

Review of Pandolfi et al 2017 – ACTRIS nephelometer measurements

This manuscript summarizes nephelometer measurements (in terms of monthly climatology, co-variance of scattering-related optical properties and trends) across the ACTRIS network in Europe. The manuscript also includes a few non-European sites supported by ACTRIS. A lot of work went into bringing these data sets together and summarizing station differences (instrumentation, size cut, corrections...).

General/Technical comments

The manuscript includes a lot of information and clearly a lot of work was involved in getting that information into useable form for analysis, but, as written, it's really hard to digest. Some of that would be helped by more editing (e.g., shorter paragraphs as described in my editorial comments below).

Why not compare scattering seasonality with Zanatta seasonality of absorption where possible?

Why not present SSA seasonality/trends at the same time? I realize an absorption measurement is needed to do so – but Zanatta's ACTRIS paper means those data exist for some sites and should be in some sort of consistent form.

I understand you want to include all ACTRIS supported light scattering measurements, but the non-European sites (TRL and CHC) are a distraction. They seem to only be included because they are part of ACTRIS but not for any strong scientific reason and are barely discussed in the text. I think it is fine to include these stations in the supplemental materials if you must, but they should be eliminated from the main manuscript.

Page 6, Line 23 – the nomenclature 'regional' and 'continental' is a bit confusing. Perhaps change continental to remote or rural? Or combine to 'continental' and note that some are more polluted than others? This would make sense as I think they are usually discussed together in the manuscript.

Page 7 – somewhere in Section 2 (under data treatment) you should explain where the data came from, i.e., was it downloaded directly from EBAS (if so, what level (presumably level 2)). Or was it provided by data providers? Or some combination of the two? You should also say whether you performed additional quality checks on the data or did you take them "as is" from EBAS/data providers? I would hesitate to assume that even Level 2 data downloaded from EBAS it is OK to use without further review (particularly if you are working with backscatter values of any wavelength and blue or red scattering values).

Personal experience with the Level 2 nephelometer data in EBAS suggests that much of the data provider QC focuses primarily on the green scattering and the other nephelometer parameters may not get as much attention. A further issue is that it seems that often data QC is done by different individuals in different years

and they don't look at years before and after their year to see if there are obvious differences.

Page 7, Line 15-18 – Later on you note that there are differences in operations from the GAW protocols – the measurement RH is one obvious difference from GAW protocols at many sites, so the statement ‘the nephelometer instruments are run following the ACTRIS/GAW standards (WMO-GAW Report, 2016)’ is not strictly true. Please revise.

Page 7, Line 29 – ‘Due to the non-homogeneity of the light source’ this should be made clearer. Was the light source causing measurement problems and is there a reference report describing this? Do you use the data from before the light source change? Why were different replacement light sources used at SIR and CMN and does this have implications for the data?

Page 8, Line 15-16 – “However, for $\text{SSA} < 0.8$ a correction scheme based on particle number size distribution should be used” Was this type of correction necessary for any of the stations? It looks from the table like it may have been performed for some stations, but was it necessary for those stations?

Page 8, Line 19-20 - Only at SIR, FKL, and CMN, σ_{sp} data are not corrected for truncation because σ_{sp} at these observatories was measured at one wavelength. Anderson and Ogren suggest corrections for the nephelometer when the SAE is not available. Doesn't the Mueller scheme also provide an option for correction when no SAE is present? I would expect FKL (a coastal site presumably with large sea salt aerosol) will be quite sensitive to not being corrected for truncation and uncorrected scattering will significantly underestimate the actual scattering.

Page 9, Line 10-11 –“However, the scattering enhancement due to a change in RH between 40% and 50% should be small.” This very much depends on the aerosol type. See hygroscopicity work at several of these ACTRIS sites by Paul Zieger as well as more recent (2017) work on sea salt. How much do the results change if $\text{RH} < 40\%$ is chosen instead of $\text{RH} < 50\%$? That would (a) be a useful finding and (b) indicate whether the statement above is reasonable (though it would not prove it one way or the other).

