

Review of ‘Seasonal cycle and source analyses of aerosol optical properties in a semi-urban environment at Puijo station in Eastern Finland’ by Leskinen et al., ACPD, 12, 4719-4754, 2012.

Science type comments

P4726 – lines 1-3 – the authors note that they reject scattering coefficients below the detection limits given by Anderson et al. (1996). First they should state what these detection limits are. Did they take the detection limits directly from the Anderson paper or did they calculate them for their nephelometer (every neph will have slightly different characteristics)? How much data was thrown out for being BDL? Also, I believe the nephelometer detection limits depend on averaging time – if you are throwing out BDL values for 1 minute averages and then averaging the data up to an hour you are not treating the data properly. Rejecting the high frequency low values will bias your lower frequency (i.e., hourly) averages to be higher than reality. A better approach would be to either set the BDL values equal to zero and include them in the analysis – or even better to leave the values as they are (even if they are negative) they should average out to a positive scattering if hourly averaged data is being used.

P4726 – lines 5-6 – the authors applied the Anderson and Ogren corrections to the nephelometer to account for truncation error and lamp non-idealities. They applied the values for no size cut, but that is inappropriate for the data after (Nov 2009) which was behind a 1 μ m impactor.

P4726 – the authors rejected angstrom exponents less than zero. I believe those are physically possible and should not be rejected. Regardless the authors should say what percentage of data is thrown out when such a choice is made.

P4727 – you should not mix averages and percentiles – they are different statistics. Use averages and standard deviations together and medians and percentiles together.

P4728 – lines 10 – do you have any indication that such inefficient mixing is occurring?

P4729 – lines 11-24 – I’m not sure why you are comparing Puijo with both clean remote Arctic measurements (e.g., Delene and Ogren, 2002) and highly polluted regions in China. Your title characterizes Puijo as a ‘semi-urban environment’. Given the location, the Arctic comparisons may be relevant depending on wind direction, but surely there is a better point of comparison than measurements in China? Please defend or change. Also – unlike Puijo data presented here, the Delene and Ogren Arctic measurements are screened for local contamination using wind speed and wind direction from the town of Barrow so that those measurements are representative of the region with minimal local influence. I believe the University of Manchester has some studies of diurnal variation of aerosol optical properties both near and farther from urban areas (look for Hugh Coe as one of the co-authors in ACP).

P4730 – lines 25-30 - based on time series plot of absorption and scattering (Figure 2) it looks like the Russian fires did not increase/change the monthly absorption at all. This is somewhat surprising to me – I think it is usually to see scattering and absorption both increase when a site is influenced by smoke. For long range transport the increase in scattering is typically more significant than absorption, which is primarily attributed to gas to particle conversion during transport, but both parameters do increase (discussed in Fischer et al., 2010 and Andrews et al., 2004). Do you see an increase in absorption in the hourly data that is masked in the monthly data? If not – where did the absorbing aerosol go?

P4731 – lines 13-19 – similar to a previous comment, I’m not sure why you are comparing Puijo SSA to SSA values reported in India, Beijing, airborne measurements from Ace-Asia and equatorial Pacific ocean measurements. Surely there are other, more relevant measurements to compare with? Are there no semi-urban SSA measurements for other places in Europe/Scandinavia? Also – for the measurements you are comparing with – are they all at the

same wavelength, please confirm that they are or note the wavelengths – SSA is wavelength dependent.

P4732 – Fig 4 shows plot of SSA vs scattering similar to that done in Delene and Ogren. Why not continue that similarity and also make the scattering angstrom vs scattering plot? It could also be interesting to plot scattering angstrom vs SSA. See for example discussion in Toledano et al (2007) and Andrews et al (2011)

P4734 – Line 15 – you note there were 260 other cloud events. Do you see any trends in any of the aerosol optical properties for those other cloud events or does the inlet used preclude this?

