Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-1058-RC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "How much of the global aerosol optical depth is found in the boundary layer and free troposphere?" *by* Quentin Bourgeois et al.

Anonymous Referee #2

Received and published: 18 December 2017

This is a concise, well-written paper that addresses a potentially important topic for radiative forcing and atmospheric processes and transport – i.e., the vertical distribution of optically important aerosol. The authors utilize aerosol extinction profiles from CALIPSO and split the aerosol optical depth into boundary layer air and free troposphere air based on boundary layer heights determined from ECMWF ERA interim analysis. It does however need some major revisions in terms of details about uncertainties and the in-situ comparison. I've first provided major science comments and then some minor technical notes and editorial comments.

Major science comments: (1) There is very little space given to uncertainties in the

CALIPSO extinction and AOD retrievals. The authors at some point note that CALPISO underestimates extinction at values below 0.001 km-1. This should be in the methods section. Additionally, this is actually a pretty high extinction value, corresponding to 100 Mm-1. Most background surface observatories in North America and Europe measure aerosol scattering values less than 0.0005 km-1 (see for example, Pandolfi et al ACPD 2017 and Sherman et al ACP 2015). Scattering tends to be around 90% of extinction (assuming a single scattering albedo of 0.90) so this suggests that CALIPSO retrievals of extinction will also underestimate BL AOD in many locations. The authors give no indication of the magnitude of the underestimation, whether it scales with aerosol loading below 0.001 km-1 or even what the uncertainty is. This is critical information when comparing relative loading of BL and FT.

(2) I appreciate the authors' desire to put the satellite retrievals in the context of in-situ measurements (the LOAC) but feel that this either requires more work or should be removed from the manuscript. The limited nature of the comparison doesn't particularly strengthen the paper and indeed raises more questions than it answers. Some things that should be included if it stays: a) how representative of the 2x2 grid is the region from which the LOAC balloon is launched (it looks like the launch site is close to the Pyranees and the coast and Toulouse which could wreak havoc with the BL height determination and be subject to significant subgrid variability in the aerosol in the BL (and FT) b) provide the size range and assumed refractive indices for converting from size distribution to extinction. c) does the LOAC measure dry aerosol or ambient? If dry what assumptions are made about hygroscopicity to convert to ambient extinction? d) what is the balloon flight path – does it stay in the 2x2 grid around the launch site? If not how does that affect results? e) why not include a plot of the comparisons of the CALIPSO and LOAC profiles? You could show median profiles for the two instruments and use shading to indicate the variability.

(3) I am surprised that the authors did not cite a similar (but better characterized and constrained and with more cases) comparison between CALIPSO and in-situ aerosol

vertical profiles by Sheridan et al in ACP (2012). This paper demonstrates (albeit with an older version of CALIPSO data) the lack of sensitivity of the CALIPSO extinction profiles to extinction values below ∼25 Mm-1 (0.00025 km-1) which is likely larger than the extinction in much of the free troposphere.

(4) Comparisons with ground based and airborne lidar have also explored the FT vs BL loading (e.g., Giannakaki et al (2015) and Rogers et al (2014). The Rogers paper also discusses CALIPSO detection limits (for an earlier version of the data).

Minor comments and editorial notes Page 1 line 5 – need to make clear the limited nature of this comparison in abstract and note location of LOAC flights

Page 1 line 12 – replace process with processes

Page 2 line 24 – replace govern with governs

Page 3 line 20 – some discussion of subgrid variability – a 2x2 degree grid can be pretty variable in terms of aerosol loading – see for example Weigum et al (2016)

Page 3 linen 21 – the Arctic is going to be almost as clean as the Antarctic for the vast majority of its area, particularly in terms of CALIPSOs sensitivity to aerosol extinction. I would suggest some caveats here.

Page 4 line 16-17 – awkward sentence, I'd suggest something like 'AODs are computed for each of the seven aerosol types consider by CALIOP, for both the BL and FT.'