Page 9, Line 31 – is 1.5 a reasonable Angstrom exponent for a marine site? Seems a bit high to me

Page 10, Line 28 – I think it would be worthwhile to explain why the blue and red scattering have less coverage. Are the consistent problems with the wavelengths in particular nephelometers or are there data QC issues? (I realize some sites only have green measurements but it's unclear from your statement if those are the sites that don't have red/blue coverage (it wouldn't be expected!) or if it is other sites that do have those measurements but for whatever reason they aren't available. My experience with Ecotech neph data suggests the red wavelength is pretty unstable at many sites within the ACTRIS network and that

has implications for SAE climatologies and trends.

Page 12, Lines 4-10 – it would be good to include the physical interpretation of the skewness as well as the mathematical

Page 11, Line 34 - PAL is often considered an Arctic site (see e.g., Backman et al., AMT, 2017) so it may make more sense to group it with ZEP)

Page 14, Lines 19-21 – you have the data and can determine whether the inlet size cut had an effect on the Angstrom exponent. Could plot monthly medians of SAE before and after the inlet was changed.

Page 15, Line 22 – why do BIR and PLA have a large particle peak?

Page 20, Lines 20-26 – these percentage comparisons are misleading – scattering Angstrom exponent is constrained between the values of approximately 0 and 3 and therefore you will not see anywhere close to the same percentage change as you would see for scattering which is unconstrained. I would remove lines 20-26

Page 25, Line 32 – PAL trends in SAE – you may want to discuss with John Backman – the Level 2 PAL data in EBAS has some suspicious red values for several years.

Page 27, Line 29 – are there day/night differences at coastal sites due to onshore/offshore flow?

Editorial comments

It would be good to have a native English speaker read and edit the paper before resubmission. It would also be really helpful to the reader to organize the discussion better – the paragraphs are really long and it's occasionally hard to follow the arguments because of that. I've suggested some places where the really long paragraphs could be broken into smaller paragraphs but some transitional sentences may be needed.

Page 4

Break into smaller paragraphs: line 11 – start new paragraph, line 19 – start new paragraph, line 32 – start new paragraph

line 15 – there are evidences → there is evidence

line 17 – phrasing of 'would eventually unmask the global warming'

line 22-23 – phrasing of 'Several international projects are providing in the last decades important information on the atmospheric particle properties worldwide'

line 25 completed → complemented

line 26 - 27 USA or EMEP → USA and EMEP

line 30 – define RTD

Line 36 – You should be careful here. (1) EBAS also includes data from the IMPROVE network nephelometers which are operated outside at ground level and at ambient conditions with no size cut. These IMPROVE data aren't really comparable to the ACTRIS data sets discussed here. (2) Additionally there are other sites making nephelometer measurements that aren't providing the data to EBAS (see comment below for page 5, lines 20-22).

Page 5

Line 7 'that decreasing or' → that a decreasing or

Lines 13-22 – should cite Sherman et al 2015 – it is an updated version of Delene and Ogren 2002

Lines 20-22 – this statement only relates to multi-station studies although Andrews et al included 3 mountain sites in Asia (WLG (China), PYR (Nepal), LLN(Taiwan)). Please rephrase to make clear that there are measurements outside of Europe and the US, but that many of those measurements have primarily been written about in isolation (e.g., not in the context of other sites). For example, FMI has reported on long-term optical properties in Saudi Arabia and South Africa. Paolo Laj et al have reported on aerosol optical properties in Nepal, and Paolo Artaxo in Brazil. Note: this manuscript doesn't really change this since it is focused on European sites as well and I think you should remove the non-European sites you do include to improve the discussion and flow.

Line 26 - 'related with aerosol phenomenology' → related to aerosol phenomenology

Line 31 – 'Zanatta' → 'and Zanatta'

Page 6

Lines 1-13 should be in methods section 2.2.3, not introduction

Line 1 – delete 'In fact,'

Line 1 – add the word 'spectral' in front of σ_{SP}

Line 5 – particles → particle

Line 7 – associated to → associated with

Line 7 - course → coarse

Line 11 – a better reference is the Andrews et al. 2006 reference which you also cite later. Ogren 2006 is gray (not peer-reviewed) literature and the Andrews paper is the peer-reviewed version of it

Line 18 – performed → characterized

Line 19 – observatories → observatory

Line 19 – measurements → measurement

Line 20 – divided in -→ divided into

Line 22 – coastal – how close to the sea coast?

