This paper presents airborne Sun photometer measurements acquired from the NASA P-3 aircraft over Alaska, Canada, and California during the ARCTAS mission. The paper focuses on the Angström exponents derived from this instrument and the use of these measurements to investigate aerosol type and infer FMF fraction. Comparisons with airborne in situ measurements are also presented. The AOD measurements are also used to investigate horizontal variability of AOD and how this varies with location.

The paper provides analyses of a new airborne Sun photometer dataset collected over Alaska and Canada where such measurements have been sparse. The paper uses techniques published earlier and applies these techniques to this new dataset. The paper is generally well written. Most of the figures are satisfactory, but there a few noted below that are much too small and must be clarified to understand what is plotted.

My recommendation is to publish after the authors satisfactorily address the comments listed below. Most of these comments are minor. There are two particular areas that should be addressed. The first is the impact of cloud contamination and cloud screening should be discussed in much more detail. This is barely mentioned in the paper. Given the prevalence of ice particles over Alaska during April, there is a good likelihood that these ice particles will interfere with the AOD measurements. Also, with the very high AODs associated with the smoke over Canada, it is not clear how cloud interference can be completely removed given the large spatial and temporal variability of AOD which would mask the cloud variability. The second is the retrievals of fine mode fraction (FMF) from the AOD spectra. The authors use the method of O'Neill to derive this. This is fine, but there is no mention of how this compares with previous studies (which included some of the authors) in using methods described by Anderson et al. (2005) and Redemann et al. (2009). The differences between these methods should be described in this paper.

1. (page 1, line 5) This lists the 499 nm Angström exponent. Typically the Angström exponent refers to a pair of wavelengths or a range of wavelengths. The range of wavelengths should be indicated here.
2. (page 1, line 5) The 499 nm Angström exponent is listed as 1.4+/-0.3 but the value between 2-4 km is listed as 1.6-1.8? Why not be consistent in indicating these values? Why not indicate that the value between 2-4 km is 1.7+/- 0.1?
3. (page 1, line 7) Presumably "this" refers to the Angström exponents in the altitude regions mentioned in the previous sentence. However, it is not at all clear how these values indicate that the aerosols in these layers are from anthropogenic emissions and biomass burning. This seems to imply that pollution and biomass burning aerosols can have only these values of Angström exponent and this is not conclusively demonstrated.

It may be true that the AMS and black carbon measurements indicate this; if so, the wording should be changed to indicate that these in situ measurements are the prime reason for making the statement about the type of aerosols in these altitude ranges.

4. (page 1, line 8) Should be AOD spectra.
5. (page 1, line 15-17) This sentence describing comparison between AERONET and P3 AOD measurements should indicate the wavelength (500 nm?) corresponding to the 0.02 difference and the direction of the difference (which is higher?).
6. (page 1, line 13-15). Likewise, when indicate the rms difference in FMF, should indicate which measurement is higher or lower.
7. (page 5, line 10-11) There is not enough information that describes how cloud screening was done. This is of particular concern for the cases in Alaska during April 2009 when there were many times when ice particles were present. These ice particles were apparent in other ground based and airborne datasets, are especially apparent in lidar data. For the Sun photometer measurements, setting a criteria to screen out all the ice would likely result in removal of also much aerosol; likewise, relaxing the criteria to allow more aerosol measurements would also likely allow ice to be present. The authors need to discuss in much more detail how the cloud screening was done and provide some discussion about the potential presence of ice. The impact of ice would likely be much larger than the sources of error listed on page 6.
8. (page 10, last paragraph) What is the correlation between Angstrom exponent and organic mass fraction? Likewise between Angström exponent and pollution? The paper currently does not provide enough information to show that there is a high correlation. The paper leads one to believe that smoke was present above 2 km and pollution below 2 km; however, there were occasions (ex. April 19) when smoke was observed by surface instruments at the DOE ARM NSA site and so smoke is not necessarily confined to 2-4 km.
9. Figure 3a, 3b are way too small. I could not read the legends in these figures (even with my reading glasses.) Consequently, I can not determine what is actually plotted in these figures.
10. (page 11, line 24) replace us with NASA P-3.
11. (page 12, line 9) When AOD at 499 nm changed by 2.6, was this due to vertical variability or horizontal variability of the smoke?
12. (page 12, line 23-24) What was the source of the dust? What other information or data are present to corroborate this inference of dust?
13. (page 13, line 8-9) This analysis does not account for cloud or ice contamination.
14. (page 14, line 5) What is the horizontal distance corresponding to 5 seconds?
15. (page 14, line 18) What were the values of particle hygroscopicity?
16. (page 14, Figure 14b) Why not plot both as either AOD or extinction?

17. (page 16, line 24-25) What was the top altitude of the AOD profile?
18. (Page 19, line 13-14) The analysis uses O'Neill's method to derive FMF from spectral AOD. It would be good if this paper had also used or at least commented on the methods of Anderson et al. (2005) and Redemann et al. (2009) in the use of AOD spectra to infer FMF. These authors had developed empirical relationships to infer FMF using AATS-14 data; it would be good to know how well these relationships would work in these studies.
19. (Figure 8) Both Fig. 8a and 8b show outliers. What do these represent? How were they deemed outliers?
20. (Page 23, lines 11-21) This is also relevant to determine whether a proposed satellite sensor needs to have very high resolution or whether lower resolution would be sufficient.

References

Anderson, T. L., Wu, Y., Chu, D. A., Schmid, B., Redemann, J., and Dubovik, O.: Testing the MODIS satellite retrieval of aerosol fine-mode fraction, J. Geophys. Res., 110, D18204, doi:10.1029/2005JD005978, 2005.

Redemann, J., Zhang, Q., Livingston, J., Russell, P., Shinozuka, Y., Clarke, A., Johnson, R., and Levy, R.: Testing aerosol properties in MODIS Collection 4 and 5 using airborne sunphotometer observations in INTEX-B/MILAGRO, Atmos. Chem. Phys., 9, 8159-8172, 2009b.