Reviewer 2:

Title: Intra-annual variations of regional aerosol optical depth, vertical distribution, and particle types from multiple satellite and ground-based observational datasets

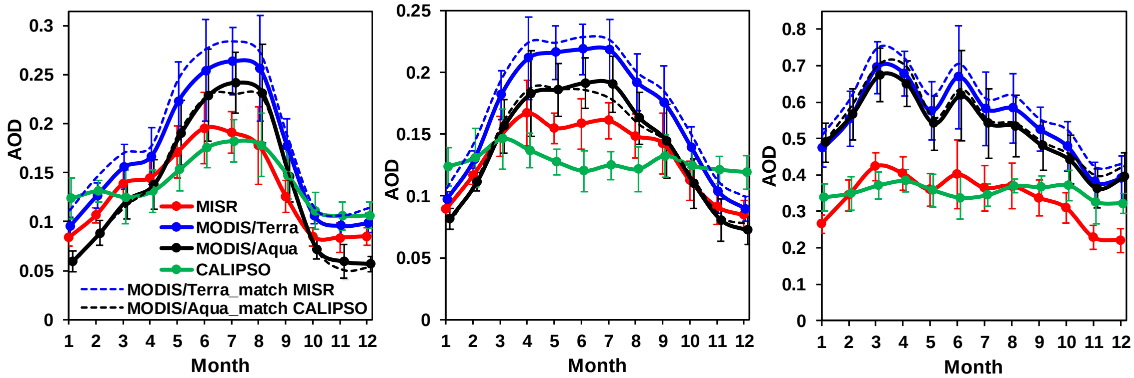
Summary: The paper combines retrievals and observations from multiple satellites (active and passive) and ground based (in-situ and remote sensed), in order to characterize the seasonal and diurnal variations of aerosol properties in three heavily-populated regions (EUS, WEU and ECC). The aerosols are separated into lower (< 800 m) and higher levels (> 800m), monthly averages are calculated, and annual cycles plotted. Analysis and interpretation and some speculation are presented. The main conclusions are that in all three regions, column AOD and higher level AOD all peak in the summer, whereas AOD in lower levels peaks during the winter. AOD from fine-sized particles peaks in the spring/summer and is attributed to anthropogenic sources. Dust and sea-salt peaks in the winter in WEU but are nearly constant in the other two regions. There appears to be larger nighttime/daytime AOD difference in summer than winter.

Response: We thank the reviewer for the insightful comments. We have carefully addressed these comments in revising the manuscript. Point-to-point responses are given below.

This paper is logical and easy to read. The English language usage is satisfactory. The idea of separating into low level (e.g. < 800 m; presumably a proxy for boundary layer) and higher level (> 800 m) is unique. I wish I could believe all of the conclusions. But I don't yet. Like I explained in the "initial quality" review, I have strong concerns about data sampling. For example, Colarco et al., (2014, [10.5194/amt-7-2313-2014]) explains that "sampling matters", and that when we develop climatology from different types of orbits (and coverage), we need to deal with this problem. Because of this, I don't think that "the impact of the sampling issue is expected to be much smaller than that on the AOD retrieval in an individual month at a specific location" (Lines 162-167). If MODIS calculates monthly mean based on all 30 days and CALIPSO based on 2-4 times month (every 8 or 16 days, if lucky), then we don't expect the monthly means to match. Of course, if there are clouds, this could be MODIS making monthly means from, say 10 days, and CALIPSO making only one. One more paper to think about is Chin et al., (2014; [10.5194/acp-14-3657-2014]). Although they study multi-year data, they make points about comparing datasets with all kinds of sampling differences.

Response: We thank the reviewer for this valuable comment. To investigate the impact of data sampling on seasonal variation of AOD, we design two sensitivity cases: a "MODIS/Terra_match MISR" case in which the monthly mean AOD of MODIS/Terra is calculated using only the days when MISR overpasses, and a "MODIS/Aqua_match CALIPSO" case in which the monthly mean AOD of MODIS/Aqua is calculated using only the overpassing days of CALIPSO. The results are illustrated in the following figure (Fig. 2 in the revised manuscript). In all three regions, the monthly mean AODs are slightly different for "MODIS/Terra" and "MODIS/Terra_match MISR", but the seasonal variation patterns are largely the same. The same results are found for "MODIS/Aqua" and "MODIS/Aqua_match CALIPSO". As such, we conclude that sampling has little effect on the AOD seasonal variation patterns reported in this study. In fact, this conclusion is compatible with the findings of Colarco et al. (2014). Colarco et al. (2014) revealed that the spatial sampling artifacts were significant for fine aggregation grid (e.g., 0.5°), but they are reduced at coarse grid scales (e.g., 10°). In this study, we use only the mean AOD over three large regions (about 20°×20°) across 10 years, therefore the sampling artifacts are expected to be even smaller.