Page 6-lines 26-36 and Page 7 lines 1-4 - Delete and say the categories for each station are given in table 1. No need to have in paragraph form also.

Page 7

Line 5 - Earlier you say mountain sites are higher than 1km, but then you characterize HPB at 985 m as a mountain. Should change previous statement and say mountain sites are at or higher than 985 m to be consistent.

Line 9 – investigation → investigations

Lines 11-15 – delete - you say this in the trends section.

Line 25 - 'Most used nephelometer models are the' → The most common nephelometers in the ACTRIS program are the

Line 27 - 'Other used models' → Other models used

Line 37 – 'guarantee the quality and comparability of the data.' This is a strong statement. There are RH control issues, wavelength, differences, data QC issues and many of the systems operate with different inlets/size cuts (e.g., whole air, pm2.5 pm 10). Do stations have different procedures for dealing with negative values close to zero? How do they deal with local contamination? There are unfortunately a lot of issues and while the nephcs themselves are probably quite comparable when they are operated side-by-side as happens at a Leipzig workshop the operating conditions at the stations will vary and make the measurements less comparable. What happens if a nephelometer fails a performance check – is the data prior to that time invalidated? You discuss some of these issues below in the sections below but I recommend changing the text as follows:

{Recommended quality assurance procedures during on-site operation as described in GAW (WMO/GAW, 2016), guarantee the quality and comparability of the data. Moreover, most of the integrating nephelometers involved in ACTRIS have undergone performance checks at scheduled times at the World Calibration Center for Aerosol Physical properties of ACTRIS/GAW.}

Change text in red to text in blue→

{Recommended quality assurance procedures during on-site operation as described in GAW (WMO/GAW, 2016), help to ensure the quality and comparability of the data. Additionally, most of the integrating nephelometers involved in ACTRIS have undergone performance checks at scheduled times at the World Calibration Center for Aerosol Physical properties of ACTRIS/GAW.

2.2.2 Data treatment

The σ_{sp} and σ_{bsp} data reported to EBAS and used in this work are referenced to standard T (273.15 °C) and P (1013 hPa) conditions. There are however station-to-station differences (e.g., sizecut, RH control, wavelength, data processing, etc) which are addressed in the sections below.

2.2.2.1}

Page 8

Line 30 – should say here in the first sentence that the GAW (and ACTRIS?) protocol is RH<40%. You say it later in the paragraph but it should be at the very beginning of the paragraph.

Page 9

Lines 14-16 - this should be moved elsewhere – it is not related to the RH discussion. See my suggestion above which puts it after section heading 2.2.2

Line 20 – other used wavelengths → other wavelengths used

Line 23 – most used → most common

Line 24 – following Sections → following sections,

Line 27 – (and at CMN → (or at CMN

Line 28-29 - measured at different wavelength than 550 nm → measured at additional wavelengths to 550 nm

Page 10

Line 6 - delete (with $\lambda_1 > \lambda_2$): this is not a necessary condition for the SAE equation you provide - you can flip things around so long as you are consistent,

e.g., if $\text{ssp550}=40$ and $\text{ssp700}=30$, then $\text{SAE} = -\log(30/40)/\log(700/550) = -\log(40/30)/\log(550/700) = 1.19$

Line 8-9 – ‘Here, the SAE is calculated as linear estimation of σ_{SP} measured at the three available wavelengths.’ This statement is inconsistent with the equation describing Angstrom exponent (eq 1). Further the relationship is not linear – that’s why the equation has logs in it. Please clarify what was meant.

