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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Properties of biomass burning aerosol mixtures derived at fine temporal and spatial scales from Raman lidar measurements: Part I optical properties" *by* Lucja Janicka and Iwona S. Stachlewska

Anonymous Referee #2

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The manuscript introduces a dataset of lidar observations of aerosol backscatter, aerosol extinction, and aerosol depolarization at multiple wavelengths plus relative humidity, for a heat wave event in Warsaw comprising many identified aerosol layers over two nights. It provides aerosol type identification for these layers and gives the mean lidar properties for layers of each type.

Unfortunately, most aspects of aerosol type analysis are not clear and there are many results with inadequate support or no support whatsoever. The overall motivation or objective of the manuscript also seems confused. The lack of clarity about the objec-

tives and especially the flaws in the analysis make it impossible for me to recommend the manuscript for publication in ACP.

Before getting into the details, I will say that I agree that the dataset itself (as distinct from the aerosol type analysis) is potentially quite valuable, and I am particularly impressed by the inclusion of relative humidity profile information which adds significant potential for science value when combined with the lidar aerosol measurements. One option might be to make a streamlined manuscript without the aerosol type analysis, but with a more in-depth uncertainty analysis for the primary lidar measurements (backscatter, extinction, intensive parameters, and RH), plus an explanation of the quality control, and submit this alternate paper as a data paper in a data journal such as Earth System Science Data.

I will organize my more detailed comments by manuscript section.

Section "Introduction":

My primary criticism in the introduction is that it does not adequately state the objective and motivation. The key statement of the objective in the introduction seems to be at page 2, lines 30-33, "The algorithms that deal with the inversion problem of microphysical retrieval require an accurate aerosol layer selection and a high quality 3beta + 2alpha optical data set as well as the depolarization information [Veselovskii et al. 2002; Bockmann et al. 2005; Muller et al. 2016]. Thus the manual data evaluation which allows for an insightful analysis of the lidar signals with the individual approach to the considered case is still much-needed." In this study, the high quality 3beta + 2alpha optical data set and the QA on that dataset is provided by the retrieval algorithm which is considered to be an input to this study and not part of the study itself. Similarly, although the layer selection is done here, the explanation of how it's done is presented as a black box. The statement is a non-sequitur if the "insightful analysis" is taken to mean the aerosol typing analysis, since that is not needed for microphysical retrievals. I also don't understand why automated aerosol typing algorithms are acknowledged but discarded. In short, I just don't understand the motivation for doing this study this way.

A less important point is that there is a lot of information in the introduction, such as the first several paragraphs about the effect of aerosol properties on radiative forcing, whose relevance to this study is not explained at all. On a positive note, I appreciate the introduction to the specific heat wave event being studied, and also the statement about the uniqueness of the dataset.

Section "Methodology":

The methodology describes the retrieval methodology for obtaining the measurements of e.g. backscatter and extinction but not the analysis methodology. Most of the analysis in this paper is about aerosol type attribution, but there is no description of the methodology for this (here or anywhere in the paper), leaving me completely without a foundation for understanding the rest of the paper. For a few other key aspects of the analysis, the hints in the methodology are inadequate, for example "The sub-layers selection was based mainly on RH and extinction profiles". This is too vague to be useful for understanding how the layers are selected.

Also, I think it is important to understand the uncertainties on the lidar measurements. Page 5, line 30 says the uncertainties for the intensive parameters are propagated from the backscatter and extinction uncertainties, but does not say how the backscatter and extinction uncertainties are calculated, and anyway, it doesn't seem that the uncertainties are actually presented in the paper. Error bars in the figures and tables seem to be just the standard deviation calculated for selected layers, not measurement uncertainties. Standard deviation might be a reasonable stand-in for measurement random uncertainties, but for some of the analysis, such as understanding quite small particle depolarization values, the authors need to develop an understanding of the expected systematic uncertainty.

The statement "The columnar value of the extinction related Angstrom exponent of 1

was assumed for the extinction coefficient profiles calculations" confuses me. I don't understand why you would need this assumption and it seems that this assumption would have too much impact on the layer angstrom exponents. I think I'm probably just misunderstanding what this means. I would like to be able to read a reference about the retrieval to be sure, but I can't find one cited. Did I miss it?

Section 4.3 "Aerosol source analysis"

On first read-through of the paper, I thought the aerosol source analysis was meant to be a major part of the analysis. There is a significant amount of analysis to calculate and collate the backtrajectories in an attempt to identify source regions, and this could potentially be very informative in the inference of aerosol type and source for the observed layers. However, the analysis stops far short of that. There is very little attempt to link the backtrajectories to specific layers and the two segments of analysis (aerosol source analysis vs. aerosol typing) seem to be separate, disjointed sections.

There are also several confusing or contradictory statements. For example, page 8, lines 14-15 say where two-days old smoke would have come from but then lines 17-19 says that above 3 km, it's about 3 days old and below 3 km, it's older; so where is the 2-days old smoke? Then at line 20 is a statement that aged biomass burning aerosol is possible in the lowermost levels but at line 22, it's "moderately fresh". I'm not sure if all of this is actually contradictory or if the authors are expressing the thought that there are multiple possibilities and a lot of uncertainty. If there are any layers at all where the airmass analysis allows for assessing probable sources or a range of ages of the aerosol layer, it would be very helpful to see that information included explicitly in Table 1 where it can be easily used for understanding the aerosol types.