We have added the preceding discussions in the revised manuscript. (Line 208-224)



(a) (b) (c)

Figure. Monthly mean AOD observed by MISR, MODIS/Terra, MODIS/Aqua, and CALIPSO during 2007-2016 in (a) EUS, (b) WEU, and (c) ECC. For CALIPSO, only clear-sky daytime profiles are averaged in order to be consistent with the MISR and MODIS products. "MODIS/Terra_match MISR" is a sensitivity case in which the monthly mean AOD of MODIS/Terra is calculated using only the days when MISR overpasses, and "MODIS/Aqua_match CALIPSO" is a case in which the monthly mean AOD of MODIS/Aqua is calculated using only the overpassing days of CALIPSO. The error bars denote the standard deviation of the monthly mean AOD values obtained over all years. Note the different scales on the y-axes of the plots.

Of course, the low bias (Fig 2, lines 216-218) between CALIPSO -derived AOD and the other satellites (MODIS, MISR), can be because of assumed lidar ratios (Ma et al., AMT; [10.5194/amt-6-2391-2013]), or undetected aerosol layers (Kim et al., JGR, [10.1002/2016JD025797]). I guess that the Thorson (2017) reference already listed could be a reason as well.

Response: Thank you! We have added these explanations to the revised manuscript, citing the two references. (Line 315-318)

Why the non-confident statements ("probably") in lines 227-231? I think you should be able to find references. What are the sources of these SOA? What about long-range transport in the summer?

Response: To address the reviewer's comments, we have revised these sentences as follows:

Considering the high accuracy of AERONET, we conclude that AOD peaks in summer/spring and dips in winter. An important reason for the higher AOD in summer is that the stronger radiation and higher temperature accelerate the formation of secondary aerosols (Timonen et al., 2014), including sulfate, nitrate, ammonium, and secondary organic aerosol (SOA). SOA is produced by photo-oxidation of volatile organic compounds (VOCs) and intermediate volatility organic compounds (IVOCs), as well as the chemical aging of primary organic aerosol (Zhao et al., 2016). Another reason is that more abundant water vapor in summer favors the hygroscopic growth of aerosols (Liu et al., 2012; Zheng et al., 2017). The different patterns of long range transport as a function of season is also partly responsible for the seasonable variation of AOD (Tian et al., 2017; Yang et al., 2018; Garrett et al., 2010). (Line 232-242)

Considering lines 235-236, I again ask about sampling? Are the monthly means for AERONET and satellites computed based on the same days? Or is mean AERONET = mean (of AERONET data) and mean satellite = mean (of SATELLITE data)? We know from validation exercises that when actually collocated in space and time (both AERONET and satellite are free of clouds) that they match overall well (yes, sometimes small biases, e.g. Remer et al., 2005). However, I do not expect matches if using different samples (days). Note that the Remer et al., (2005) study has been updated for MODIS (Collection 5 and Collection 6), and there are also updates for MISR evaluations. The "instrument calibration issue" (lines 243-244) would not cause such a large bias.

Response: For each satellite-borne sensor, only those days for which the satellite overpasses an AERONENT site were used in the comparisons. In other words, the monthly means for AERONET and satellites were indeed computed based on the same days. In addition, to match coincident measurements, the AERONET AOD retrievals for each site were averaged within a 2 h window centered on the satellite overpass times (about 10:30 for MISR and MODIS/Terra, and 13:30 for MODIS/Aqua and CALIPSO, depending on site location), and compared with the satellite AOD retrievals in a 1° × 1° grid box that contains the corresponding AERONET site. (Line 172-177)

Thank you for pointing out that the Remer et al. (2005) study is outdated. For MISR, however, the Kahn et al. (2010) study is applicable to the product used in this paper. We have revised the descriptions about the discrepancies among MODIS, MISR and AERONET as follows:

