Review of the ACP manuscript acp-2017-278
"Sub 500 nm refractory carbonaceous particles in the polar stratosphere"
by K. Schütze et al., 2017

The above manuscript deals with electron microscopy (TEM, SEM) analysis of stratospheric particles sampled mainly in the Arctic polar vortex. As such measurements are rare, the presentation of the measured data is well suited for ACP, even if the results are not totally conclusive. However, there are some points, which should be improved before publication, some work, but feasible.

General remarks:
The first thing, which immediately leaped out at me when reading the abstract, was the big difference between the time of sampling and the time of publication. The samples were taken in 2000, now we have 2017. When was the analysis done? If it was in recent years, how where the samples stored in-between? How might the particles have changed during this long storage time? If the analysis was performed shortly after sampling, why did the publication take so long? The authors have to address this issue in a new paragraph.

Secondly, concerning the samples, section 2.2. There are 11 samples, OK, but I got confused how many particles where analyzed with which method. Were some particles analyzed with both methods? Moreover, on page 4, line 21 you even mention STEM, which is not mentioned somewhere else. Was this an additional method? Then it should be listed in the abstract as well. To make it easier for the reader to understand what you did and not to put too much workload on you, I suggest to include another table, where the reader gets an overview how many particles where analyzed with which method (and detector).

Another point, there are many statements in the manuscript, which are not specific enough. This occurs quite often, when citing the literature (which might not be the fault of the present authors, maybe the original authors were not specific enough). I tried to list some examples of that below. Please have a look throughout the manuscript and improve the text.

Finally, there are another two important issues (measurement artifacts and particle aging), which are explained in the following section in detail.

Specific remarks:
Abstract:
- p. 1, l.15: "… approximately 28-82% of the particles are refractory carbonaceous ..."
This statement is not very specific. You can nearly find all fractions of refractory carbonaceous particles, well OK, everything is possible, not very useful, but how likely is that? Moreover, isn´t the range much smaller, 52-82% (Fig. 3), considering that sample G seems to be a special one?

- p. 1 l. 17: "20-830 nm" this contradicts the manuscript title, i.e. the "500 nm"

- p. 1 l. 21: The ratios to C: It would help the reads imagination if you would provide the ratios as fraction, i.e. instead of for instance "0.001" use "1/1000". Same for the detection limits in section 2.2. As the first place is the abstract, showing your major findings, you should

also asses the meaning of these numbers, are they common or rather rare, what do they indicate, etc.

- p. 2 l. 25: Ebert et al. 2016, from the same group, what are the similarities, what are the differences between this paper and the current manuscript? It should be possible to compare the results.

- p. 3 l. 4: redistribution vs. sedimentation: Currently your statement reads like an exclusive "or", but both processes can happen to a specific trace gas, it can be redistributed and be removed by sedimentation, or?

- p. 3 l. 6: In this paragraph, you list a bunch of sources for stratospheric refractory particles. However, the reader does not know, which one is the more important (e.g. with respect to mass or frequency of occurrence). Could you please provide the reader with such an additional information.

- p. 4 l. 2: It is stated that the vortex was stable between mid-January and mid-March. This was exactly the time of sampling and you should mention here that the presented data are from this period.

- p. 4 l. 11: the selection criteria "substrate area covered by particles", what does it mean? I´m not an electron microscopy specialist.

- p. 4 sect. 2.2: The two silicon-EDX detectors from Oxford, are they the same? Once Oxfordshire, once Wiesbaden? Isn´t it the same company?

- p. 5 l. 10: Not being an electron microscopy specialist: what would you expect the scattered electrons do? Hit the housing and generate x-ray emissions there? Please clarify.

- p. 5 l. 17: "small but systematic differences". Please specify what "small" means, e.g. give a percentage range. Same for line 21.

- p. 6 l. 15: "all particles" in the world? The stratosphere? On a sample? Which diameter does the spot have?

- p. 6 l. 25: For me, sample G seems to be special. Did you check how the sampling conditions of sample G are compared to the other samples?

- p. 7 l. 1: In Fig 4, sample E and F show a very different distribution width. Did you check for reasons?

- p. 7 l. 7: The minor components you have found (Fig. 5): From the literature (e.g., Murphy et al., AS&T, 2004; Martinsson et al., AMT, 2014) it is well known that ice crystals hitting the aerosol inlet can remove inlet material, bring it into the air and thus can generate artificial particle signals. Fe and Ni are known for this. Did you check the correlation between the occurrence of these elements with the ice crystal number concentration or the sampling time spent in ice clouds? This is **important** and must be addressed in order to trust your data.

