these data are shown. To my knowledge the present study is the first ambient study where HOM, O_3, OH, HO_2 and RO_2 were measured simultaneously. Therefore, a lot could be learned about the different HOM formation pathways (e.g., if certain HOM originate rather from reactions with OH or O_3). It would be great if somehow the HO_x data could be incorporated in the data analysis.

Figure S1: please show the (normalized, see comment above) H_2SO_4 signals on a log scale

L209: delete one of the "that"

L221: I think some of the signals cannot be unambiguously identified, e.g., the mentioned sum formula could also be written as $C_5H_8O_2(HNO_3)_2$ or $C_5H_9NO_5(HNO_3)$, where the HNO_3 could be coming from the charger ions (i.e., $(HNO_3)_2NO_3^-$ or $(HNO_3)NO_3^-$ rather than NO_3^-). One way to test this hypothesis is to check if the m/z 288 signals correlates with m/z 225 (this could be the same neutral molecule just with one less HNO_3 from the charging process). I also think that this possibility of ambiguity exists for some other nitrogen containing species, which affects the evaluation of the oxidation state values shown in Figure 1. Although the question of ambiguity cannot be ultimately resolved it should be mentioned and discussed briefly.

L245/246: Schobesberger et al. (2015) provide a detailed list of observed signals in the nucleating system of sulfuric acid and ammonia. From their observations prominent signals for

the reported masses (m/z 344 and m/z 362) seem rather unlikely. I would also be surprised if just these two mixed ammonia-sulfuric acid peaks show up in the spectra without any others. Have the authors considered the isotopic distributions of the assigned signals in their analysis? Sulfur has a distinct isotopic pattern; therefore, the assigned formulas in Table S2 for the sulfur-containing species could be checked by considering the isotopes.

L267/268: As mentioned before, it would be great if more information on HO_x and RO_2 could be provided.

L295: the plot does not show concentrations but the raw signals

L344: J(O1D) is not shown in Figure S1

L347: neither O_3 nor HOM are shown in Figure S2

L410: in the PSM particles are grown within the condenser

L411 and L412: Can the authors at least speculate what compounds cause these signals? If they are from (in)organic compounds (H_2O, NH_3, H_2SO_4 and maybe amines) the number of possible combinations should not be too large.

L420 to 430: The possibility of sulfuric acid-amine nucleation should be further discussed. To me it seems very unlikely that only selected SA-DMA clusters show up in the spectra. For nitrate CI-APi-TOF measurements a detailed study of sulfuric acid-dimethylamine clusters has recently been presented (Kürten et al., 2014). That study has also shown that DMA together with sulfuric acid forms new particles very efficiently; therefore, tiny amounts (pptv) should suffice for efficient nucleation and the presence of DMA in clusters is already evidence that DMA is assisting in NPF. I suggest to search for further DMA (or other amine) containing clusters and to check if ambiguity can be ruled out, e.g., that the clusters with DMA and sulfuric acid are not due to some other (organic) compound. This can be done by taking into account the isotopic patterns. In addition, in Table S2 one of the listed clusters is $C_2H_7NHSO_4^-$ (i.e., a C_2-amine clustered with the bisulfate ion). This cluster does, however, not exist as the Lewis base (HSO_4^-) does not form a stable cluster with a strong base (C_2-amine) unless at least two further acids (H_2SO_4) are present in the cluster (Ortega et al., 2014; Kürten et al., 2014).

Figure 2: Is this MD plot corresponding to a period when NPF is occurring? It would be good to show a second MD plot for another day (same time of day) when no NPF is occurring just to see what signals could make the difference. In addition, there seem to be really prominent peaks (negative MD) at m/z of ~500 and ~700. Have the corresponding compounds been identified? Do these signals show a distinct diurnal pattern with higher concentrations during NPF?

References:

Kürten, A., et al.: Calibration of a chemical ionization mass spectrometer for the measurement of gaseous sulfuric acid, *J. Phys. Chem. A*, 116, 6375–6386, https://doi.org/10.1021/jp212123n, 2012.

Kürten, A., et al.: Neutral molecular cluster formation of sulfuric acid-dimethylamine observed in real-time under atmospheric conditions, *Proc. Natl. Acad. Sci. USA*, 111, 15019–15024, https://doi.org/10.1073/pnas.1404853111, 2014.

Ortega, I. K., et al.: Electrical charging changes the composition of sulfuric acid–ammonia/dimethylamine clusters, *Atmos. Chem. Phys.*, 14, 7995–8007, https://doi.org/10.5194/acp-14-7995-2014, 2014.

Schobesberger, S., et al.: On the composition of ammonia–sulfuric-acid ion clusters during aerosol particle formation, *Atmos. Chem. Phys.*, 15, 55–78, https://doi.org/10.5194/acp-15-55-2015, 2015.