Reviewer 1:

Aerosol radiative effect is a hot topic in the science community, which is dependent on the aerosol optical properties, size distribution, aerosol types, and their vertical distribution. While many studies have examined the seasonal and diurnal variations of aerosols, few studies have examined the vertical distributions of aerosol amount and types, which can strongly affect the aerosol radiative effect and corresponding thermal impacts on the profiles of temperature. Using multi-satellite observations along with the surface observations of aerosols, this study examines the seasonal and diurnal variations of aerosol column loading, vertical distribution, and particle types over three populous regions: the Eastern United States (EUS), Western Europe (WEU), and Eastern and Central China (ECC). Interesting statistical characteristics about the dominant aerosol types and vertical distributions have been obtained. The paper is also organized and written well.

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

Detailed Comments:

Line 45-48: I am not sure if the climate effects of CO2 also depends on its intra-annual variability, particularly considering the higher outgoing longwave radiation from surface in summer than in winter.

Response: We no longer mention the climate effects of CO_2 in the revised manuscript. The revised sentence is as follows:

Therefore, the climatic and health effects of aerosols are not only induced by inter-annual concentration changes, but also strongly depend on their intra-annual variability. (Line 41-43)

Line 49-54: Not only the scattering and absorption properties, but also the size distribution and vertical distribution of aerosols can also cause problems. Zheng et al. (2017, ACP) have shown that the vertical distribution of AOD could have strong impacts on the aerosol concentration (mass concentration) within PBL, which directly affect cloud properties and then change cloud radiative forcing. Garrett et al. (2004, GRL) showed that nucleation mode aerosols and accumulation mode aerosols have very different scattering radiative effects.

Response: Following this comment, we have described the impact of size distribution and vertical distribution and cited these two references. The revised text is as follows:

However, the wide ranges of particle optical properties and size distribution mean that even for the same AOD, different aerosol components have different effects on not only the magnitude, but also the sign, of aerosol radiative forcing (IPCC, 2013; Gu et al., 2006; Garrett et al., 2004). (Line 46-49)

Besides aerosol type, the aerosol vertical distribution influences its mass concentration within the planetary boundary layer (PBL) (Zheng et al., 2017) and the vertical profile of heating rate (Johnson et al., 2008; Guan et al., 2010; Zhang et al., 2013), which subsequently modifies the atmospheric stability and convective strength (Ramanathan et al., 2007), with potential changes in cloud properties (Johnson et al., 2004). (Line 54-58)

Line 54-58: Besides to the TOA radiative balance, convective clouds development, absorptive and non-absorptive aerosols have different impacts on the surface radiative cooling effects, as shown by Yang et al. (2016, JGR) which demonstrated that more absorptive aerosol can cause more surface cooling effects.

Response: We have mentioned the different impacts of absorptive and non-absorptive aerosols on surface radiative cooling effects in the revised manuscript:

Furthermore, absorbing and non-absorbing aerosols have been found to have very different impacts on the surface radiative cooling effects (Yang et al., 2016) and the development of convective clouds (Massie et al., 2016; Ramanathan et al., 2005; Rosenfeld et al., 2008). (Line 51-54)

Line 59-63: The vertical distributions are also important for aerosol-cloud interaction since only the aerosols near cloud bases have strong interaction with cloud properties. Zhao et al. (2018, Earth and Space Science) showed Twomey effect using in-situ aircraft observations in East China instead of anti-Twomey effect found using column aerosols based on satellite observations.

Response: Following the reviewer's comments, we have added this point in the revised manuscript:

Understanding aerosol variability as a function of height is also important because the indirect effect of aerosols is mainly dependent on those mixed with the clouds (Zhao et al., 2018b). (Line 59-60)

Line 101-103: You may indicate the data observation time, such as day time and night time.

Response: We have added the following description in the revised manuscript:

The aerosol retrievals from MISR and MODIS are only available for clear-sky conditions in the daytime. CALIPSO provides retrievals during both day and night, but only clear-sky daytime profiles are used in order to be consistent with the products from MISR and MODIS. (Line 99-102)

Line 158: I would suggest "variations of AODs with heights below 200 m"

Response: Done. Thank you! (Line 155)

Line 162-167: The different time representation errors could be noticeable for monthly average. For example, Wang and Zhao (2017, JGR) showed that the MODIS cloud time representation errors could be up to 3-4% for monthly average (10 years data) while much smaller for yearly average and much larger for daily average. However, this might not affect the findings/conclusions of this study. You may simply indicate/inform the noticeable time representation error of 3-4% (Wang and Zhao, 2017) while what they studied are cloud coverage instead of aerosols.

Response: In the revised manuscript, we have investigated the impact of spatiotemporal sampling on seasonal variations of AOD using two sensitivity scenarios, and cited this reference. The added text is shown as follows:

As described in Section 2.1, MODIS provides near-daily global coverage but MISR and CALIPSO do not. As a result, the monthly mean AOD from different sensors is calculated based on different sets of days, which might lead to uncertainties in the estimation of monthly mean AOD (Colarco et al., 2014; Wang and Zhao, 2017). To rule out the impact of spatio-temporal sampling on seasonal variation patterns, 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 Fig. 2. 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. (Line 208-224)

Line 267-269: I highly agree with this analysis. By doing this, the effects of PBL and relative humidity could be minimized.

Response: Thank you!

Line 303-304: Yang et al. (2018, AR) also showed the winter vs summer patterns (high in winter, low in summer) of inter-regional transport (between pearl river delta and Hongkong) of aerosols; Garrett et al. (2010, Tellus B) also showed the strong transport in winter and weak transport in fall for aerosols from mid-latitudes to Arctic. Actually, the vertical distribution of aerosols is very interesting in the Arctic, with high values at heights around 2-7 km, mainly caused by long-range transport from other regions such as mid-latitudes.

Response: Thank you for providing these useful references. We have included them to support the point that the seasonal variations of AOD at different vertical levels are influenced by the seasonal patterns of inter-regional transport of aerosols. (Line 328-331)

Line 374, acts -> act, strengthens -> strengthen

Response: Done. Thank you! (Line 401-403)

Line 400-405, Since AOD includes the impacts of relative humidity, is it possible that relative humidity also contributes to the diurnal variation of AOD? Another possible contribution might be

the growth of fine aerosols, as indicated by Zhao et al. (2018, AAS), the growth rates of fine mode aerosols generally starts from the morning time with a growth rate of around 2-7 nm/hour (1.7-6.5 nm/hour near Beijing region in summer). They could become accumulation mode from fine mode aerosols at night time or on next day (then day time) making AOD larger.

Response: Thank you for your suggestions. We have removed the section about diurnal variations in the revised manuscript following the 2nd reviewer's comments. We hope this is acceptable for you.

Line 462-465: I agree that absorbing aerosols could play different roles to convection and clouds from non-absorbing aerosols. However, I think both absorbing aerosols and non-absorbing aerosols will reduce the direct solar radiation reaching the surface, causing surface cooler and further affect convection and clouds in similar way (of course, no absorption of solar radiation in the air), while the effects could be weaker than absorbing aerosols.