P4735 – lines 21-24 – most places will look relatively clean compared to Asia. Additionally the title calls Puijo a semi-urban site which comes with certain implications of pollution levels. Are you saying Puijo is a relatively clean semi-urban site or a relatively clean Arctic site? As mentioned in previous comments I think the comparison with measurements at other sites needs to be improved.

Editorial type comments

General comments:

I do not think the introduction to this paper (pages 4721-4722) properly cites the relevant literature. I am not saying that the papers cited aren't relevant, but typically the cited work is not the seminal work, but rather a recent paper and no acknowledgement is made of original work in the field. If the authors wish to have minimal citations for the sake of brevity that is their choice, but at the very least they should use the strategy of citing papers something like this: (e.g., Virkkula et al., 2011 and references therein) to provide an acknowledgement that the Virkkula paper is one of many that has built on the work of those that came before. I think a better strategy would be to provide a set of papers spanning the literature from older examples to more recent. For example, the Virkkula et al., (2011) paper is given as the single citation for ground-based, in-situ aerosol optical property measurements. Ground-based, in-situ aerosol optical property measurements of measurements have been made for decades – Bodhaine, 1983, and Bodhaine 1995 describe long-term, ground-based, in-situ aerosol scattering and absorption measurements from the 80s-90s.

In the section of the introduction describing long-term measurements at a variety of locations, the authors again seem (to me) to pick and choose citations, giving the incorrect impression that they are presenting a comprehensive list of aerosol climatology papers. I think this paragraph should be re-written to reflect the fact that they are only listing examples of long term measurements papers. They could reorganize this section by noting the literature contains both papers describing climatologies at multiple sites (e.g., Delene and Ogren, 2002; Andrews et al., 2011...) as well as climatologies for single sites (e.g., Virkkula, Zieger, Coen Collaud, etc...) and that the advantage of focusing on measurements at a single site is that it allows for more in-depth exploration of what causes temporal cycles.

P4720

Line 9 – delete 'the' in '...by *the* plumes...'

Lines 10-12 – rearrange: The absorption coefficient peaked in the winter when soot-containing particles derived from biomass burning were present. The higher relative absorption coefficients resulted in lower single scattering albedo in winter.'

Line 18 – change '*Changes* in the...' to '*Decreases* in the'

Line 20 – clarify whether the decrease in scattering and absorption due to clouds was primarily for paper mill aerosol or all aerosol.

Lines 23-27 – not appropriate for abstract – remove 'What happens to ... their activation into cloud droplets.'

P4721

Line 2 – change '...forcing has still a great uncertainty...' to 'forcing is still highly uncertain...'

Lines 4-5 – change ‘i.e. light scattering from and absorption to the particles.’ to ‘i.e., due to the combined effects of aerosol light scattering and absorption.’
Line 5 – change ‘Evaluation of its magnitude...’ to ‘Evaluation of the direct effect’s magnitude...’
Line 9 – change ‘...carbon, predominately...’ to ‘...carbon predominately...’
Line 9 – change ‘...black carbon, absorb it.’ To ‘black carbon absorb it.’
Lines 10-11 – change ‘...internal mixing...dramatically...’ to ‘internal mixing (e.g., of organics and black carbon) may dramatically influence...’
Line 13 – change ‘...Lidar...ranging...’ to ‘...Lidar (light detection and ranging)...’ (notes: I actually think it is not necessary to spell where the word lidar comes from at this point in time. Also, I don’t think you need to specify the type (Polly) of lidar as you do – it’s not consistent with your general description of in-situ instruments.
Line 19 – change ‘...so-called extensive properties...’ to ‘...intensive properties...’
Line 22 – delete ‘so-called’
Line 22 – delete ‘(Å)’ because you have individual symbols for scattering and absorption angstrom exponent
Line 26 – you should add some information about absorption angstrom exponent and backscatter fraction since you discuss them later in the paper.

