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
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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.
We would like to thank the referee for these positive words as well as the careful review. We very much appreciated the suggestions and comments that helped us significantly improve the manuscript.

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.
The CALIOP detection sensitivity is added to the method section. However, we are surprised by the second part of the comment. An extinction of 0.001 km-1 does not correspond to 100 Mm-1 but to 1 Mm-1 (0.001 km-1 => 0.000 001 m-1 => 1 Mm-1). As a consequence, the whole comment is irrelevant because the reviewer supposes that 0.001 km-1 is a large extinction value while it is actually very low.

(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.

a) **Particles found in Aire sur l’Adour are representative of a rural aerosol background because the closest mountain (Pyrenees), big city (Toulouse) and ocean (Atlantic Ocean) are located at about 100 km away. As a consequence, LOAC remains in this background aerosol**

environment within the first kilometers of the balloon ascension and the choice of a 2x2 degree grid (about 200x200 km) is relevant.

b) **The size range of LOAC is 0.2-20 microns. Typical refractive indices are used: 1.45 for liquid and transparent particles, and 2+0.6i for absorbing particles. This is now mentioned in the method section.**

c) **LOAC measures ambient aerosols so no assumptions have been made about hygroscopicity.**

d) **As mentioned in the answer a), measurements are always made within the 2x2 degree grid, at least within the troposphere. It is however possible that LOAC flies outside of this grid box in the stratosphere.**

e) **A plot showing the mean vertical profile of aerosol extinction values over Aire sur l'Adour for the 23 LOAC flights and for CALIPSO orbit tracks passing in the 2x2 degree grid box centered on Aire sur l'Adour a few days before or after each LOAC flight (because there is only one single LOAC flight collocated in space and time with a CALIPSO overpass) was included (Figure 6).**

(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.
Same unit conversion mistake, 25 Mm-1 corresponds to 0.025 km-1 (not 0.00025 km-1). However, following the recommendation of the referee, we repeated the calculations of Sheridan et al. [2012] with CALIOP v4.10 data and 28 collocated flights over Illinois (USA). We arrived at the same conclusion with regards to extinction at the limits of CALIOP detection (i.e. an underestimate of the lowest aerosol extinction values). As a consequence, the global statistics of the study are very likely biased low, in particular in the free troposphere. We expanded the discussion about this matter but we did not add any new figure because it would probably be redundant with Sheridan et al. [2012] results.

(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).
We now include the Rogers et al. [2014] paper in the discussion. However, we cannot really compare Giannakaki et al. [2015] findings with our results because Giannakaki paper uses the PollyXT algorithm for the retrieval of the PBL while we use ECMWF. Note also that the Giannakaki study uses nighttime data only. According to Korhonen et al. [2014], the PBL is found during nighttime at about 1100m in PollyXT vs 200m in ECMWF (see their Figure 10). As a consequence, Giannakaki et al. find a nighttime FT contribution of 46% to the AOD while we find a nighttime FT contribution of 93% (because only the first 200 m are found in the BL in our study).
Korhonen et al. [2014]: https://www.atmos-chem-phys.net/14/4263/2014/acp-14-4263-2014.pdf

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
The location of LOAC flights (Aire sur l'Adour, France) has been added.
Page 1 line 12 – replace process with processes
Done.
Page 2 line 24 – replace govern with governs
Done.
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)

The effect of subgrid variability would be relevant if a model with a "coarse" resolution was used in the study but we average individual vertical profiles having a horizontal resolution of 5 km on a 2x2 degree grid.

Page 3 line 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.

We just mention that we do not use CALIOP data over the Antarctic region because they are likely spurious as also reported in Winker et al. [2013]. Indeed, high aerosol values are reported with CALIOP despite it is known to be a pristine region.

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.'

Done.

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.

See answer to the major comment above.

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.

This sentence has been removed since it was confusing and it has been added earlier that Mie scattering theory is used for two different particles (liquid and transparent particles, refractive index = 1.45 & solid and absorbing particles, refractive index = 2+0.6i).

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

Done.

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.

The CALIOP aerosol extinction detection sensitivity is now mentioned in the method section. A range is given because it also depends on the nature of the particles (i.e. the lidar ratio).

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

Toth et al. [2016] attribute the AOD decrease in China to a decrease in aerosol loading after the 2008 Olympic games and Ridley et al. [2014] attribute the AOD decrease in Northern Africa to a decrease in dust emissions due to a reduction in surface winds over the dust source regions. This is now mentioned.

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

Done.

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

Since the contribution of clean continental aerosols to the AOD is almost null, we do not think that it is relevant to include them into table 3.

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

Done.

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

Done.

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

Done.

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

We changed the sentence by "This can partly be explained by the location of the emission sources (near the convective regions)".

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.'
Done.
Page 7 line 20 – replace 'contribute to about' with 'contribute about'
Done.
Page 7 line 27 – delete '(not shown)'
Done.
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).
This sentence has been removed since it was confusing.
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.
The sentence about the aerosol number concentration has been removed.
Done.
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.
See answer to major comments. An extinction of 0.001 km-1 is very low and the extinction in the BL is usually substantially larger than that.
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.
A plot showing the mean vertical profile of aerosol extinction values over Aire sur l'Adour for the 23 LOAC flights and for CALIPSO orbit tracks passing in the 2x2 degree grid box centered on Aire sur l'Adour a few days before or after each LOAC flight (because there is only one single LOAC flight collocated in space and time with a CALIPSO overpass) was added (Figure 6).
Page 9 line 9 – how does the vertical distribution of particles affect their size distribution? Isn't it the other way around?
The sentence has been changed for "the distribution of aerosol particles between BL and FT affects their atmospheric residence time".
Page 15 Table 3 – one not include 'clean continental' in the table for completeness?
As mentioned above, we do not think that it is relevant to include them into table 3 because their contribution to the AOD is almost null.
Page 15 Table 3 – presumably these are based on averages not medians?
The reviewer is right. We added that results in Table 3 are averages.
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.
We included a plot (Figure 3) showing the annual and global FT contribution to the AOD. As supposed by the reviewer, the peak at the Equator in Figure 4 is mostly due to the dust/biomass burning in Africa and Indonesia, and the Arctic FT contribution is more obvious.
Page 19 Figure 4 caption – replace 'full' with 'solid'; replace 'show' with 'indicate'
Done.

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
Weigum et al 2016: https://www.atmos-chem-phys.net/16/13619/2016/