Response: Following the reviewer's comment, we have revised these sentences as follows:

Both absorbing and non-absorbing aerosols could invigorate deep convection by serving as cloud condensation nuclei and affect convection by reducing downward solar radiation and causing surface cooling (Rosenfeld et al., 2008). However, absorbing aerosols play unique roles in convection and cloud development by heating the atmosphere. This inhibits convection in most situations (Ramanathan et al., 2005; Massie et al., 2016; Zhao et al., 2018a) but may enhance convection and cloud formation above the PBL (Wang et al., 2013; Bond et al., 2013), depending on the vertical distribution of absorbing aerosols. (Line 445-452)

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)

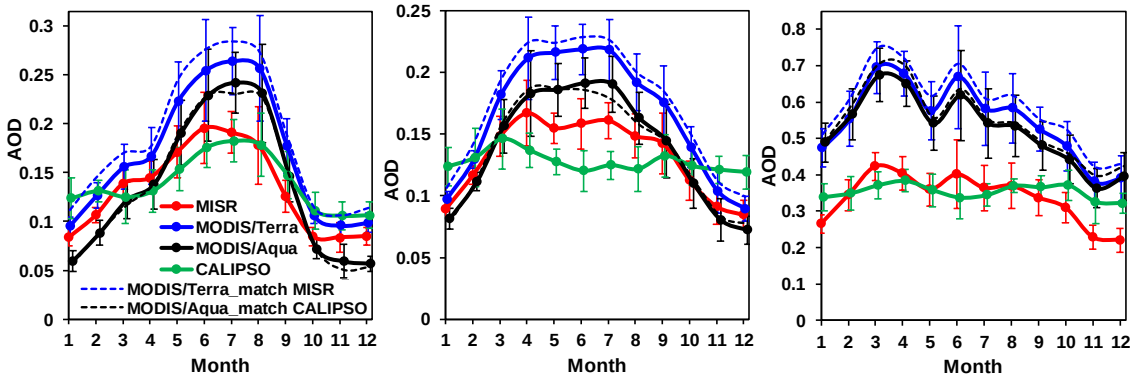

Figure. Monthly mean AOD observed by MISR, MODIS/Terra, MODIS/Aqua, and CALIPSO during 2007-2016 in (a) EUS, (b) WEU, and (c) ECC. For CALIPSO, only clear-sky daytime profiles are averaged in order to be consistent with the MISR and MODIS products. "MODIS/Terra_match MISR" is a sensitivity case in which 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.

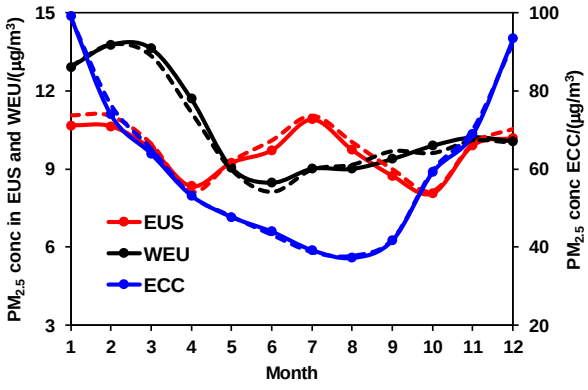

Figure. Monthly mean surface $PM_{2.5}$ concentrations during 2007-2016 in three target regions. The solid lines represent monthly mean $PM_{2.5}$ concentrations calculated using observations in all days, while the dashed lines are calculated using only the days when CALIPSO overpasses an observational 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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Intra-annual variations of regional aerosol optical depth, vertical distribution, and particle types from multiple satellite and ground-based observational datasets

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Abstract

The relatively short lifetimes of aerosols in the atmosphere result in climatic and health effects that are strongly dependent on intra-annual variations in particle concentrations. While many studies have examined the seasonal variations of regional aerosol optical depth (AOD), understanding the temporal variations in aerosol vertical distribution and particle types is also important for accurate computation of aerosol radiative effects. In this paper, we combine the observations from four satellite-borne sensors and ground-based AOD and fine particle ($PM_{2.5}$) measurements to investigate the seasonal variations of aerosol column loading, vertical distribution, and particle types over three populous regions: the Eastern United States (EUS), Western Europe (WEU), and Eastern and Central China (ECC). In all three regions, column AOD, as well as AOD higher than 800 m above ground level, peaks in summer/spring probably due to accelerated formation of secondary aerosols and hygroscopic growth. However, AOD at height below 800 m mostly peaks in winter except that a second maximum in summer occurs over the EUS region, which is consistent with observed temporal trends in surface $PM_{2.5}$ concentrations. AOD due to fine particles (< 0.7 μm diameter) is much larger in spring/summer than in winter over all three regions, whereas coarse mode AOD (> 1.4 μm diameter) generally shows less variability, except for the ECC region where a peak occurs in spring, consistent with the prevalence of airborne dust during this season. When aerosols are classified according to sources, the dominant type is associated with anthropogenic air pollution, which has a similar seasonal pattern as total AOD. Dust and sea-spray aerosols in the WEU region peak in summer and winter, respectively, but do not show an obvious seasonal pattern in the EUS region. Smoke aerosols, as well as absorbing aerosols, present an obvious unimodal distribution with a maximum occurring

in summer over the EUS and WEU regions, whereas they follow a bimodal distribution with peaks in August and March (due to crop residue burning) over the ECC region.

1 Introduction

Aerosols have adverse effects on human health (Lelieveld et al., 2015) and play a key role in Earth's climate through aerosol-radiation interactions (McCormick and Ludwig, 1967) and aerosol-cloud interactions (Twomey, 1977; Albrecht, 1989; Garrett and Zhao, 2006). Compared with long-lived climate forcers such as CO_2, aerosols have relatively short lifetimes and hence large spatiotemporal variability (Unger et al., 2008; Shindell et al., 2009). Therefore, the climatic and health effects of aerosols are not only induced by inter-annual concentration changes, but also strongly depend on their intra-annual variability.

Aerosol optical depth (AOD) has been widely used to represent the column aerosol loading and to assess the aerosol impacts on radiation, clouds, and precipitation (Ma et al., 2014; Niu and Li, 2012; Zhao et al., 2018b; Song et al., 2017). However, the wide ranges of particle optical properties and size distribution mean that even for the same AOD, different aerosol components have different effects on not only the magnitude, but also the sign, of aerosol radiative forcing (IPCC, 2013; Gu et al., 2006; Garrett et al., 2004). IPCC (2013) estimates that the historical global mean direct radiative forcings due to sulfate, organic carbon (OC), black carbon (BC), and mineral dust are −0.40, −0.19, +0.36, and −0.10 W m^{-2}, respectively. Furthermore, absorbing and non-absorbing aerosols have been found to have very different impacts on the surface radiative cooling effects (Yang et al., 2016) and the development of convective clouds (Massie et al., 2016; Ramanathan et al., 2005; Rosenfeld et al., 2008). Besides aerosol type, the aerosol vertical distribution influences its mass concentration within the planetary boundary layer (PBL) (Zheng

et al., 2017) and the vertical profile of heating rate (Johnson et al., 2008; Guan et al., 2010; Zhang et al., 2013), which subsequently modifies the atmospheric stability and convective strength (Ramanathan et al., 2007), with potential changes in cloud properties (Johnson et al., 2004). Understanding aerosol variability as a function of height is also important because the indirect effect of aerosols is mainly dependent on those mixed with the clouds (Zhao et al., 2018c). Meanwhile, the health impacts of aerosols are only associated with those present near the surface, where they are inhaled. For these reasons, systematic analyses of the intra-annual variations of aerosol vertical distribution and particle types, in addition to total column AOD, are necessary to improve our understanding of aerosol climatic and health effects.

