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Using MODIS Cloud Regimes to Sort Diagnostic Signals of Aerosol-Cloud-Precipitation Interactions

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Abstract

Coincident multi-year measurements of aerosol, cloud, precipitation and radiation at near-global scales are analyzed to diagnose their apparent relationships as suggestive of interactions previously proposed based on theoretical, observational, and model constructs. Specifically, we examine whether differences in aerosol loading in separate observations go along with consistently different precipitation, cloud properties, and cloud radiative effects. Our analysis uses a cloud regime (CR) framework to dissect and sort the results. The CRs come from the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor and are defined as distinct groups of cloud systems with similar co-variations of cloud top pressure and cloud optical thickness. Aerosol optical depth used as proxy for aerosol loading comes from two sources, MODIS observations, and the MERRA-2 re-analysis, and its variability is defined with respect to local seasonal climatologies. The choice of aerosol dataset impacts our results substantially. We also find that the responses of the marine and continental component of a CR are frequently quite disparate. Overall, CRs dominated by warm clouds tend to exhibit less ambiguous signals, but also have more uncertainty with regard to precipitation changes. Finally, we find weak, but occasionally systematic co-variations of select meteorological indicators and aerosol, which serves as a sober reminder that ascribing changes in cloud and cloud-affected variables solely to aerosol variations is precarious.

1. Introduction

While the number and quality of observations of aerosol, clouds, precipitation, and radiation has been steadily growing and improving in quality, using these observations to unequivocally detect interaction pathways among them has remained a formidable challenge. To distinguish cause and effect, while concurrent changes in the surrounding meteorological environment also take place is a tall order that usually forces us to resort to circumstantial evidence to support our predictions and theoretical constructs. In the end, with only imperfect or compromised observations at hand, we do not have a high degree of confidence about the details of the interactions and their usefulness in a predictability context.

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Investigations of aerosol-cloud-precipitation interactions (ACPI) commonly focus on a few standard paradigms. The first paradigm that springs to mind at most times is the “cloud albedo effect” [Twomey, 1977] or “first indirect effect” (FIE) or “Twomey effect” whereby liquid clouds become more reflective when aerosol number concentration, and therefore the amount of available cloud condensation nuclei (CCN) increases. Clouds become brighter because their optical thickness increases, a consequence of the more numerous but smaller droplets formed under constant liquid water content conditions when more droplets are activated due to ampler CCN availability. When searching for observational signals of the FIE, evidence of either higher cloud albedo (or some closely-related radiative quantity), or increased cloud optical thickness, or decreased cloud effective radius (or a combination of all the above) is sought. “Ship tracks” (narrow streaks of brighter clouds along ship paths, e.g., Radke et al. 1989) are the most renowned manifestations of FIE, although in a strict sense the main requirement of the pure “Twomey effect”, namely that the total cloud condensate remains constant, is not necessarily satisfied [Chen et al. 2012].

Also associated with liquid-phase clouds is the so-called ‘lifetime’ effect, commonly referred to as the “second indirect effect” (SIE), where aerosol effects on precipitation constitute an additional process in the aerosol-cloud interaction cycle, leading thus to what we now call ACPI. In this mechanism, a heavier aerosol load ultimately yields more persistent warm clouds, higher cloud amounts, less precipitation, and higher liquid water contents [Albrecht, 1989]. A simplified description of the physical pathway states that more aerosols, (via greater number of CCNs), more widespread droplet activation, larger droplet concentration, and smaller droplets lead to suppression of warm precipitation development (via reduced collision-coalescence), and a resultant increased tendency for clouds to retain more condensate, be more extensive and long-lived since suppressed precipitation is thought to inhibit their demise [Stevens and Feingold, 2009]. Not as broad a consensus exists for the SIE as there is for the FIE, given that increased droplet concentration does not universally suppress precipitation across all types of warm clouds [Stevens and Seifert, 2008]. If one were to look for observational evidence of systematic SIE occurrences, then positive correlations of aerosol loading with cloud fraction or domain-averaged reflected solar flux, and negative correlations with precipitation would provide support for the hypothesis.