P4722

Line 1 – Delene and Ogren (2002) includes a remote Arctic site.
Line 2 – Andrews et al (2011) summarizes the climatology of aerosol light scattering, absorption, SSA, backscattering fraction, scattering angstrom exponent and radiative forcing efficiency at 12 high altitude sites. It’s in the journal Atmospheric Research.
Line 4 – Fischer et al (2010) is NOT a coastal paper. It describes measurements made at a high elevation mountain site quite a distance inland (100s? km) from the ocean.
Line 6 – Zeiger et al (2010) is a nice paper, however it is not perhaps the best representation of a longterm Arctic monitoring paper – only covers ~4 months of measurements. Better papers would be Delene and Ogren (2002), and/or an overview paper on Arctic Haze by Patricia Quinn which appeared Tellus a few years ago or some especially ancient papers by Bodhaine (1983 and 1995)) describing long term measurements of aerosol light scattering and absorption at Barrow. All the papers I’ve mentioned describe multi year data sets which Zeiger does not. Tomasi et al (2007) also has a extensive overview of both in-situ and remote sensing measurements at both Arctic and Antarctic sites which I think includes both in-situ scattering and absorption.
Lines 16-17 – change ‘Furthermore, we examine...’ to ‘Finally, we examine...’
Line 24 – change ‘...city center of Kuopio’ to ‘...city center of Kuopio (population=???)’
Line 26 – change ‘The surroundings of the Puijo hill belongs to the...’ Puijo hill is in the ...’

P4723

Lines 2-3 – change ‘...moderate height hills and lots of long lakes in the northwest-southeast direction.’ to ‘...moderate hills and many lakes.’
Lines 4-10 – might be useful to include a map with the sectors and sources.
Line 28 – change ‘The samples to the instruments are drawn...’ to ‘Sample air for the instruments is drawn...’

P4724

Line 1 – delete ‘and sampling line’
Line 3 – delete ‘and sampling line’
Line 7 – the ‘in-cloud’ and ‘out-of-cloud’ terminology is confusing. Stick with the ‘interstitial’ and ‘total air’ description that is consistent with the inlet description in the previous sentences.
Line 8 – add a sentence something like: ‘Unless otherwise noted, all measurements described here were made through the interstitial inlet.’
Lines 14-15 – change ‘The backscattering is measured when a shutter blocks the light scattered to over 90° angles.’ to ‘The aerosol backscattering coefficient is measured when an internal shutter restricts measured scattering to between 90-170°.’
Line 15 – change ‘The nephelometer was drawing...’ to ‘The nephelometer drew...’

Line 16 – delete ‘cloud’

Line 17 – change ‘We checked the nephelometer...’ to ‘The nephelometer calibration was checked periodically (say approx how often – weekly?monthly?seasonally?) ...’

Line 18 – Add a sentence saying the frequency at which the neph data was collected prior to averaging if applicable (e.g., 1-min, 5-min, etc)

Line 21 – actual wavelength of MAAP has been reported to be closer to ~637 nm (Mueller et al 2011). If this is the case with the Puijo MAAP it will change your wavelength adjustment and make your SSA bigger (scattering will not decrease as much).

Line 24 – change ‘...that scatter light on the back-scattered radiation.’ to ‘...that scatter light creating a scattering aerosol artifact.’

Line 25 – add the references Bond et al (1999) – I think she was the first to come up with a scattering correction for filter based measurements – and also the Petzold reference describing this specifically for the MAAP.

P4725

Line 10 – Add a sentence saying the frequency at which the MAAP data was collected prior to averaging if applicable (e.g., 1-min, 5-min, etc)

Line 13 – change ‘...reveals...’ to ‘...measures...’

Line 17 – define the actual size range for the AMS in-case the reader is not familiar off the top of his/her head with the size range for Aitkin/accumulation mode aerosol.

P4726

Line 14 – delete comma after ‘estimate’

Line 24 – change ‘...MAAP unmatched...’ to ‘...MAAP do not match...’