Numerous studies have investigated the seasonal variations of AOD at global and regional scales using satellite observations (e.g., Kim et al., 2007; Song et al., 2009; Mehta et al., 2016; Mao et al., 2014). By comparison, most previous studies of the temporal variations of aerosol vertical distributions and aerosol types have been confined to only a few sites due to coverage limitations associated with reliance on ground-based instruments (e.g., Liu et al., 2012; Matthias et al., 2004). Despite continuous advancement of remote sensing technology and emergence of new spaceborne sensors, only limited number of studies have utilized satellite observations to examine the seasonal variations of aerosol profiles and/or types at regional or larger scales (Huang et al., 2013; Kahn and Gaitley, 2015; Yu et al., 2010; Li et al., 2016). Huang et al. (2013) analyzed the seasonal variations of aerosol extinction profile and type distribution using 5-year observations from the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO). Kahn and Gaitley (2015) examined the spatiotemporal variations of aerosol types retrieved by the Multi-angle Imaging SpectroRadiometer (MISR). Different satellite-borne sensors, such as MISR, CALIPSO, and Moderate resolution Imaging Spectroradiometer (MODIS), employ different

principles of measurement and retrieval, and therefore provide different sensitivities to column AOD, aerosol types, and vertical profiles. Therefore, integration of data from multiple satellites and ground-based observational networks makes it possible to deepen our understanding of the intra-annual variations of aerosol loadings, profiles, and types.

In this study, we investigate the seasonal variations of aerosol column loading, vertical distribution, and particle types using multiple satellite and ground-based observational datasets covering the period from 2007 to 2016. The purpose is to assess the consistency among various datasets and provide a comprehensive characterization of aerosol properties in polluted regions to facilitate future studies of aerosol climate effects and local air quality issues. The data are from MISR, MODIS, CALIPSO, Aerosol Robotic Network (AERONET), and surface $PM_{2.5}$ monitors. Consistent with our previous study (Zhao et al., 2017), we selected three populous regions which have experienced substantial anthropogenic pollution (Wang et al., 2017; Wang et al., 2014) and have received considerable attention in other climate studies: the Eastern United States (EUS; 29º-45º N, 70º-98º W), Western Europe (WEU; 37º-59º N, 10º W-17º E), and Eastern and Central China (ECC; 21º-41º N, 102º-122º E). The geographical boundaries of these regions are shown in Fig. 1.

2 Data and Methods

2.1 Satellite data

We obtain retrievals of total column AOD as well as AOD for various height ranges and aerosol types during 2007-2016 from MISR (flying on the Terra satellite), MODIS (Terra and Aqua), and the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) on CALIPSO. The aerosol retrievals from MISR and MODIS are only available for clear-sky conditions in the

daytime. CALIPSO provides retrievals during both day and night, but only clear-sky daytime profiles are used in order to be consistent with the products from MISR and MODIS.

MISR observes the Earth with moderately high spatial resolution (275 m to 1.1 km) at 9 along-track viewing angles in each of 4 visible/near-infrared spectral bands, which enables the partitioning of AOD by particle type over both land and ocean, in addition to retrieval of total AOD (Kahn and Gaitley, 2015; Kahn et al., 2001). Its observations provide near-global coverage every 9 days (Diner et al., 1998). We make use of the Level 3 daily global aerosol product (MIL3DAE) version F15_0031, which is generated at a spatial resolution of 0.5° × 0.5°. The variables used in the analysis are total AOD at 555 nm as well as AODs for six aerosol components, namely small (< 0.7 μm diameter), medium (0.7-1.4 μm diameter), large (> 1.4 μm diameter), spherical, non-spherical, and absorbing. Based on comparison with ground-based AERONET measurements, the errors in MISR AOD data are on the order of ±0.05 or ±(0.20 × AOD), whichever is larger (Kahn et al., 2005; Kahn et al., 2010). In addition, retrieval of MISR particle property information from individual retrievals is considered to be reliable when AOD > 0.15, and has diminished sensitivity at smaller AOD (Kahn and Gaitley, 2015; Kahn et al., 2010). In this study we use only monthly mean values, for which the uncertainties are expected to be smaller than those for individual retrievals. Note that we did not do a relative humidity (RH) correction to AOD retrievals from MISR as well as other sensors. The seasonal variations of AOD represent a combined effect of variations in aerosol abundance, vertical distribution, chemical constituents, and meteorological conditions.

The MODIS sensors onboard the Terra and Aqua satellites observe the Earth with multiple wavelength bands over a 2330 km swath (King et al., 2003), which provides near-daily global coverage. In this study we obtain column AOD data at 550 nm with a 1° × 1° resolution from the

Level 3 daily atmosphere products Collection 6 (MOD08 and MYD08 for the Terra and Aqua platforms, respectively). Comparison studies with AERONET have estimated the accuracy of AOD retrievals to be about $\pm(0.05 + 0.15 \times AOD)$ over land and $\pm(0.03 + 0.05 \times AOD)$ over ocean (Levy et al., 2010; Remer et al., 2005). For both MISR and MODIS data, we calculate regional mean AOD by averaging valid AOD values over all grids within the three target regions.

CALIOP is a dual-wavelength polarization lidar on the CALIPSO satellite, and is designed to acquire vertical profiles of aerosols and clouds at 532 and 1064 nm wavelengths [Winker et al., 2007]. CALIPSO flies in formation with Aqua, and all three satellites employed in this paper fly in orbits having 16-day repeat cycles. In addition to vertical extinction profiles, CALIPSO categorizes an aerosol layer as one of seven types based on a number of parameters including altitude, location, surface type, volume depolarization ratio, and integrated attenuated backscatter [Omar et al., 2009]. The seven aerosol types are dust, smoke, clean continental, polluted continental, polluted dust, clean marine, and dusty marine. For most profiles, this aerosol classification is consistent with that derived from AERONET inversion data (Mielonen et al., 2009). In this study, we adopt the Level 2 aerosol profile product (05kmAPro, V4.10), which has an along-track horizontal resolution of 5 km and a vertical resolution of 60 m or 180 m, depending on whether the aerosol height is below or above 20.2 km altitude. We do not use the CALIOP Level 3 product because it is difficult to collocate with AERONET observations (see Section 2.2) due to its coarse resolution ($2° \times 5°$). For each clear-sky profile, we calculate the column AOD at 532 nm by vertically integrating extinction coefficients of the features that are identified as "aerosols" and have valid quality control (QC) flags, i.e., $-100 \leq$ cloud aerosol discrimination (CAD) score ≤ -20, extinction QC = 0/1, and extinction coefficient uncertainty < 99.9 (Huang et al., 2013). In addition, we employ two quality filters used in generating the Level 3 product in

order to eliminate features that probably suffer from surface contamination, i.e., near-surface features with large negative extinction coefficients and contaminated features beneath the surface-attached opaque layer (NASA CALIPSO team, 2011). Following the same method, we also bin the 532 nm AODs into various height ranges, i.e., 0-200 m, 200-500 m, 500-800m, 800-1200 m, 1200-2000 m, and > 2000 m above ground level (AGL). Finally, we derive monthly mean AODs by averaging all clear-sky aerosol profiles within each month over the three target regions. Although aerosol extinction coefficients with heights below 200 m AGL are considered to be uncertain despite the application of quality filters (NASA CALIPSO team, 2011), we include them for completeness but exercise with caution when interpreting variations of AODs below 200 m. It should be noted that CALIPSO AOD is reported at a different wavelength (532 nm) from those used in the MISR and MODIS products (555 nm and 550 nm, respectively); this slight wavelength difference is not expected to affect our conclusions regarding AOD seasonal variations.