Existing evidence on the nature of ACPI is even less definitive and prone to diverging interpretation for cold (mixed- and ice-phase) clouds. The expected eventual outcomes are highly uncertain as well. In Table 1 of the review paper by Lohmann and Feichter [2005], the ultimate radiative effect of mixed-phase clouds remains unspecified for the three types of aerosol-cloud interactions listed (“thermodynamic effect”, “glaciation indirect effect”, and “riming indirect effect”) while predictions for precipitation changes as a result of these interactions include both increases and decreases. The most recent IPCC AR5 synthesis [Boucher et al. 2013] does not shed more light: a statement is included about anthropogenic (and presumably natural) perturbations to aerosol potentially affecting glaciation, liquid and ice optical properties, and ultimately cloud radiative effects, but the direction of the changes remains unclarified.

For ice clouds, it has been hypothesized that a “Twomey”-like effect with similar outcomes as in liquid clouds takes place [Lohmann and Feichter 2005], namely increases in ice

particle numbers, smaller crystal sizes, larger ice optical thicknesses, and increased brightness (more reflected solar radiation) as well as “blackness” (higher emissivity in the thermal infrared). Boucher et al [2013] highlight two studies that have documented negative correlations between aerosol optical depth (AOD) and ice particle size in deep convective clouds [Sherwood, 2002; Jiang et al., 2008], while another study has found a negative correlation in simulations of anvil clouds [Fan et al. 2013]. Evidence for such behavior in cirrus clouds embedded in weaker dynamical environments remains however lacking: in their modeling study Lee and Penner [2010] find that it is actually increases in ice water path under increased aerosol loading that have the most impact on cirrus cloud optical thickness rather than decreases in effective particle size.

The most intriguing of aerosol effects on clouds is perhaps the so-called “invigoration effect” hypothesized to occur within mixed phase clouds with precipitation potential [Rosenfeld et al. 2008; Altaratz et al. 2014]. Invigoration, as the name suggests, results in an expansion and deepening of clouds by aerosol. This means physically and optically thicker clouds (accompanied by changes in the vertical distribution of condensate), wider convective anvils (hence larger cloud fractions) where applicable, higher cloud tops (lower cloud top pressures) and more intense rainfall. Despite extensive observational and modeling efforts, the widespread occurrence of the invigoration effect, the mechanism and sequence of the interactions, and the potential magnitude of climatic responses still remain uncertain [Tao et al., 2012; Altaratz et al. 2014], with some of the low degree of confidence due to discrepancies between simulations and observations. Also unknown is whether ice particles sizes near cloud tops as observed by passive sensors respond consistently in a specific direction.

As will become clearer in the following, even if all mechanisms involved in ACPI were known and neatly broken down by cloud thermodynamic phase, observational confirmation would not necessarily be straightforward. Setting aside the fact that non-aerosol factors can potentially have big impacts on cloudiness, global or near-global studies have to employ rather coarse datasets which rarely have homogenous single-type clouds within individual grid cells. Furthermore, in passive imager observations cloud phase is determined by conditions near cloud top so that the thermodynamic phase throughout full cloud depth can not be unambiguously determined. Another major impediment is that aerosol observations in cloudy areas decrease both in quantity because fewer confidently clear pixels can be identified (Levy et al. 2013), but also in quality because of clouds affecting the reflectance of nearby clear areas (Várnai and Marshak 2009). Finally, the full lifecycle of clouds under aerosol influence is usually not monitored because of the sparse temporal coverage of some of our most capable space-based observing systems.

Given the success of the Cloud Regime (CR) concept as a meaningful foundation on which to base the breakdown and interpretation of a variety of cloud properties and impacts [Oreopoulos et al., 2014; 2016], we examine here whether its proven usefulness can extend to the study of ACPI. Successful aspects of such an approach have been previously demonstrated by Gryspeerdt et al. [2014a;b]. Regime-specific examinations of cloud and precipitation responses to aerosols have been advocated earlier by Stevens and Feingold [2009] as vital for capturing the expected full diversity of outcomes, but without an explicit

guidance on how a “regime” should be defined. Our choice of CRs as the major modes of cloud height and opacity co-variations provides a means to systematically group cloud systems that appear similar from space and to meaningfully organize the diagnostics of ACPI. While this is in our view a step in the right direction, the nature of the satellite observations from which cloud and aerosol information is retrieved does not permit a more illuminating analysis that would include monitoring of the temporal evolution of clouds as aerosol conditions change. Instead, we resort to comparing similar instantaneous cloud states (i.e., “snapshots” of regimes) in different aerosol conditions, a comparison from which we can only diagnose prevailing tendencies in cloud-affected variables which are not as revealing about the underlying physical processes.

