Interactive comment on “Chemical composition of free tropospheric aerosol for PM1 and coarse mode at the high alpine site Jungfraujoch” by J. Cozic et al.

Anonymous Referee #2

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This paper summarizes results from long term measurements of aerosol chemical and physical properties at the high altitude site of Jungfraujoch, Switzerland. While a synthesis of data of the kind reported in the paper is appropriate and should be published in a journal like the ACP, there are some particular problems with this particular paper that need attention before the paper is accepted for publication.

There are too many details in the paper that have been broadly brushed over; some of the details need to be examined for subsequent data discussions. In particular, the comparison between OC from AMS and OC from the Sunset analyzer has many problems (as outlined further below). Yet the results of the comparison were used to
derive data for further presentation and discussion in the paper. Another example is the use of the EC data from the Sunset analyzer as a surrogate for BC to estimate the absorption coefficient. As pointed out in detail below, BC and EC are different in concept; BC really should be referred to as an \textit{equivalent black carbon}; but really the absorption could be due to many absorbing materials including soil dust and HULIS in addition to black carbon. Calculations based on the calibrated absorption coefficient are prone to errors and these errors should be discussed in detail in the paper.

Specific comments are listed below.

Abstract: p. 12146, line 14. "...while calcium and nitrate were the two major measurement components..". Both components seem quite small to me.

p. 12147. line 3. "..dependant." should be "..dependent."

p. 12147, line 6-7. Clarify "Size discrimination into ... is often applied." I would say "Aerosols are often separated into a coarse fraction..and a fine fraction during sampling."

p. 12147, line 15. Comment: DMS is often oxidized first to SO2 and then onto sulfate. How do you distinguish aerosols through SO2 from DMS and SO2 from anthropogenic sources?

p. 12148, line 23. Add "site" after "Jungfraujoch"


p. 12149, line 25. Comment: what are the uncertainties in the BC measurements that arise from other absorbing materials in the aerosols? This may be quite significant in cases where dust particles are present, or that HULIS are present.

p. 12150, line 9 and 10. How are the absorption coefficients determined and are they relevant to the ambient aerosols measured at the site?
p. 12151, paragraph 1. This calibration procedure only calibrate the response of the NDIR detector. The conversion efficiency and sampling efficiency are all ignored and uncalibrated.

p. 12151, line 27. "two size classes" Typically they are referred to as "two size fractions" in most literature.

p. 12152, line 1. "two size classes" See comment above.

p. 12152, line 2. "scientific advisory group for aerosol" Should these be capitalized to refer to a specific organization?

p. 12152, line 8. "treated with high-vacuum grease to prevent bouncing" Does this introduce organics into air stream from blow offs?

p. 12152, line 11. "the particulate matter" You need to be consistent with terminology. Either use "aerosols" or "particles" here to be consistent with the rest of the text.

p. 12152, line 12. "for collection of nitrate," Is this for the gaseous nitrate from the particle evaporation? Should be "for collection of gaseous nitrate, e.g., HNO3,"

p. 12152, line 19. What is a "clean room 0.1"?

p. 12152, line 26. Should "MSA gradient" be "MSA eluent"?

p. 12152, line 29. Ditto for "KOH gradient"

p. 12153, line 4. Change "samples" to "sample"

p. 12153, line 14. d from AMS is really "vacuum aerodynamic diameter, or dva" and can be substantially smaller than geometric diameter.

p. 12154, line 2. "vaporize" should be "vaporizes"

p. 12154. section on AMS. The reduce ambient pressure at Jungfraujoch can impact the reported concentration and size distribution measurements of the AMS. Has this been properly addressed?
p. 12155, line 5. morphology can also affect the SMPS measurements.

p. 12156, line 16. To use EC for BC is not appropriate. In addition to EC, there are potentially other chemical components that can absorb light. In particular, some soil dust minerals can absorb light. At the very least, some discussion on the uncertainty this substitution may have caused should be given here.

p. 12156, line 19. "may be partly caused by" Not "may be", but certainly "is"

p. 12156, line 21. reference not quoted properly.

p. 12157, line 6. Give references for the orthogonal regression technique.

p. 12157, line 10. Would not the coating be more likely in summer than winter due to stronger photochemistry? If true, should the absorption coefficient be smaller in summer?

p. 12157, line 15. Be specific about the differences in procedures and how these differences can account for the observational differences.

p. 12157. line 25. Are the filter samples TSP or fine fraction samples? Need to clarify because AMS only samples submicron particles.

p. 12128. comparison of AMS OC with OC from Sunset analyzer. Sunset analyzer OC includes OC at high temperature 615°C; whereas AMS measures OC typically at 550°C (unless specifically set to 615°C, in that case measurements for other components such as SO4 and nitrate must be treated with different fragmentation tables; this is not clarified in the text). Also, it appears that OC/EC from Sunset analyzer is for TSP, whereas AMS is for submicron particles only.

p. 12159, line 2. Again, this conversion factor can be problematic because of the issue above. See comments above. At least some uncertainty must be included.

p. 12159, line 8-9. "this is not expected to have strong influence at the JFJ." Unless there is little or no OC in particles > 700 nm, this is not true.
p. 12159, line 9. I have a real issue here with using the same coefficient for the winter and summer, for the reasons outlined above. Uncertainties are not given and no confidence can be gained for the extrapolation.

p. 12159, line 27. Why not use the AMS total mass values to derive the effective density of PM1?

p. 12160, line 9 to 15. There is no reason that PM1 and TSP should be correlated.

p. 12160, line 24-25. What about Len Barrie's work from the Arctic? As far as I know, that data series have been published many times and the data date back to early 1980s and are continuing.

p. 12161, line 17. Work by Barrie and Sirois (JGR) should be cited for long term trends at the GAW site at Alert, Canada.

p. 12161, line 21 to 23. From a time series sampling point of view, the discrete sampling of 1 in 6 should not mask periodicities of 12 days or longer; and thus this argument is not true.

p. 12162, line 9. To me this ratio of 80% indicates acidic aerosols; if the acid is on the surface then the proton concentration can be quite high on the coating. This is plausible since one does not expect to see high NH3 up on the mountain to neutralize all the acidities due to sulfuric acid in aerosols.

p. 12163, line 6. This justification depends on the particle size distribution variation at the location. This is a very bold and unjustified JUSTIFICATION.

p. 12163, line 25 to 27. The problems here are that (1) timing seems different and (2) PM1 has larger ND than coarse mode.

p. 12164, line 1. Change "can not" to "cannot"

p. 12164, line 18. A correlation coefficient of 0.57 cannot be stated as a "good correlation". One has to give the confidence level for the correlation (probability of no
correlation) based on the number of data points.

p. 12165, line 2-3. "area above UK"? north of UK perhaps? Was there any vertical mixing while the air mass traverses France and UK? Afterall, 2000 agl is not a lot of vertical distance.