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Comment on acp-2021-94

Anonymous Referee #2

Referee comment on "Differentiation of coarse-mode anthropogenic, marine and dust particles in the High Arctic islands of Svalbard" by Congbo Song et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-94-RC2, 2021

Comments to the Authors

This study focuses on coarse-mode aerosol particles and discusses their possible sources based on cluster analysis, aerosol chemical composition analysis, and air-mass backward trajectory analysis. The authors categorize the coarse particles into 5 clusters based on the difference of the spectra of their number-size distribution, and then suggest that the clusters C1 and C2 (27%) and the clusters C3, C4, C5 (73%) are attributable to anthropogenic and natural origins, respectively. Because previous studies have overlooked the possible importance of coarse-mode aerosols in the Arctic, the datasets presented here would be valuable for the science community. However, the methods and discussion on the source of coarse-mode aerosols might be biased, because there are some major problems with the methodology. As a result, it is hard for me to believe the percentages presented here (e.g., Lines 5-10) are indeed valid. I would like to ask the authors to answer the following comments and clearly explain the possible biases of their approaches.

Major Comments

- Based on the cluster analysis of the number-size distributions of coarse-mode aerosols (0.5 to 20 um), the authors divided them into 5 categories. I can agree that this categorization is one of the reasonable approaches for understanding the characteristics of spectra of their number-size distributions. The authors also explained that the percentage of the clusters C1, C2, C3, C4, and C5 are 9%, 18%, 34%, 32%, and 7%, respectively (Lines 8-10; Table 1). However, it seems that there is a large seasonal bias of the available data (Figure 1). In particular, the wintertime data are very lacking. The authors would need to explain the possible influence of this bias on their cluster analysis.
- Although the authors describe that they have used the aerosol chemical composition data from several online and offline measurements (Lines 5-6), these datasets are

mainly based on ion chromatography (i.e., water-soluble particles) and PSAP (i.e., black carbon). For this reason, the results presented here would underestimate the contribution of "insoluble" particles to the population of coarse-mode aerosols. For example, although they discuss the contribution of mineral dust particles based on Ca content (Ca/Na ratio), Ca-containing components (e.g., calcite, dolomite) are not necessarily major mineral components. Actually, the mineral composition analysis of mineral dust particles in high latitudes using SEM/EDS and/or XRD analysis (e.g., Moroni et al, 2015; Tobo et al., 2019; Sanchez-Marroquin et al., 2020; Adachi et al., 2021) has shown that mineral dust particles contained few or no Ca components. Also, the presence of insoluble organic particles and biological particles (e.g., bacteria, fungal spores, pollens) has been reported by filed measurements at the Gruvebadet and Zeppelin Observatories in Svalbard (Geng et al., 2010; Tobo et al., 2019). For these reasons, additional analysis of insoluble particles would be required if they would like to quantify the dominant aerosol types in each cluster (C1 to C5).

- I doubt if each category was indeed characterized by anthropogenic or natural sources. As far as I check the results in Figures 4 and 7, each cluster were likely to be influenced by both anthropogenic and natural origins in most cases. For example, the aerosol population in the clusters C1 and C2 might be largely influenced by Arctic haze (Lines 244-261); however, I cannot think that the influence of natural aerosols on the population of coarse-mode aerosols was negligibly small. Although the clusters C3 and C4 are categorized as sea spray aerosols (Lines 262-291), a case study at the same site (Gruvebadet Observatory) in summer (Geng et al., 2010) demonstrated that both sea salt and mineral dust particles were always detected in the same samples. It is also skeptical that the cluster C5 can be identified as mineral dust only (Lines 292-300).
- There would be some problems with the use of backward trajectory analysis of air masses. Although the authors discuss the possible source of BC, sea spray, and/or mineral dust based on the trajectory analysis, their emission in high latitudes would be largely altered by the seasonal variability of snow/ice cover over land and sea. The authors would need to check the variation of snow/ice cover as well as air mass history (Figure 8). The explanations presented in Lines 230-239 are insufficient. In addition, the trajectory analysis might overlook the possible contribution of local sources (namely, Svalbard and its surroundings).

Specific Comments

- Line 30: Where is the Zeppelin Observatory?
- Figure 3: Was there no difference in the diurnal variation of aerosol volume size distribution between the periods of polar day (summer) and night (winter)?
- Lines 206-208: It was hard for me to understand this explanation. Why did you think that "the polar plots show that high concentrations of the five clusters were typically found when wind speed was higher than 5 m s^{-1} (Fig. 3a), indicating limited impact of local emissions on coarse-mode particles at the GVB station except for wind lifted particles"?
- Sections 4.1.1 to 4.1.5: The title of these sections would not be appropriate. I would recommend the titles like "4.1.1 Possible sources of C1" or something like this. In addition, more detailed discussion on possible sources of each cluster would also be required in each section.
- Lines 281-282: Why do you think that the main difference of C4 relative to C3 is "less source contribution from the open ocean (Fig. 8d)"? It seems that most air masses originate from the North Atlantic Ocean, which is rarely covered with sea ice during all seasons.

References

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