- p. 7 l. 24: For how many particles was this element distribution imagine done? Fig. 6 shows just one. Is there any statistics on the results of this analysis?

- p. 7 l. 25: I do not know how the element distribution images work, hence I do not know what “measuring several” (how many?) “points on the particles” means. Please explain this more in detail.

- p. 7 l. 31: Why did you generate these four groups? If I did not overlook it, they are not used afterwards.

- p. 9 l. 22: The differences between your study and the results in Nguyen et al., 2008 is likely due to the different atmospheric measurement regions and different measurement altitudes. You should mention that, otherwise the reader might take the Nguyen reference as a contradiction to your findings, which is, in my point of view, not the case.

- p. 10 l. 11: Pyro-convection is defined as fire-started or fire-added convection, hence the definition given by you is incomplete.

- p. 11 aircraft exhaust section: The Mazaheri et al. reference here, and later on also the Tumolva et al. and Torvela et al. references in the wood burning section, here you compare freshly emitted particle properties to your particles, which are, because they were measured in the polar vortex, likely more than one year old. This comparison can only fail, the particles aged and strongly changed. I miss this time effect in all potential source paragraphs, but this point is **important** and must be considered in the discussion section.

- p. 11 l. 26: Consider to add “(dominant meteorite fraction)” or something similar after “chondrites”, in order to explain what this thing is.

- p. 15 l. 1: The summary is too short. You did a lot of work, please expand the summary.

Technical corrections:

- p. 1 l. 29: Please remove the empty line, the last sentence of the abstract belongs to the upstream paragraph and should not be separated.

- p. 2 l. 3: “sulfur” is an “element”, not a “component”.

- p. 2 l. 13: which “groups” where identified? “Particle” or “morphology” or …

- p. 2 l. 15: “a large refractory particle load”, what does this mean? With respect to particle mass or particle number or just fraction of particles containing refractory material?

- p. 2 l. 20: “widely distributed”, what does this mean? All over the globe? Or at all altitudes (which ones?) in the area of investigation (which was?)?

- p. 2 l. 22: I´m not a native speaker, but shouldn´t it be “Earth´s”?

- p. 2 l. 31: “condensation of saturated gases”, it is not necessary to provide seven (!) references for this textbook process. As it disturbs reading the paper, you should reduce the number.

- p. 3 l. 25: Please insert a comma after “impactor”.

- p. 3 l. 26: Please remove the “The” before “MACS”.

- p. 3 1. 32: “It was weaker …” What is “it”? The “Arctic winter”? But then the sentence does not make sense.

- p. 4 1. 4: Please use “Θ” instead of “PT”.

- p. 4 1. 20: Please move “software” before the brackets.

- p. 4 sect. 2.2: Please use “EDX” instead of “energy-dispersive X-ray” throughoutly, after you defined it once.

- p. 6 1. 29: The whole statistical analysis section reads like a bullet point list. Please make it more a coherent text or a real bullet point list, with an introductory text.

- p. 6 1. 7: Please remove “applying a significance level of 5%”, this is redundant, as it is mentioned in the last sentence of this paragraph.

- p. 6 1. 15: Please move the comma after “(Figure 1)”.

- p. 7 1. 1: Please use “indicated” instead of “shown”, you do not show real particle size distributions, e.g. dN/dlogDp.

- p. 7 1. 6: Please move “besides C” to the beginning of the sentence.

- p. 7 1. 25: Please replace “contained in the whole” with “found everywhere in”.

- p. 8 1. 18: Please replace “The samples” with “All samples”.

- p. 10 1. 27: Please replace “emissions” with “eruptions”.

- p. 11 1. 15: A space is missing before “The”.

- p. 11 1. 16: Please replace “at” with “in”.

- p. 12 1. 27: “comprised … to” sounds strange, better use “contribute …to” or something similar.

- Fig. 2: Please specify Kα and Kβ in the figure caption. What does “all particles” mean? In the stratosphere or all sample or all refractory? Is the peak height/area linearly representative for the number of atoms? This should be mentioned somewhere in the text.

- Fig. 3: The given particle numbers are the total number of analyzed particles or only the refractory ones? Please specify this “n” in the figure caption.

- Fig. 6: The colors in the lower row of pictures are hard to see. I believe to use bright red or even white as occurrence indicator color in all pictures would improve the figure.