Line 14 – given radiation → given direction

Line 18 – see previous comment about the Ogren 2006 citation

Page 11

Section 3.1 – two points: (a) Why no separate section for Arctic sites? Could move discussion of ZEP and PAL to an arctic section (as suggested above I would remove discussion of TRL from main manuscript). (b) Why not order the discussion of sites from typically cleanest (arctic) → mountain → coastal → ... → to typically dirtiest (urban). That would make the discussion easier to follow I think.

Line 20 - placements → locations

Line 32 – because their → because of their

Line 35 – since these are → because they are

Line 27 – low scattering are → low scattering is (or low scattering value are)

Page 11, line 16 – page 12, line 10 – Split the paragraph into several smaller paragraphs: Start a new paragraph at page 11 line 25 for discussion of figure 3. Start a new paragraph at line 29. Start a new paragraph page 12 line 4.

Lines 29 to page 12, line 4 – This discussion is hard to follow – in part because the site types as defined earlier aren’t used consistently.

Page 12

Line 13 – placements → environments

Line 30 – here you combine regional and continental stations for discussion – I do think it makes sense to call them clean and polluted continental stations – it would make the discussion simpler.

Line 31 – present → exhibit

Line 33-34 – I think ‘linked to strong stable air with thermal inversion’ this could be better phrased: linked to stable air due to strong thermal inversions

Line 33-34 – delete ‘On the other side,’

Page 13

Line 2 – instead of continental should say Nordic/Baltic continental sites because those are the subset of sites VHL is being compared to rather than all continental sites.

Page 14

Section 3.2 – maybe would be easier to read if had subsections by station type or could have two subsections: (a) by geography (east to west) (b) by type (coastal, mountain, etc). Right now they are a bit intertwined.

Line 16 – start new paragraph

Line 37 – the SAE data → the frequency plot of the SAE data

Page 15

Line 13 – start new paragraph

Line 18 – cite work by Zieger et al 2010 at ZEP (their figure 4) which shows presence of sea salt at ZEP.

Line 24 – ‘Differently than σ_{sp} , ...’ → Unlike σ_{sp} , ...

Line 34 – start a new paragraph with ‘Also at...’

Page 16

Section 3.3 – maybe would be easier to read if had subsections by station type or could have two subsections: (a) by geography (east to west) (b) by type (coastal, mountain, etc). Right now they are a bit intertwined.

Line 14 - start new paragraph

Line 14 – “..thus the lower BF the higher is g ..” → change to “...with lower BF corresponding to higher g ...”

Line 16-17 – “Higher g median values are in some cases observed at mountain sites compared to regional or urban environments” → At some mountain sites higher median g values are observed relative to the g values obtained at regional or urban locations.

Line 18 – “...European sector or HPB...” → ...European sector and for HPB...

Line 19 – “However, exceptions are observed for example for CMN...”
→However, exceptions are observed. For example, at CMN, ...

Line 23 – start a new paragraph

Line 35 – start a new paragraph

Page 17

Line 10 – start a new paragraph

Line 10 – “Moreover, the refractive...” →The refractive ...

Line 13 – “...did non linearly...” →...non-linearly decreased...

Line 16 – higher → larger

Line 17 – “On the other side, Obiso et al. (2017) showed...” → Obiso et al. (2017) also showed...

Line 20 – “This kind of...” →These kind of ...

Line 36 – retitle section BF and g vs scattering relationships. Both are talked about in the section so the title is a little confusing

Page 18

Line 7 – Sherman et al., ACP 2015 also.
www.atmos-chem-phys.net/15/12487/2015/

Line 37 “or higher 1.5” →or higher than 1.5

Page 18, Line 20 – page 19, line 17 – this paragraph should be better organized and split into two smaller paragraphs. It's a bit hard to follow in its current form.

Section 3.4.1 – general comment – I would recommend first discussing the SAE/scattering relationships and then relating it to the g/BF relationships. Move lines 20-23 to a paragraph at the end of the section. Also, perhaps it would make sense to split this section into aerosol types ‘marine/dust’ and ‘anthropogenic’.