Lines 26-27 – delete ‘, where the scattering angstrom exponent as700-450 was calculated from the scattering coefficients at 450 and 700’ (you’ve already defined the scattering angstrom exponent equations so you don’t need this.)

Lines 28-29 – delete ‘, where $\sigma_{ap,670}$ is the absorption coefficient at 670 nm’ (you’ve already defined this)

P4727

Lines 21-22 – change ‘...we analyzed the monthly...such as the traffic.’ to ‘...we inspected the (*average or median?*) diurnal cycles for each month in order to identify the effects of daily (e.g., traffic) and seasonally (e.g., fires?) time-dependent sources.’

P4728

Line 14 – change – ‘In order to reveal the air mass arrival patterns, ...’ to ‘In order to explore long range transport of air masses to the site,...’

Line 16 – change – ‘...by using FLEXTRA ...’ to ‘...using the FLEXTRA...’

P4729

Line 3 – delete ‘, as expected,’

Line 5 – change ‘...were found exceptionally...’ to ‘...were found to be exceptionally...’

Line 9 – say something about seasonal cycles of RH and visibility since you have them plotted (or delete those plots)

Line 11 – change ‘...average, based on 20175 hourly...’ to ‘...average scattering, based on 20175 hourly-averaged...’

Line 14 – change ‘...that observed in the Arctic region...’ to ‘...that observed at other Arctic sites...’

Line 18 – Delene and Ogren (2002) report absorption at 550 nm, not black carbon. If you are going to change units need to explain how did conversion. Sangeeta Sharma and Patricia Quinn present long term measurements of BC at other Arctic sites – if you wish to compare in terms of BC they would be better sources for comparison.

Line 22 – change ‘...reach occasionally comparable values...’ to ‘...occasionally reach values comparable...’

Line 25 – change ‘...The monthly average...being at its...’ to ‘The annual cycle of bsp,550 showed strong seasonality, being at its...’

P4730

Line 1 – change ‘...2010 made an...’ to ‘...2010 was an...’

Line 6 – change ‘...its visit to...’ to ‘...its deployment at...’

Line 8 – change ‘...when its measurement...’ to ‘...when this measurements...’

Line 8-9 – change ‘...results, for example, as an average...’ to ‘...results in an average...’

Line 11 – change ‘...cannot be stated...’ to ‘...cannot be determined...’

Line 14 – change ‘...corresponds to the value measured at various places (Delene...’ to ‘...is in the same range as *b* reported for a wide variety of location types (e.g., Delene...’

Line 16 – define spring months? In this section you mix references to seasons and months and occasionally it makes the text confusing.

Line 21 – biomass burning – is it wood/peat/other?

Line 23 – delete comma after ‘observed’

P4731

Line 3 – change ‘...nearly equal, between...’ to ‘...nearly equal, ranging between...’

Lines 3-4 – delete ‘monthly average of the’

Lines 4-5 – change ‘...2006 and ... summertimes...’ to ‘...2006 and highest in the summertime, reaching values of 2.1-2.2...’

Lines 6-12 – combine this paragraph with previous paragraph (lines 2-5)

Lines 3-12 – why not compare your measured scattering angstrom exponents with those made at other places? And with AERONET, as that would not require the high AOD₄₄₀ values required for calculation of AERONET SSA.

Lines 20-28 – Delete the discussion about the comparison with AERONET SSA. Also a comment on the phrasing in that paragraph– the range in SSA can be explained by uncertainty, but presumably the uncertainties in scattering and absorption don’t bias the SSA to low values.

P4732

Line 5 – change – ‘...mean values, calculated...’ to ‘...mean values of SSA, calculated...’

Line 6-9 – Delene and Ogren only presented statistically relevant points in their binned plots of scattering vs SSA – you should do the same. Look in that manuscript for discussion of mean standard error (MSE).