2.2 AERONET and surface $PM_{2.5}$ data

We use AOD observations from AERONET to compare with the AOD seasonal variations derived from satellite datasets. AERONET sunphotometers directly measure AOD at seven wavelengths (approximately 340, 380, 440, 500, 675, 870, and 1020 nm) with an estimated uncertainty of 0.01-0.02 (Holben et al., 2001; Eck et al., 1999), which is much smaller than the uncertainties associated with satellite measurements (Kahn et al., 2010; Levy et al., 2010; Schuster et al., 2012). Therefore, we consider AERONET as “ground truth” for AOD temporal variations. We adopt the AERONET Level 2 Version 2.0 direct-sun measurements of spectral AODs, which are subsequently interpolated to 550 nm using a second-order polynomial fit to ln(AOD) vs. ln(wavelength) as recommended by Eck et al. (1999). A fundamental difference between satellite and AERONET AOD observations is that a satellite acquires data at a single overpass time (or

spread over 7 minutes for MISR's nine views) and over an extended spatial area in the case of MISR and MODIS, whereas AERONET obtains a time series of point data at each surface station. To match coincident measurements, the AERONET AOD retrievals for each site are 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 (consistent with the grids used in the MODIS Level 3 products) that contains the corresponding AERONET site. Only those days for which a satellite overpasses an AERONENT site are used in the comparisons. Since AOD variation has a large spatial correlation length of 40-400 km (Anderson et al., 2003), spatial averaging over a 1° × 1° grid should not bias the seasonal variations of AOD but has the benefit of increase the number of data points with valid AOD retrievals that are used in the comparisons. To assure data quality, only the AERONET sites that span at least 5 years with at least 10 months of valid data in each year are included in the comparison. After screening, 28, 54, and 13 sites are used in our analysis of the EUS, WEU, and ECC regions.

To provide additional information on the seasonal variations of satellite-observed aerosol loadings near the surface, we obtain surface $PM_{2.5}$ concentrations from several observational networks over the three target regions. Hourly $PM_{2.5}$ concentrations for 225 sites over the EUS region are achieved from the Air Quality System (AQS), which is a large observational database containing ambient air pollution data collected by the United States Environmental Protection Agency (USEPA), as well as state, local, and tribal air pollution control agencies in the United States (USEPA, 2015). For the ECC region, we obtain hourly $PM_{2.5}$ concentrations from the Ministry of Environmental Protection of China (MEP, http://datacenter.mep.gov.cn/), which provides continuous measurements at 496 sites located in 74 major cities in China. Hourly/daily

193 $PM_{2.5}$ concentrations for 52 sites over the WEU region are taken from the European Monitoring
194 and Evaluation Programme (EMEP). Similar to the processing of AERONET data, we only include
195 sites whose data span ≥ 5 years with ≥ 10 months of data in each year, except in the case of the
196 ECC region where at least 2 years' data are required because the $PM_{2.5}$ concentrations have been
197 only publicly available since January 2013.

198 **3 Results and Discussion**

199 3.1 Seasonal variations of column AOD

200 Figure 2 illustrates the monthly variations in column AOD observed by MISR,
201 MODIS/Terra, MODIS/Aqua, and CALIPSO during 2007-2016 in the three target regions. All
202 satellite-borne sensors show that AOD in the EUS region is the highest in summer and lowest in
203 winter, though CALIPSO reports a noticeably smaller difference between the summer and winter
204 extrema compared with the other three satellite instruments. For the WEU and ECC regions, MISR,
205 MODIS/Terra, and MODIS/Aqua also reveal consistent seasonal patterns in which AOD peaks in
206 spring and/or summer and reaches its lowest valley in winter. CALIPSO, however, shows little
207 intra-annual variation in AOD, with small peaks occurring in spring and fall.

208 As described in Section 2.1, MODIS provides near-daily global coverage but MISR and
209 CALIPSO do not. As a result, the monthly mean AOD from different sensors is calculated based
210 on different sets of days, which might lead to uncertainties in the estimation of monthly mean
211 AOD (Colarco et al., 2014; Wang and Zhao, 2017). To rule out the impact of spatio-temporal
212 sampling on seasonal variation patterns, we design two sensitivity cases: a "MODIS/Terra_match
213 MISR" case in which the monthly mean AOD of MODIS/Terra is calculated using only the days
214 when MISR overpasses, and a "MODIS/Aqua_match CALIPSO" case in which the monthly mean
215 AOD of MODIS/Aqua is calculated using only the overpassing days of CALIPSO. The results are

illustrated in Fig. 2. 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.

In view of the substantial differences between CALIPSO and the other three sensors, we compare satellite retrieved AOD seasonal variations with point-based ground measurements from AERONET (Fig. 3). As in other studies, AERONET data are treated as "ground truth" for column AOD due to its smaller uncertainty compared with satellite data (Kahn et al., 2010; Levy et al., 2010; Schuster et al., 2012; Fan et al., 2018). Figure 3 shows that, in all three regions, the AOD seasonal variations measured by AERONET are similar to those retrieved by MISR, MODIS/Terra, and MODIS/Aqua, but are quite different from CALIPSO data. Reasons for the different seasonal patterns between CALIPSO and other sensors will be discussed in Section 3.2. 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).

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.

3.2 Seasonal variations of aerosol loadings as a function of height

In addition to column AOD, the climatic effects of aerosols are also strongly dependent on their vertical distribution. To explore intra-annual variations in aerosol vertical profile, Fig. 4 presents CALIPSO-observed monthly variations of AOD as a function of height in the three target regions. A striking pattern is that the AOD seasonal variations are dramatically different at lower and upper heights. Over the WEU and ECC regions, AODs of the vertical layers below 800 m AGL generally peak in winter, while those above 800 m AGL peak in summer/spring. As a result, the CALIPSO-observed column AOD for these two regions presents a rather uniform seasonal pattern. For the EUS region, the maximum AOD above 800 m AGL also occurs in summer; however, AOD below 800 m AGL shows two peaks, one in summer and the other in winter. The integration of various layers thus yields a nearly unimodal distribution with maximum occurring in summer.

To provide an independent evaluation of the CALIPSO-observed AOD variations at lower heights, we examine the seasonal variations of near-surface $PM_{2.5}$ concentrations at hundreds of surface monitor locations within the three target regions (Figure 5). The aerosol extinction coefficient, and hence AOD at lower heights is affected by not only the particle mass concentrations, but also aerosol type (absorbing vs. nonabsorbing aerosols, coarse-mode vs. fine-mode aerosols) and meteorological parameters such as RH, wind speed and direction, and planetary boundary layer height (Zheng et al., 2017). 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). For this reason, it is reasonable to qualitatively compare

the seasonal variation patterns of near-surface $PM_{2.5}$ concentrations and low-level AOD. We calculate monthly mean $PM_{2.5}$ concentrations using only the days when CALIPSO overpasses an observational site to enable a better comparison. Figure 5 shows that, over the ECC and WEU regions, surface $PM_{2.5}$ concentrations are largest in winter and smallest in summer. In the EUS region, the maximum $PM_{2.5}$ concentration occurs in summer and a second maximum occurs in winter. These trends are generally consistent with the seasonal variations of AOD at low heights, implying that CALIPSO data can generally capture the seasonal changes in low-level aerosol abundance.

