

Interactive comment on “Compositions and mixing states of aerosol particles by aircraft observations in the Arctic springtime, 2018” by Kouji Adachi et al.

Anonymous Referee #1

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Review of Adachi et al., ACP, 2020

This manuscript evaluates particle samples collected by impaction upon electron microscope grids and analyzed by transmission and scanning transmission electron microscopy with x-ray compositional analysis. The samples were collected on an aircraft in the Arctic in springtime, when the Arctic haze phenomenon is a maximum.

The methodology and analysis is very clear, and provide useful information on the mixing state, morphology, and size-dependent composition of the aerosol in the Arctic in springtime. The complexity of the aerosol is surprising, with mineral dust, sea-salt, soot, fly ash, and dust particles adding to the more widely recognized sulfate-organic

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mixtures in this region. The manuscript is a carefully written and useful addition to the literature on Arctic haze, and should be suitable for publication following minor revision.

Below are major comments, followed by technical issues.

Major comments:

1) One overall disappointment is the relative lack of integration of the electron microscopy data with on-line aerosol instruments on the aircraft. For example, according to the project website, an aerosol optical particle spectrometer was operated on the aircraft during PAMARCMiP 2018, and an SP2 instrument provided information on black carbon abundance and coating thickness. Instead of combining these measurements with the TEM and STEM data, the microscopy results are analyzed and interpreted alone. Online single particle mass spectrometers, which provide statistical information on the size resolved mixing state and composition, are being combined with independently measured particle size distributions to provide a more quantitative description of the aerosol (e.g., Froyd et al., 2019; <https://doi.org/10.5194/amt-12-6209-2019>). For future publications (not for this one), I urge the authors to consider blending their very useful, but quantitatively limited, compositional data with online techniques to place the results on a more quantitative footing.

2) In the abstract, and elsewhere in the manuscript, the Arctic aerosol is described as being "internally mixed". However, it's clear that there are really separate aerosol types—sulfate, sea-salt, mineral-dust, K-bearing, and carbonaceous—that were in different particles. This is the definition of an external mixture. I think the authors mean to say that all of the types were coated with sulfate/organic materials, and that some particles were composed of two or more different compositions that had coagulated. But definitely one could NOT say that the aerosol was composed of a single, internally mixed composition.

3) Line 123. Is the area-equivalent diameter just the observed diameter as viewed on the microscope grids, or is there some reconstruction to a three-dimensional form from

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the (flattened?) images from the microscope? Please clarify.

4) Line 139. The classifications based on fractional elemental composition seem non-specific. For example, a particle could be considered both "carbonaceous" if it has a C + O weight percent >90%, but also a "sulfate" particle if it also has a S weight percent >2%. In fact, I'd expect all carbonaceous particles to have some significant S component. Can you comment on the fraction of particles that could be ambiguously classified, and how these are resolved by your scheme?

5) Lines 168-170. How does the model represent particulate components? Is it a bulk (mass) model, or does it have a binned or modal representation of the size distribution? A little more detail (a couple of sentences) here would be very helpful.

6) Line 299. Groot Zwaftink et al., 2016 (<https://doi.org/10.1002/2016JD025482>) report on local sources of dust in the Arctic, so your data are very pertinent to their hypothesis that Arctic dust emissions are substantial.

7) Lines 320-322. Is the number of "tarball" particles on each grid correlated with the number of potassium-rich particles on each grid? This would provide a useful link to biomass burning as a source of the tarballs.

8) Section 3.7. Are these particle types counted in the "other" category in e.g., Fig. 3? I'm a little confused between the detailed description of complex particle inclusions and blended types in Section 3.7 with the very discrete particle categories discussed elsewhere in the manuscript.

9) Lines 358-359, how does the soot number fraction compare with values from the SP2 instrument?

10) Please provide 2-sided linear regressions and slopes to Fig. 9. It would be better to show molar values rather than weight percents.

11) The figures are very nice and clear!

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Technical comments:

a) Line 42, change to "However, air pollution over the Arctic, named 'Arctic haze', due to"

b) Line 50, change to "source regions and to altitudes above the polar dome. . . ."

c) Line 64, remove hyphen between "soot" and "mixing".

d) Line 87, the acronym PAMARCMiP is already defined in the abstract.

e) Line 126-128. I don't understand this sentence regarding AED and mixing state.

f) Line 130, change "element" to "elemental"

g) References. The formatting of the references is inconsistent, and also not consistent with Copernicus guidelines. For example, journal names are not abbreviated, and some entries, such as Buseck et al., have capitalized the paper titles. Please review and manually correct; don't rely on your reference manager software.

h) Fig. 2 caption. These are normalized size distributions, not raw number concentrations.

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