Line 11 – Here you are comparing the relationship between SSA and scattering at Puijo to a continental site (Bondville) in Delene and Ogren. Why not compare to the relationship at Barrow?

Line 18 – change ‘...the in situ SSA...’ to ‘...the ambient scattering coefficient and SSA...’

P4733

Line 1 – change ‘...turns fast...’ to ‘...quickly converts...’

Line 6 – put comma after ‘small-scale’

Line 8 – this is unclear because both sectors 4 and 5 are simply termed ‘RA’ but sector 5 has less absorption than sector 4 and the papermill sector has less than sector 4 also.

Line 14 – change ‘...southerly sector...’ to ‘...southerly sector with highway and large residential area...’

Line 15 – change ‘In our AMS analysis we observed a 2-3 fold particulate...’ to ‘The AMS data showed a 2-3 fold larger particulate...’

Lines 24-29 – what about other parameters? Does scattering angstrom exponent increase when SSA decreases?

Lines 28-29 – change ‘...seasons which resulted also as higher SSA values.’ to ‘...seasons, with a corresponding decrease in scattering, resulting in higher SSA values at night.’

P4734

Line 1 - Make a section header on effect of long range transport to match with section 2.5 about trajectory analysis.

Line 1 – Change ‘...northwesterly sector without local sources...’ to ‘...northwesterly sector with only small residential sources’ (if that’s correct!)
Line 4 – delete ‘Actually,’
Line 7 – change ‘...as a good background.’ to ‘...as background air.’
Lines 7-13 – this discussion is confusing and should be clarified.
Lines 1-13 – the conclusions discuss changes in scattering angstrom exponent due to long range transport so this section should too.
Line 25 – change ‘...resulting as a...’ to ‘...resulting in a...’

P4735

Lines 1-10 – the term ‘out-of-cloud’ is confusing. I think it would be better and more consistent with the literature to use the terms interstitial and total (which would include both interstitial and cloud drop residuals)
Line 3 – change ‘...for the absorption.’ to ‘...for absorption.’
Line 14 – change ‘...scattering from and absorption into aerosol...’ to ‘scattering and absorption by aerosol...’
Line 18 – change ‘...exponent and hemispheric backscattering fraction’ to ‘...exponent and the diurnal cycle of hemispheric backscattering fraction’ (since you don’t have enough backscattering data to do the annual variation)
Line 21 change ‘...areas) to the...’ to ‘...areas) on the..’
Line 24 – ‘a cloud event’. In the results section you discuss two cloud events.

P4736

Line 2 – change ‘...higher than at other...’ to ‘...higher for the NE sector than for other...’
Lines 6-8 – change ‘...combustion, the single scattering albedo decreases, when the absorption coefficient increases and the organic concentration increases’ to ‘...combustion, the absorption coefficient and organic concentration increases, while single scattering albedo decreases.’
Note: don’t include references to figures in conclusions section.,
Line 11 – delete ‘Since’
Line 12 – change ‘...areas, we conclude that...’ to ‘...areas, suggesting that...’
Line 17 – delete ‘(Fig 7a)’
Line 18 – change ‘...observed increase...’ to ‘...observed an increase...’
Line 18 – delete ‘also’
Line 20 – delete ‘(Fig 5d)’
Line 22 – delete ‘(Fig 6c)’
Line 23 – delete ‘(Fig 8)’
Line 25 – ‘...which are known to be larger in size and more oxygenated than the fresh emissions.’ This should be mentioned in the results section with the appropriate citations.

Figure 1 and 2 – should not combine means with percentiles. Change to means to medians **or** 10 and 90 percentiles to standard deviations (I think 2 standard deviations are cover about the same range as the 10 and 90 percentiles, but you should check that if that’s the way you decide to change the plot.

Figure 3 – what are the ranges given in the plots? Stdev or percentiles?

Figure 4 – remove statistically insignificant data – see related comment above.