The aerosol vertical distribution is an important factor in reconciling CALIPSO and other sensors with regard to AOD seasonal variations. MISR, MODIS, and AERONET all measure column-integrated AOD using spectroradiometers, whereas CALIOP is an active lidar which estimates vertically-resolved AOD based on vertical profiles of attenuated backscatter. By comparing CALIPSO with the Atmospheric Radiation Measurement (ARM) program's ground-based Raman lidars, Thorsen et al. (2017) showed that CALIPSO does not detect all relatively significant aerosols due to insufficient detection sensitivity, and that the fraction of aerosols detected in the upper levels (> 800 m AGL) is much smaller than that in the lower levels (< 800 m AGL) because the upper-level aerosols tend to be optically thin. As a result, the CALIPSO-observed AOD seasonal variations are significantly weighted toward lower heights. Note that the aerosols with heights below 200 m AGL are frequently undetected because of surface contamination (Kim et al., 2017; NASA CALIPSO team, 2011), but this does not alter the key feature that the AOD is weighted toward lower heights. Over WEU and ECC regions, the unimodal AOD distributions with a summer peak at higher levels are largely counteracted by the opposite seasonal variations at lower levels, resulting in rather uniform seasonal variations of column AOD.

For the EUS regions, due to the bimodal AOD distribution at lower heights, the summer peak in column AOD variations remain but the difference between peak and valley is smaller than implied by the observations of MISR/MODIS/AERONET. Although the integrated CALIPSO column AOD does not agree well with AERONET, it does provide valuable information with respect to seasonal variations of aerosols within a specific height range, since the optical thickness and hence the detection fraction has a smaller variability at a given height. 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), while it decreases by about 2 orders of magnitude with an increase of height (Kim et al., 2017; Thorsen et al., 2017). Besides the seasonal variations, the difference in the magnitude of AOD between CALIPSO and other sensors are also largely explained by the undetected aerosol layers by CALIPSO (Kim et al., 2017; Thorsen et al., 2017) as well as the assumed lidar ratios in CALIPSO retrievals (Ma et al., 2013).

Why are the AOD seasonal variations different between the lower and upper levels? The atmosphere in winter is generally more stable and vertical mixing is weaker, therefore more aerosols, particularly primary aerosols, are confined to lower heights, resulting in the peak of low-level AOD in winter (Guo et al., 2016; Liu et al., 2012; Zheng et al., 2017). At higher levels, the maximum AOD in summer can be explained by two reasons: (1) more aerosols, especially primary aerosols, are transported to the upper levels in summer due to stronger vertical mixing (Guo et al., 2016; Liu et al., 2012; Zheng et al., 2017), and (2) secondary aerosol formation is more rapid in summer because of stronger radiation and higher temperature, and much of the secondary aerosols are produced in the upper levels (de Reus et al., 2000; Minguillon et al., 2015; Heald et al., 2005). In addition, the seasonal variations of AOD at different vertical levels may also be influenced by the variations of water vapor amount which affects the hygroscopic growth (Liu et al., 2012; Zheng

et al., 2017) as well as the seasonal patterns of inter-regional transport of aerosols (Tian et al., 2017; Yang et al., 2018; Garrett et al., 2010).

3.3 Seasonal variations of aerosol types

Besides column AOD and vertical profiles, another factor influencing aerosol climate impact is aerosol type (i.e., partitioning by size and chemical composition). The MISR and CALIPSO products classify aerosols based on distinct principles of measurement and retrieval algorithms. Analysis of the two datasets in combination can potentially lead to a deeper understanding of the factors driving temporal variations of aerosol type.

Figures 6 illustrates the seasonal variations of type-specific AODs retrieved by MISR. MISR distributes AODs into three size ranges, i.e., small (< 0.7 μm diameter), medium (0.7-1.4 μm diameter), and large (> 1.4 μm diameter). The ambient aerosols are comprised of primary aerosols (dust, sea-spray aerosols, and primary anthropogenic aerosols) and secondary aerosols (sulfate, nitrate, ammonium, and SOA). Among these constituents, dust and sea-spray aerosols are predominantly coarse particles and secondary aerosols are dominated by very fine particles, while primary anthropogenic aerosols span a large size range, leading to a mean size intermediate between dust/sea-spray and secondary constituents (Seinfeld and Pandis, 2006). Fig. 6 indicates that the small-size AOD is much larger in spring/summer than in winter over all regions, primarily due to accelerated secondary aerosol formation and enhanced hygroscopic growth (see Section 3.1). In contrast, large-size AOD generally shows rather uniform distributions, except for the ECC region where a peak occurs in late winter/early spring. AOD of primary anthropogenic aerosols are less influenced by seasonal effects than secondary aerosols, which partly accounts for the rather uniform distributions of large-size AOD. Additionally, the seasonal variations of large-size AOD are also affected by dust and sea-spray aerosols, as discussed below.

In contrast to MISR's partitioning of aerosol type by size and absorption, the CALIPSO-retrieved aerosol types are characterized by emission source (Fig. 7). As discussed in Section 3.2, relative variability in CALIPSO-derived AOD at different height ranges appears to be more reliable than integrated column AOD, therefore we show aerosol types below and above 800 m separately in Fig. 7. Particles associated with anthropogenic air pollution (polluted continental and polluted dust) comprise the dominant type in all three regions. The seasonal variation patterns of polluted continental/dust are in accordance with those of the total AOD. Specifically, at higher levels, the maximum AOD of polluted continental/dust aerosols occurs in spring/summer in all regions. At lower levels, however, the maximum occurs in winter (plus a second maximum in summer in EUS).

With regard to dust and clean marine (sea-spray) aerosols, the AOD in the EUS region does not show an obvious seasonal pattern. In the WEU region, AOD of dust aerosols peaks in summer, consistent with previous surface-based observational studies which show that dust events in Europe predominantly occur during summer due to transport from the Sahara region (Stafoggia et al., 2016). The AOD of dust is primarily located above 800 m, supporting the conclusion that dust aerosols in WEU mainly originate from long range transport. Since the dust AOD is subject to a large inter-annual variability (denoted by the large error bars in Fig. 7), we use the Student's t-test to demonstrate the statistical significance of the seasonal variations. The dust AOD in summer is statistically larger than that in any other season at the 0.05 level, indicating the robustness of the peak in summer. Contrary to dust, the AOD of sea-spray aerosols in WEU is much higher in winter than in summer, probably because winter is the relative windy season with large low pressure systems over the Atlantic Ocean and the North Sea (Manders et al., 2009). The offset of the opposite variation trends in dust and sea-spray aerosols partly accounts for the rather uniform

distributions of large-size AOD in WEU (see Fig. 6). Over the ECC region, sea-spray aerosols make a negligible contribution to total AOD. The dust AOD is much larger in spring than in any other season (significant at the 0.05 level), which is tied to the outburst of springtime Gobi desert dust storms (China Meteorological Administration, 2012). The high dust AOD explains the peak in large-size AOD in spring over the ECC region (see Fig. 6).