Page 5 lines 9-17 – please see my major comment above. This paragraph needs to have more detail included to make it useful for the reader.

Page 5 lines 16-17 – '. . . particles in the BL and FT are reported as mostly absorbing and scattering, respectively, . . .' I'm not sure what this sentence is supposed to say. In-situ measurements suggest that the aerosol in the BL are primarily scattering – typical single scattering albedo values are ∼0.9 (or higher!) meaning they are ∼90% scattering and 10% absorbing. Sea salt aerosol, which the authors note typically stays

in the BL is pretty much 100% scattering. Please clarify what is meant here.

Page 6 line 3 – put numbers on the FT contribution in polar regions as is done for the other regions further down.

Page 6 line 4 '. . . detection limits of the instrument.' Please state what these are here and/or in the methods section where the CALIPSO retrievals are described.

Page 6 line 16 – to what does Toth et al 2016 ascribe the AOD decreases observed in Africa and China?

Page 6 line 19 – replace 'indicates' with 'suggests'

Page 6 line 24 – include clean continental aerosol in table 3 and revise this sentence.

Page 6 line 30 – replace 'is' with 'are'

Page 6 line 32,33 – replace 'contribute to about' with 'contribute about'

Page 7 line 5 – replace 'contribute to' with 'contribute'

Page 7 line 7 – the emissions sources will be on the surface (except for airplanes). Do you mean vertical transport or something like that?

Page 7 line 12-13 – rewrite sentence as 'It should be noted that while the AOD can provide a rough measure of total particulate mass, particle residence time and cloud interactions depend strongly on the particle size distribution.'

Page 7 line 20 – replace 'contribute to about' with 'contribute about'

Page 7 line 27 – delete '(not shown)'

Page 8 line 8 – '. . .scattering or absorbing particles only,. . .' why would only absorbing particles be considered? The only place those will exist is at the tailpipe of a diesel engine and even there they will have some amount of scattering (SSA∼0.3-0.4).

Page 8 line 11 – delete the sentence about the fraction of number concentration. Since

the LOAC only goes down to ∼0.2 micrometers it's missing a lot (most) of the number concentration.

Page 8 line 19 – See major science note#1. If 0.001 km-1 is where CALIPSO starts underestimating extinction then AOD in much of the BL (except in highly polluted regions) is also going to be underestimated.

Page 8 line 21 – A plot comparing LOAC and CALIPSO for the one coincident profile would be good and a plot showing the statistical comparison with all the LOAC profiles in the 2x2 grid would also be good to give the reader confidence in your results.

Page 9 line 9 – how does the vertical distribution of particles affect their size distribution? Isn't it the other way around?

Page 15 Table 3 – one not include 'clean continental' in the table for completeness?

Page 15 Table 3 – presumably these are based on averages not medians?

Page 17 Figure 2 – it would be interesting to see these maps plotted as the ratio (or difference?) of FT to BL AOD. Doing so would better highlight regional differences. To some extent this is shown in figure 3 but Figure 3 masks the longitudinal differences. For example, is the peak at the equator in figure 3 primarily due to the dust/biomass burning in the 60W to 60E region or does the aerosol get transported outside that longitudinal band and there's actually a FT/BL discrepancy for the full 360? Similarly it would make the Arctic FT contribution more obvious.

Page 19 Figure 4 caption – replace 'full' with 'solid'; replace 'show' with 'indicate'

Cited references:

Giannakaki et al 2015: https://www.atmos-chem-phys.net/15/5429/2015/acp-15-5429-2015.pdf

Rogers et al 2014: https://www.atmos-meas-tech.net/7/4317/2014/amt-7-4317-2014.pdf

Pandolfi et al 2017: https://www.atmos-chem-phys-discuss.net/acp-2017-826/

Sheridan et al 2012: https://www.atmos-chem-phys.net/12/11695/2012/

Sherman et al 2015: https://www.atmos-chem-phys.net/15/12487/2015/acp-15-12487-2015.pdf

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