Figure 5 and 6 – indicate the sectors with vertical lines? change axis to degrees for easier comparison with text (or add second axis with degrees).

Figure 7 – is Puijo the same time zone as UTC? Please note in text/figure caption. If it’s not then please add a vertical bar indicating local noon.

Figure 9 – Probably more interesting to show time series of SSA. Also, do you see a change in scattering angstrom exponent or backscattering fraction for cloud events?

References mentioned above

Andrews, E., Ogren, J.A., Bonasoni, P., Marinoni, A., Cuevas, E., Rodriguez, S., Sun, J.Y., Jaffe, D.A., Fischer, E.V., Baltensperger, U., Weingartner, E., Collaud Coen, M., Sharma, S., Macdonald, A.M., Leaitch, W.R., Lin, N.-H., Laj, P., Arsov, T., Kalapov, I., Jefferson, A., Sheridan, P.J., "Climatology of aerosol radiative properties in the free troposphere", *Atmospheric Research*, 102, 365-393, 2011.

==> in-situ aerosol optical properties at 10 ground-based, high altitude sites and 2 aircraft sets of measurements

Andrews, E., Ogren, J.A., Sheridan, P.J., Ferrare, R., "Vertical properties of aerosol optical properties at the ARM Southern Great Plains CART site," *J. Geophys. Res.*, 109 (D6): Art. No. D06208, 2004.

==> airborne in-situ measurements, but discusses in one section differences between local smoke and long range transport smoke

Bodhaine, B., "Aerosol Measurements at Four Background Sites," *J. Geophys. Res.*, 88, 10753-10768, 1983

Bodhaine, B., "Aerosol absorption measurements at Barrow, Mauna Loa and the South Pole," *J. Geophys. Res.*, 100, 8967-8975, 1995.

==> i think these two are first in-situ optical prop measurements in arctic

Collaud Coen, M., Weingartner, E., Furger, M., Nyeki, S., Prévôt, A. S. H., Steinbacher, M., and Baltensperger, U., "Planetary boundary influence at the Jungfraujoch analyzed by aerosol cycles and synoptic weather types," *Atmos. Chem. Phys. Discuss.*, 11, 1-40, 2011.

==> long term measurements at a site, seasonal cycles of various optical properties

Fischer, E. V., D. A. Jaffe, N. A. Marley, J. S. Gaffney, and A. Marchany-Rivera, Optical properties of aged Asian aerosols observed over the U.S. Pacific Northwest, *J. Geophys. Res.*, 115, D20209, doi:10.1029/2010JD013943, 2010.

==> plumes closer to source have lower single scattering albedo than long range transported plumes. see figure 6 and paragraphs 65-68

Mueller, T., et al. "Characterization and intercomparison of aerosol absorption photometers: result of two intercomparison workshops," *Atmos. Meas. Tech.*, 4, 245-268, 2011.

==> MAAP wavelength ~637 nm, see section on MAAP, p253.

Quinn, P.K., Shaw, G., Andrews, E., Dutton, E.G., Ruoho-Airola, T., Gong, S.L., "Arctic Haze: Current trends and knowledge gaps," *Tellus*, 59B, 99-114, 2007.

==> summary of arctic chemistry measurements for a bunch of surface sites. gives a bit about optical measurements too.

Toledano, C., Cachorro, V. E., Berjon, A., de Frutos, A. M., Sorribas, M., de la Morenab, B. A. and Goloub, P., "Aerosol optical depth and Ångström exponent climatology at El Arenosillo AERONET site (Huelva, Spain)," *Q. J. R. Meteorol. Soc.*, 133, 795-807, 2007.

==> groups aerosol in type as a function of relationship between loading (AOD) and angstrom exponent

Tomasi, C., et al., "Aerosols in polar regions: A historical overview based on optical depth and in situ observations," *J. Geophys. Res.*, 112, doi: 10.1029/2007JD008432, 2007.

==> mostly aerosol optical depth stuff but some insitu optical props too.