Smoke aerosols are predominantly located above 800 m in all regions. Over the EUS and WEU regions, smoke aerosols present a unimodal distribution with maximum occurring in summer. The differences between smoke AOD in summer and the other three seasons are all statistically significant at the 0.05 level, except for the difference between summer and spring over the WEU region, which is statistically significant at the 0.10 level. In the ECC region, the smoke AOD follows a bimodal distribution with peaks occurring in March and August and valleys occurring in May and December. The differences between either of the peak months and either of the valley months are statistically significant at the 0.05 level. MISR's independent retrieval of absorbing AOD (Fig. 6) presents a highly similar seasonal pattern (statistically significant at the 0.05 level) as the CALIPSO smoke AOD. In fact, smoke and absorbing aerosols are closely correlated with each other, since smoke consists of a much larger fraction of absorbing aerosols (Dubovik et al., 2002), such as BC and light-absorbing organic aerosol (Kirchstetter and Thatcher, 2012), as compared to other aerosol types. Besides, the MISR absorbing AOD and CALIPSO smoke AOD are also consistent in the order of magnitude. The variability of MISR absorbing AOD (shown in the right Y-axis of Fig. 6) is about 0.002-0.005, while the variability of smoke AOD from CALIPSO is about 0.01-0.03. The smoke AOD includes the contributions of both the absorbing and scattering portions. The MISR absorbing AOD, which is calculated using total AOD × (1 – single scattering albedo), represents only the absorbing portion but includes contributions from

aerosol types other than smoke (Bull et al., 2011). Considering that the single scattering albedo of smoke is about 0.80-0.94 (Dubovik et al., 2002), we are able to reconcile the magnitude of MISR absorbing AOD and CALIPSO smoke AOD. For the preceding reasons, the seasonal patterns of smoke and absorbing aerosols act as a cross-validation and strengthen the reliability of the observed trends. Over the EUS and WEU regions, the largest smoke AOD in summer could be explained by the highest emissions from forest and grassland fires (van der Werf et al., 2017). Over the ECC region, an additional peak occurs in March because agricultural residue burning makes a substantial contribution to total smoke emissions (van der Werf et al., 2017), and such burning takes place more frequently in March due to burning of crop residues left on the fields from the previous growing season (Shon, 2015).

4 Conclusions and implications

This study investigated the seasonal variations of aerosol column loading, vertical distribution, and particle types using multiple satellite and ground-based observational datasets during 2007-2016 over EUS, WEU, and ECC regions. Retrievals from MISR and MODIS reveal that column AOD in all three regions peaks in spring/summer and reaches its low in winter, which is consistent with observations from AERONET. This seasonal pattern is probably explained by accelerated formation of secondary aerosols in spring/summer due to stronger insolation and higher temperature. In contrast, CALIPSO shows a much weaker seasonal variability in column AOD, probably because CALIPSO-retrieved AOD is weighted toward lower heights since some thin aerosol layers in high levels are undetected due to insufficient detection sensitivity. Despite the discrepancy in integrated column AOD, CALIPSO does provide valuable information with respect to intra-annual variations of AOD as a function of height. Over the WEU and ECC regions, AODs of the vertical layers below 800 m generally peak in winter, while those above 800 m mostly

peak in summer. For the EUS region, the maximum AOD above 800 m also occurs in summer; however, AOD below 800 m shows two peaks, one in summer and the other in winter. The seasonal variations of AOD at low heights are consistent with seasonal patterns of measured surface $PM_{2.5}$ concentrations.

When aerosols are binned into different size ranges, the small-size AOD is much larger in spring/summer than in winter over all three regions. Large-size AOD generally shows rather uniform distributions, except for the ECC region where a peak occurs in spring, consistent with the largest dust AOD in this season. When aerosols are classified according to sources, the aerosols associated with anthropogenic air pollution (as well as mixtures of anthropogenic pollution and dust) are the dominant type in all three regions. AOD of polluted aerosols has a similar seasonal pattern as total AOD. Dust and clean marine aerosols in the WEU region peak in summer and winter, respectively, whereas they do not show an obvious seasonal pattern in the EUS region. Smoke aerosols, which CALIPSO indicates are predominantly located at heights above 800 m, present an obvious unimodal distribution with maximum occurring in summer over EUS and WEU regions, and a bimodal distribution with peaks in August and March over the ECC region. This pattern is in good agreement with the seasonal variations of absorbing AOD derived from MISR.

The combination of multiple satellite and ground-based observations facilitate a systematic and deeper understanding of the seasonal variations of aerosols, particularly their vertical and type distribution. Comparison of multiple measurement and retrieval methodologies enables reducing the uncertainties in the estimation of aerosol direct effects by providing improved information about aerosol vertical and type distributions, which significantly affect the aerosol-induced scattering and absorption of radiation. More importantly, the intra-annual variations of vertical distributions and types of aerosols are important for understanding their impact on atmospheric

dynamics, cloud fields, and precipitation production. For example, both absorbing and non-absorbing aerosols could invigorate deep convection by serving as cloud condensation nuclei and affect convection by reducing downward solar radiation and causing surface cooling (Rosenfeld et al., 2008). However, absorbing aerosols play unique roles in convection and cloud development by heating the atmosphere. This inhibits convection in most situations (Ramanathan et al., 2005; Massie et al., 2016; Zhao et al., 2018a) but may enhance convection and cloud formation above the PBL (Wang et al., 2013; Bond et al., 2013), depending on the vertical distribution of absorbing aerosols. Finally, the data and variation patterns presented in this study can be used to evaluate and improve model simulations, with the ultimate goal of improving model assessment of the climatic and health effects of aerosols.

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Figures

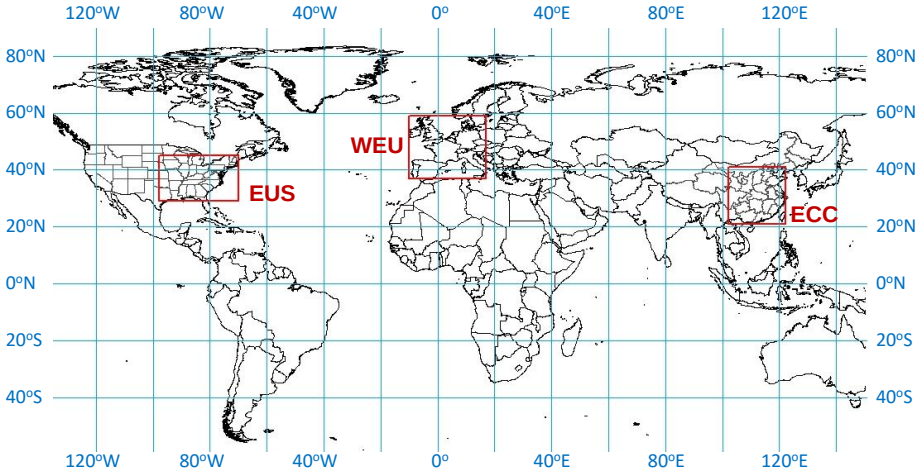


Figure 1. Target regions for this study: the Eastern United States (EUS), Western Europe (WEU), and Eastern and Central China (ECC).

