Yu et al. present findings from detailed compositional measurements of Arctic aerosol in Svalbard during August 2012. While there is obvious importance of conducting detailed physiochemical characterizations of Arctic aerosol in terms of their radiative impacts and subsequent indirect effects on frozen surfaces, there are major issues with the manuscript by Yu et al. that would need to be addressed prior to publication. These issues stem from possible misinterpretation of the data that shape the reported main findings. It would behoove the authors to provide a sufficient level of detail on the methodologies (including caveats) and results to support the main conclusions they report.

General comments:

There is a scarcity of detail regarding which samples and particles were analyzed. More specifically, which samples were analyzed, which particles were analyzed per sample and how those were chosen, how many particles per sample were analyzed, and why only select samples and particle numbers were analyzed under each method is not at all defined. For example, which of the samples constituted the 2002 and 575 particles analyzed by TEM and TEM/EDS, respectively? For certain techniques, only a few samples (i.e., 3 samples for NanoSIMS but no mention of particle number) or even only a handful of particles (i.e., only 17 particles for AFM but no mention of which sample(s) these came from) were analyzed, and in the case of SEM there is no information on sample or particle number. I understand that some of these tools, i.e., AFM, are time-consuming which is why a low number of particles were analyzed, but then the authors need to be careful about overstating result interpretations. It is important to know how many particles and from which samples to provide sufficient statistics and afford information on daily source variability. As it stands, there is no way to tell how representative the percentages (which are hidden in the text) are of summertime aerosol in general, or just of specific samples from select days.

It would be helpful to provide a figure or two of the overall picture of aerosol composition, e.g., bar graphs or pie charts. There are percentages provided in the text, but showing the relative abundance of each particle type is pretty standard. Along relative abundance, the authors report that 29% of the particles were non-sea salt. What percentage were unclassified? What percentage is the “majority of NSS-particles”? As a result, it is not clear how important these particles are in general in the context of radiative impacts, given sea salt was what seemed like the dominant particle type and also largely affects scattering and SSA. In addition, to demonstrate that these particle types are important for the Arctic energy budget, they should show extinction properties for total aerosol (including sea salt) in Figure 6. I would think given the typical sizes for these types of particles (sulfate and soot) and reported abundance for this particular study, they would not affect the scattering cross sections relative to sea salt. The emphasis on the radiative impacts of these aerosol types is a large part of the manuscript, so their properties need to be presented in broader context. Are they important within the total aerosol population or not? This would be more relevant to the actual atmospheric implications.

There are several issues with the methods as presented. For instance, there is very little detail given on how the particles were classified under each technique, there are no errors or statistical analyses reported, and certain methods have very little specification detail (i.e., SEM is a very short paragraph). Regarding the samples storage, at 20% RH, I would assume all volatile and most semi-volatile species would evaporate, significantly altering particle shape, size, and composition. I will admit, these techniques are not my area of expertise, but the authors should at least comment on potential losses and caveats with this storage method. If there are significant losses of material, how representative are the analyzed particles of the total ambient aerosol population at the time of collection? I am skeptical the authors are comparing apples to apples by possible alteration of particles during storage. Regarding the source analysis, there is very little detail given on the FLEXPART modeling and only a couple sentences on the results and discussion of the simulations. It is used to a very minimal extent and very generally summarized, even though Arctic aerosol

sources can vary drastically day-by-day, and especially given possible local contributions. Figure 5 is very difficult to discern and glean any source information from it. Also, why are only these particular days shown?

There is no background on previous relevant studies conducted at the study location, even though there is a long-term monitoring station with aerosol measurements at Ny-Ålesund (https://www.esrl.noaa.gov/psd/iasoa/stations/nyalesund). It is not the same exact location as the Chinese site, but close enough to at least use those routine, publically-available measurements to provide some broader spatial and temporal context.

There is only basic mentioning of biogenic VOCs, but none of biogenic or biological aerosol. The Arctic summer, especially in remote coastal sites, is largely affected by gases and aerosol from primary productivity due to the availability of sunlight and open water. There is no discussion on if the OM is biogenic/biological, and in general, the definition of OM is vague.

Technical corrections:

There are many typos, grammatical issues, and a lack of necessary explanation (e.g., the 3 sets of bars in Figure 6, SEM a short paragraph with no numbers, etc.).