While relative patterns of AOD seasonal variations from observations of MISR, MODIS/Terra, and MODIS/Aqua are similar to each other and to those of AERONET, the magnitude of AOD observed by these sensors shows considerable discrepancies. In all three regions, the AOD retrieved from MODIS is larger than that from MISR, consistent with the results of previous studies (de Meij et al., 2012; Zhao et al., 2017; Chin et al., 2014; Kang et al., 2016; Qi et al., 2013). This is most likely due to differences in observing strategy, retrieval algorithms, and spatio-temporal sampling (Kahn et al., 2009). The MISR-retrieved AOD agrees well with the AERONET observations in EUS and WEU regions. In the ECC region, however, MISR underestimates the AERONET AOD, probably because there is less signal from the surface at higher AOD, which creates ambiguity that can result in the algorithm assigning too much of the top-of-atmosphere radiance to the surface (i.e., a higher surface albedo), thereby underestimating the AOD (Kahn et al., 2010). The MODIS/Terra and MODIS/Aqua overestimate the AERONET AOD to some extent in all three regions. The overestimation was also reported in two previous studies (de Meij et al., 2012; Ruiz-Arias et al., 2013) using the level 3 MODIS products (Collection 5 or 5.1). We show a relatively larger overestimation than that reported by de Meij et al. (2012) and Ruiz-Arias et al. (2013), partly because we used the AERONET AOD averaged within a 2 h window centered on the satellite overpass times while the two previous studies used the daily/monthly mean AERONET AOD in the comparisons. The daily mean AOD observed by AERONET is about 10% larger than the value during the satellite overpass times (Li et al., 2013). The reasons for the overestimation are yet to be thoroughly elucidated in future studies. (Line 243-262)

I think it is a good idea that you are comparing low-level CALIPSO to ground level PM2.5 (lines 267-269) but I wonder about the temporal sampling. Also, PM2.5 is usually a "dried" aerosol measurement whereas CALIPSO is ambient RH.

Response: In the original manuscript, the monthly mean $PM_{2.5}$ concentrations were calculated using observations in all days. Here we redo the calculation using only the days when CALIPSO overpasses an observational site (dashed lines in the following figure, shown below), and compare with the original estimates (solid lines). The results show that the temporal sampling has minor effects on the monthly mean $PM_{2.5}$ concentrations. In the revised manuscript, we used the updated calculation method (dashed lines) in order to match the CALIPSO observations.

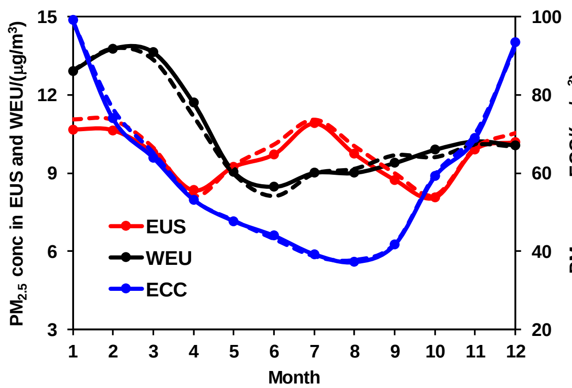


Figure. Monthly mean surface $PM_{2.5}$ concentrations during 2007-2016 in three target regions. The solid lines represent monthly mean $PM_{2.5}$ concentrations calculated using observations in all days, while the dashed lines are calculated using only the days when CALIPSO overpasses an observational site. The numbers of observational sites included in averaging are 225, 52, and 496, in the EUS, WEU, and ECC regions. Note the different scales on the y-axes for EUS/WEU and ECC.

We agree with the reviewer that the low-level AOD observed by CALIPSO is affected by ambient RH. Nevertheless, previous studies have reported fairly good correlations between extinction coefficient/low-level AOD and $PM_{2.5}$ concentrations (Cheng et al., 2013; Zheng et al., 2017). In addition, we intend to do a qualitative and not quantitative comparison. For these reasons, it appears reasonable to compare the seasonal variation patterns of low-level AOD and $PM_{2.5}$ concentrations. We have included the discussions in the revised manuscript (Line 281-284).

I don't understand the arguments in lines 279-282, in that since CALIPSO can't detect thin aerosols, that the fraction of upper-level aerosols is smaller than at the surface, and that results in the CALIPSO AOD as being weighted toward lower heights. According to the Kim et al., paper (listed above), CALIPSO is likely to miss stuff close to the ground. Anyway, the point is I don't think you can say that CALIPSO is missing stuff, and yet it "provides valuable information with respect to intra-annual variations at specific height ranges" (line 289-290).

Response: Indeed, the aerosols with heights below 200 m AGL are frequently undetected because of surface contamination (Kim et al., 2017; NASA CALIPSO team, 2011), but the overall fraction of aerosols detected in the upper levels (> 800 m AGL) is still much smaller than that in the lower levels (< 800 m AGL) because the upper-level aerosols tend to be optically thin. This is evident from Fig. 10 of Kim et al. (2017) and Fig. 1 of Thorsen et al. (2017). Therefore, the CALIPSO-observed AOD seasonal variations are significantly weighted toward lower heights.