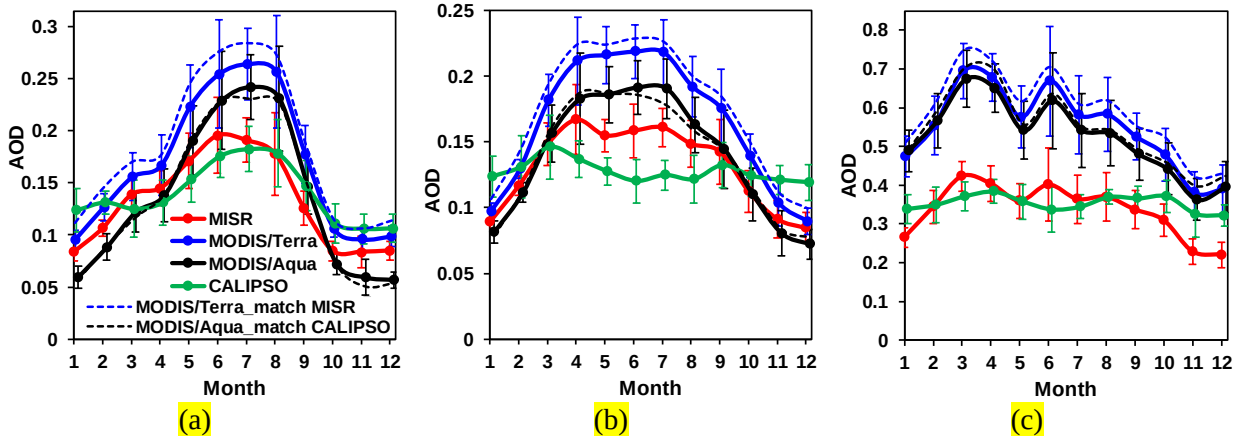


Figure 2. Monthly mean AOD observed by MISR, MODIS/Terra, MODIS/Aqua, and CALIPSO during 2007-2016 in (a) EUS, (b) WEU, and (c) ECC. For CALIPSO, only clear-sky daytime profiles are averaged in order to be consistent with the MISR and MODIS products. "MODIS/Terra_match MISR" is a sensitivity case in 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.

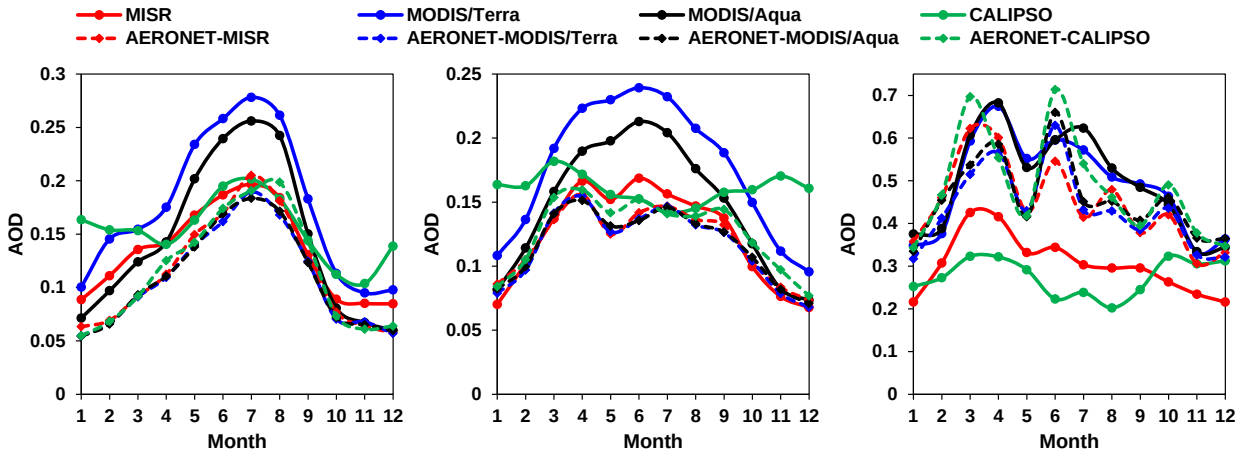
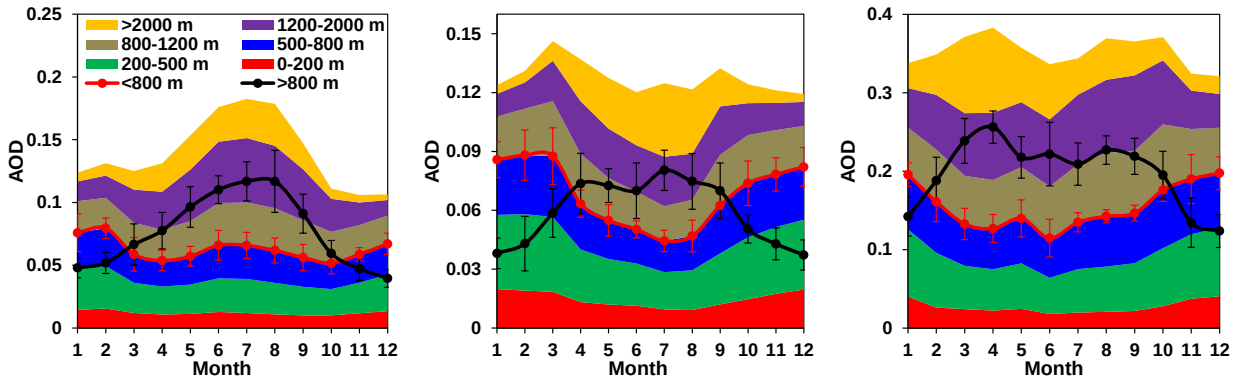
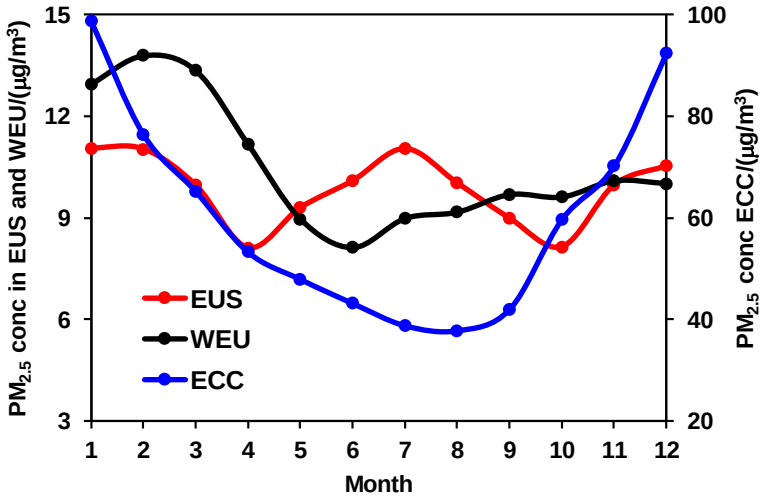