Section 5 "Optical properties of the BBA and pollution mixtures"

In this section, individual aerosol layers are labeled according to aerosol types. In a few cases, the backtrajectories are referred to, but mostly the methodology seems to be based on haphazardly comparing the aerosol intensive parameters to a few case studies in prior literature. There is no discussion of how exactly this is done, no consistent thresholds are presented, and there is no quantitative comparison with prior literature that present ranges of parameters that might be expected for different types (e.g. Muller et al. 2007, Gross et al. 2012, Burton et al. 2012).

Unfortunately, there are many, many examples of type labels being applied without adequate explanation, support, or comparison:

Page 9, line 29: “properties typical for the aged biomass burning aerosol (CRLR >1)”. No support is given for this value being “typical” for aged biomass burning aerosol. Although it is not cited in reference to this statement, I believe this argument follows from the case studies presented by Nicolae et al. (2013). However, since those are just a few case studies, it may be an overgeneralization to describe this relationship as “typical”.

Page 10, line 22: “The relatively low LR and high AE . . . indicates pollution domination.” No reference is given for LR and AE of pollution or a description of how to determine them.

Page 10, line 22: The low lidar ratios that are said to indicate pollution seem very low compared to prior literature that I know of. No references are given here. How do these values compare to other published values for pollution cases?

Page 10, line 24, "relatively high value of 532 nm particulate depolarization of 4.8 plus or minus 0.3 ... may reflect contamination by pollen". This is not a very high value of particulate depolarization, and is only for the earlier layer (L1b), and the larger AE for the earlier layer would suggest smaller particles rather than larger. These seem like subtle and confusing trends. Is it clear that these distinctions are actually significant compared to measurement uncertainty (random + systematic)? Also, why is the conclusion that pollen is present, and not some other depolarizing type such as dust? 4.8% is small enough that even smoke could have such a value. The Sicard reference cited here does not help me understand this, since it’s not about this case, but is just a

general reference for lidar measurements of pollen. (And a minor related point: if the purpose is to credit lidar measurements of pollen, much earlier papers such as Sassen et al. 2008 should get that credit, doi: 10.1029/2008gl035085).

Page 10, line 26-29 Contradictory statements that the ratio of particulate depolarization at two wavelengths “did not vary significantly” but the difference between them “confirms the hypothesis of pollen contamination”. If the variation is not significant, it doesn’t confirm the hypothesis.

Page 10, line 31. Higher lidar ratio values “indicate domination by the biomass burning aerosol.” Again, based on what methodology and what references? All the papers I’m familiar with (e.g. Muller et al. 2007, Gross et al. 2012, Burton et al. 2012, Papagiannopoulos et al. 2018) have a significant overlap for the lidar ratio of biomass burning and pollution, at whichever wavelength they look at.

Page 11, line 1, imaginary refractive index values of 0.12i or 0.20-0.30i seem extremely high. This is quote from a paper in preparation. It doesn’t seem appropriate to present something so unexpected without any support or discussion.

Page 11, line 5, “as the LR values were relatively high of 81 plus or minus 6 sr and 60 plus or minus 6 sr at 355 nm and 532 nm, respectively, the layer can be treated as fresh biomass burning aerosol . . . although the angstrom exponent value of 1.34 plus or minus 0.3 is rather low for fresh BBA”. How is this conclusion reached? What values are these measurements being compared to? Couldn’t this be urban pollution?

Page 11, lines 15-16 “Such low depolarization ratios are characteristic for aged biomass burning”. No references are given to support this statement. Aren’t low depolarization ratios also characteristic of pollution and other aerosol types?

Page 11, line 21. “The AE value of about 0.6 [i.e. the same value as measured in this study] was reported by Muller et al. 2007”. Actually, Mueller et al. 2007 reported a value of 1.0 plus or minus 0.5. Maybe that could be considered “about 0.6” but it’s a misleading way to say it.

Page 12, line 4, appears to suggests that the backscatter angstrom exponent is expected to be monotonically related to the aerosol age. I don't know of any reason to think this. Are there references?

This section also contains statements two pages apart which directly contradict each other, giving me the impression that the authors do not fully understand their subject material and this manuscript was really not ready for submission in the first place. Regarding an apparent correlation between backscatter and depolarization, on page 11, "it is probable that increased depolarization is associated with the higher participation of background aerosol in the tropical air containing some mineral dust particles." And on page 13, "the negative dependence of backscatter and particulate depolarization is unlikely related to mineral dust contamination".

Another question about the measurements. On page 11, line 28, a lidar ratio of 114 plus or minus 33 sr at 355 nm. Isn't this really very high? Are there other published observations of 355 nm lidar ratios this high? I see from the figure that this very high value occurs where there is significant slope in the backscatter and extinction profiles, on the underside of a layer with larger backscatter. The peak of the backscatter and extinction have very different shapes, with the extinction looking like a smoothed version of backscatter. If there is a difference in the resolutions of the backscatter and extinction profiles such that the edges of the layers are not being captured the same way, it creates spurious values in the ratio. In this case, it looks like the lidar ratio is large in consequence of the fact that the backscatter and extinction show the edge with different slopes. Are the backscatter and extinction resolutions compatible when the lidar ratio is computed?

The wording is quite rough. If this manuscript were to be published or resubmitted, it would be very helpful to have a round of English-language editing.

Finally, where are the data available? Given that the paper is primarily about introduc-

ing a new dataset, I would like to see a statement of data availability.

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