The detection sensitivities in the upper and lower levels differ significantly because the extinction coefficient decreases by about 2 orders of magnitude with an increase of height (Kim et al., 2017; Thorsen et al., 2017). Within a specific height range, however, the optical thickness of aerosols and hence the detection fraction has a smaller variability. This is supported by the fact that the seasonal mean AOD within a specific height range differs by at most 3 times as a function of season (Fig. 4 in the main text). For these reasons, we argue that CALIPSO could provide valuable information with respect to seasonal variations of aerosols within a specific height range.

We have added the preceding discussions in the revised manuscript. (Line 295-304, 309-315)

Thank you for adding many references in lines 292-304 to discuss why AOD seasonal differences should be different in lower versus higher altitudes. I don't know if I agree that "seasonal variations of AOD at different levels are influenced by variations of RH which affects hygroscopic growth" (Lines 301-303). Of course, RH influences AOD, but it is total column water vapor and not necessarily RH that changes drastically from season to season.

Response: Following the reviewer's comment, we have changed "RH" to "water vapor amount".

I agree that comparing MISR-derived aerosol "types (size/ absorption)" and CALIPSO derived aerosol "types" (sources) is ultimately useful. (lines 307-308). However, they are clearly different beasts, and I am getting lost reading this section (Section 3.3). Each paragraph has multiple sentences that are "A implies B, whereas (while / in contrast) sometimes C implies D". It's hard to follow. I suggest a table, or schematic cartoons, or bullets.

Response: Following the reviewer's comment, we have added a table summarizing the seasonal variations of different aerosol types in the three regions (shown below). We have also carefully revised the text of this section improve the logic and readability. (Line 333-408)

Table 1. Summary of the seasonal variations of the total, height-specific, and type-specific AOD

	EUS	WEU	ECC
Total column AOD	Peak in summer	Peak in summer/late spring	Peak in summer/spring
AOD > 800 m AGL	Peak in summer	Peak in summer/late spring	Peak in summer/spring
AOD < 800 m AGL	Two peaks in winter and summer	Peak in winter	Peak in winter
Small-size	Peak in summer	Peak in summer/late spring	Peak in summer/spring
Medium-size	Peak in summer	Peak in summer/late spring	Peak in summer/spring
Large-size	Rather uniform	Rather uniform	Peak in spring
Absorbing	Peak in summer	Peak in summer/late spring	Two peaks in Mar and Aug
Polluted continental/dust	Similar to height-specific total AOD	Similar to height-specific total AOD	Similar to height-specific total AOD
Dust	No obvious seasonal pattern	Peak in summer	Peak in spring
Clean marine	No obvious seasonal pattern	Peak in winter	Negligible amount
Smoke	Peak in summer	Peak in summer/late spring	Two peaks in Mar and Aug

I notice that regarding the aerosol typing as seen by CALIPSO, all of the regions (over land), have non-trivial amount of "clean-marine" aerosol (Fig 7). Is this transported marine aerosol to the entirety of the regional box, or should the marine aerosol be expected to be more dominant but confined only to the coastal areas of a region?

Response: All three regions used in the study cover some ocean areas (see Fig. 1 in the manuscript). The marine aerosols are predominantly located over the ocean and in coastal areas, and are much fewer over land.

The section on daytime/nighttime variability is nice, but I think it is beside the point of the rest of the paper. Why would smoke AOD accumulate at night? Higher RH at night might make bigger aerosols, but if anything, fire activity is reduced at night. You might check the PM2.5 measurements here. I suggest leaving this section out, and thinking about the questions related to the other sections. "Intra-annual", "vertical", and "particle types" is enough for one paper!

Response: Following the reviewer's suggestion, we have left out this section in the revised manuscript.

In terms of figures. I can see why the authors do this (different magnitudes of AOD and or PM2.5 at different sites), but the varying y-axes within figure captions, and from figure-to-figure are distracting. But thank you for pointing out in the caption!

Response: We tried to unify the scales of the y-axes but failed because the magnitude differs greatly according to figures. Thank you for your understanding.

What is “upper air”? I see it a few places, and assume you mean > 800 m AGL? (e.g. Line 300).

Response: Yes, it means > 800 m AGL. We have explained this in the revised manuscript. (Line 299)

The abstract suggests (lines 37-38) that results can “help to improve the current estimates of climatic and health impacts of aerosols”. Well maybe, but I would drop this from the abstract since there is no discussion in the paper.

Response: Following the reviewer’s comment, we have removed this sentence from the abstract.

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