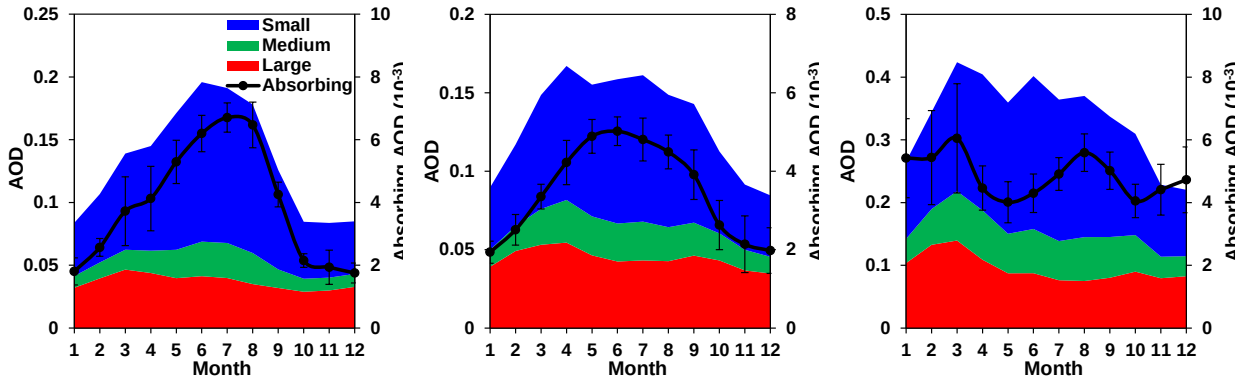
Figure 3. Monthly mean AOD observed by satellites and AERONET averaged across the AERONET sites during 2007-2016 in (a) EUS, (b) WEU, and (c) ECC. The observations from MISR, MODIS/Terra, MODIS/Aqua, and CALIPSO are averaged over 1°×1° grid boxes containing the AERONET sites. The AERONET data are averaged within a 2 h window centered on satellite overpass times. The numbers of AERONET sites included in analysis are 28, 54, and 13, in the EUS, WEU, and ECC regions, respectively. Since the four sensors overpass a site in different days and different times of day, we separately calculate the AERONET data matched to each sensor (denoted by "AERONET-×××"). The AERONET curves matched to different sensors are close in EUS and WEU, partly because there are plenty of sites in these two regions, and the discrepancy due to the sampling issue is therefore smoothed out. In contrast, there are only 13 AERONET sites in ECC, so there exists larger discrepancy between the AERONET data matched to different sensors. Note the different scales on the y-axes of the plots.



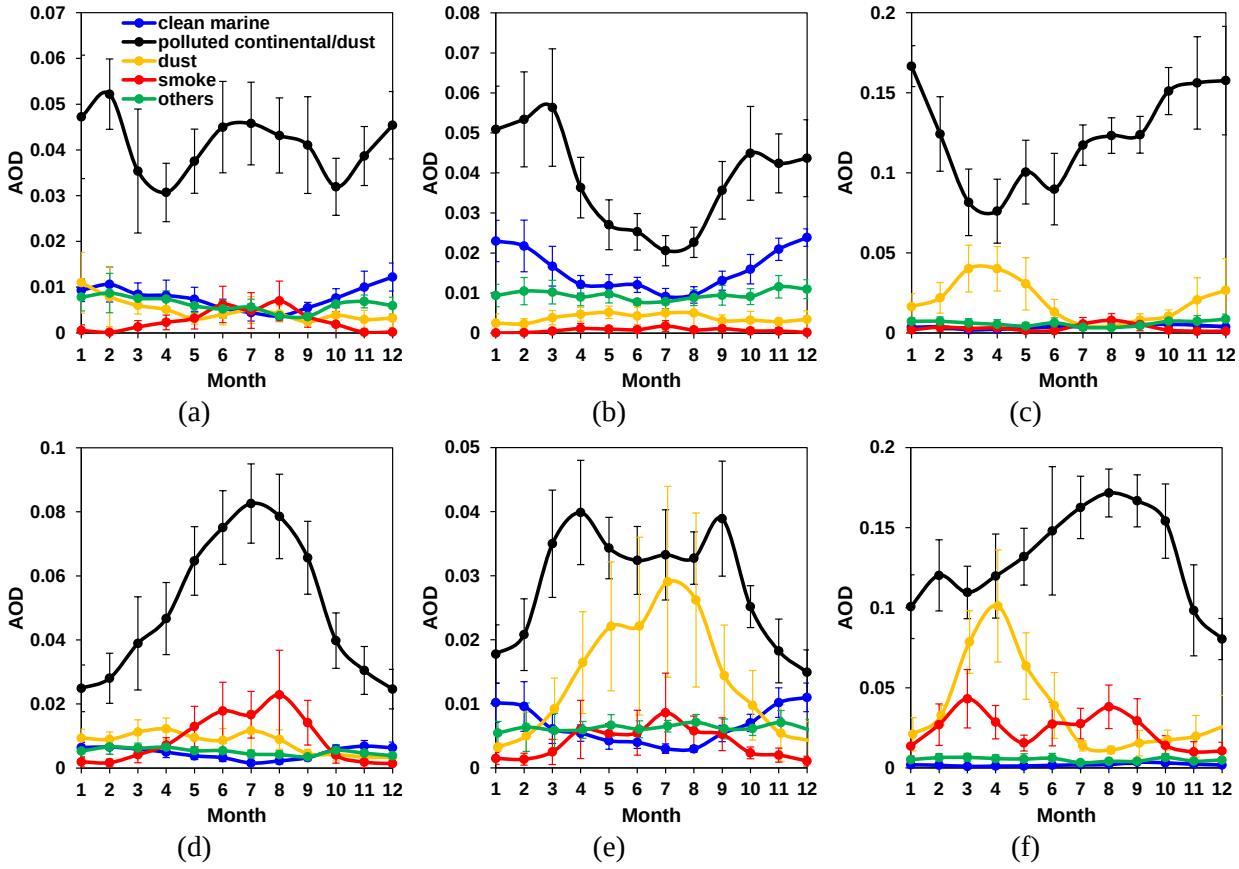
747 **Figure 4**. Monthly mean AOD as a function of height above ground level observed by CALIPSO
748 during 2007-2016 in (a) EUS, (b) WEU, and (c) ECC. Only clear-sky daytime profiles are averaged
749 in order to be consistent with the products of MISR and MODIS. The range of AOD within a
750 particular height range is depicted by the colored stacks. The integrated AODs for heights below
751 and above 800 m are shown as solid lines, for which the error bars are defined in the same way
752 as in Fig. 2. Note the different scales on the y-axes of the plots.



753
754 **Figure 5.** Monthly mean surface $PM_{2.5}$ concentrations during 2007-2016 in three target regions.
755 The numbers of observational sites included in averaging are 225, 52, and 496, in the EUS,
756 WEU, and ECC regions. Note the different scales on the y-axes for EUS/WEU and ECC.



757 **Figure 6**. Monthly mean AOD of different aerosol types observed by MISR during 2007-2016 in
758 (a) EUS, (b) WEU, and (c) ECC. The size-resolved AODs are depicted by the colored stacks (left
759 Y-axis); the integration of the three size ranges yields total column AOD, as represented by the
760 upper edge of the blue color. The AOD of absorbing aerosols is shown as solid lines (right Y-axis),
761 for which the error bars are defined in the same way as in Fig. 2. Note the different scales on the
762 y-axes of the plots.



763 **Figure 7**. Monthly mean AOD of different aerosol types (a-c) below 800 m and (d-f) above 800 m
764 observed by CALIPSO during 2007-2016 in (a, d) EUS, (b, e) WEU, and (c, f) ECC. Only clear-
765 sky daytime profiles are used in the averaging to be consistent with the products of MISR and
766 MODIS. The definition of error bars is the same as in Fig. 2. Note the different scales on the y-
767 axes of the plots.

768 **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

769