Page 19

Section 3.5 – my personal suggestion would be to switch the order of sections 3.4 and 3.5. Sections 3.1-3.3 talk about overall variability of the different parameters on an annual basis. It seems logical to talk next about the seasonal variability and tie it into annual variability. Once the individual parameter variability has been discussed then it makes sense to look at how those parameters co-vary (i.e., 3.5). Note: that's also the order those topics are presented in the conclusions.

Line 25 - ad → and

Page 20

Line 6 – start new paragraph

Lines 11-13 – “At the southern station of MSA the observed less pronounced seasonal cycle of SAE could be related with the Saharan dust outbreaks which contrast the PBL transport of fine particles observed at other mountain sites” → it's unclear what is being said here; is MSA not impacted by dust?

Line 14 - “July-August being the Saharan dust outbreaks very” → July-August. The Saharan dust are very...

Page 21

Line 7 – “being the SAE” → with the SAE

Page 22

Split the southern Europe paragraph into several paragraphs. Line 15 – start new paragraph. Line 27 start new paragraph.

Page 23

Line 3 – citation for recommendation about having more than 10 years of data for trend analysis?

Line 20 – start new paragraph

Page 24-25

This section (3.6.1) needs to be broken into smaller paragraphs! Line 9 – start new paragraph. Line 17 start new paragraph.

Line 14-15 – “A statistically significant decreasing trend of σ_{SP} at IPR was also reported by Putaud et al. (2014) for the period 2002 – 2010.” → delete from text and put as footnote to Table 2.

Line 16-17 – “As reported in Table 2 statistically significant decreasing trend for σ_{SP} is observed at around 50% of the stations considered here.” → delete from text – put the 50% number in the first sentence of the first paragraph on line 2 of this page.

Page 25

Line 8 – start new paragraph

Line 36 – start new paragraph

Page 26

Line 9 start new paragraph about BF trends

Line 21 start new paragraph

Tables and Figures

Table 1 – Explain that the ‘observatory code’ is ACTRIS’ code (or EBAS?) (or GAW?). Note where necessary any differences between GAW ids and station ids (e.g., Finokalia’s GAW id is FIK and there may be others).

Figure 2 – you could put this on log scale

Figures 2, 4 and 5 – how would this look if you had panes for different site types and then organized by geographical region? For example, you would have a pane for mountain sites and then sections for Nordic (empty), western (puy), central (jfj, cmn, hpb), etc. I think doing that would make it easier to see the east west shift and also commonalities among site types. You could keep the boxes colored by geographic region within each pane. You could do a similar thing with figure 6.

Supplemental materials

Table S1 – how is this table organized? It’s not alphabetical or by geography. Or by instrument type.

Table S2 - should provide number of points with RH>40% for each station (i.e., how many points are above the GAW and ACTRIS protocol value) as well as the number of points with RH>50%. Caption should state that table is organized by decreasing number of points with RH>50%.

Figure S1 – should make the x-axes cover the same range (0-80%?) and draw a vertical line at 40% (GAW protocol value) and 50% (value chosen for this paper). Lines could be added indicating the median RH distribution as a function of season. Explain why different widths of bars on distribution plots (presumably because different numbers of data points). I think it would make more sense to use consistent widths for the distribution plots.

Table S3 – state in caption what colors lambda1, lambda2, and lambda3 correspond with. (not the wavelength because that obviously changes with instrument and time period)

Figure S2 – remove map – that’s already a figure in the manuscript. Explain why different widths of bars on distribution plots (presumably because different numbers of data points). I think it would make more sense to use consistent widths for the distribution plots.

Figure S3 – remove map – that’s already a figure in the manuscript. Explain why different widths of bars on distribution plots (presumably because different

numbers of data points). I think it would make more sense to use consistent widths for the distribution plots.

Figure S4 – remove map – that's already a figure in the manuscript. Explain why different widths of bars on distribution plots (presumably because different numbers of data points). I think it would make more sense to use consistent widths for the distribution plots.

Figure S6b – explain the different colors of dots in the figure caption. Perhaps you could make the bars in figure S6a the same color as the dots in figure S6b and then use some other color for the dots in figure S6a

Figure S8 – has MTC instead of CMN