2. Datasets and Analysis Methodology

2.1 Datasets

We use 12 years (December 2002 to November 2014) of a variety of observations within the 50°S–50°N latitude zone. One of the main reasons we restrict our analysis area within that zone is geographical coverage of our precipitation dataset of choice, the TRMM (Tropical Rainfall Measuring Mission) Multi-satellite Precipitation Analysis (TMPA-3B42) dataset [Huffman et al. 2007]. In this dataset precipitation rate is natively resolved at a 0.25° grid and 3-hour intervals; we subsample only the time intervals that contain the overpasses of the Terra and Aqua satellites providing aerosol and cloud information, and subsequently resample rain rate to one-degree resolution to match the aerosol and cloud datasets (see below).

Aerosol loading is expressed in terms of aerosol optical depth (AOD) and comes from two distinct sources: Moderate resolution Imaging Spectroradiometer (MODIS) “Dark Target” (DT) [Levy et al. 2013] Collection 6 Level-3 daily data (10:30 morning values from the Terra satellite and 1:30 afternoon values from the Aqua satellite) at one-degree spatial resolution, which we call “MODIS DT AOD” or simply “MODIS AOD”; and Modern Era Retrospective analysis for Research and Applications version 2 (MERRA-2) analysis AOD [Randles et al. 2017] at $0.625^\circ \times 0.5^\circ$ resolution, interpolated to one degree, and subsampled to the 3-hour value containing either the Terra or the Aqua overpass, which we call “MERRA-2 AOD”. This reanalysis is based on a version of the Goddard Earth Observing System version 5 (GEOS-5) model that is radiatively coupled to Goddard Chemistry Aerosol Radiation and Transport (GOCART) aerosols and includes assimilation of bias corrected AOD from the Advanced Very High Resolution Radiometer (AVHRR, irrelevant for our period of study) over ocean, from MODIS on both Terra and Aqua satellites, from Terra’s Multi-angle Imaging SpectroRadiometer (MISR) instrument over bright surfaces, and from ground-based Aerosol Robotic Network (AERONET) observations.

The MERRA-2 AOD values are climatologically quite different from those derived by the DT algorithm from MODIS radiances. Fig. 1 compares histograms from both data sources. Direct observations suggest that morning (Terra) and afternoon (Aqua) aerosol distributions are quite different, but the extent to which the differences reflect actual aerosol variability is questionable [Levy et al. 2013]. For MERRA-2, on the other hand, AOD values are much more similar between morning and afternoon, and also systematically lower than MODIS.

The root cause of the discrepancy is MERRA-2 assimilating older Collection 5.1 radiances which are then inverted to obtain AOD using a different algorithm, based on Neural Net cloud screening and retrieval [Randles et al. 2017].

When ACPI is studied with genuine aerosol observations, contemporaneous aerosol samples are limited because aerosol data are sought in the presence of clouds. Any compromise to increase the amount of available aerosol data will therefore have some impact. Our strategy to maximize the volume of observed AOD entails the following: First, we accept that morning and afternoon values of AOD are adequately represented by a single daily (daytime) value. We assign such a daily AOD value from Terra when Aqua AOD is unavailable and vice-versa; when both retrievals are available we are taking a simple arithmetic average. We also visit immediate grid cell neighbors when both Terra and Aqua AODs are unavailable in the grid cell of interest; in this case the daily AOD value is defined as the arithmetic average of all available neighboring Terra and Aqua AOD values. This process is shown in the schematic of Fig. 2. Only when neither the grid cell of interest nor any of the immediate neighbors have valid AOD values, is the grid cell discarded from the analysis. This happens for 29.7% of MODIS Level-3 the grid cells. For MERRA-2 data we are only applying the morning-afternoon averaging (top row of graph) since an AOD value is always available.

Cloud properties also come from MODIS Level-3 data at 1° spatial resolution from Terra (MOD08) and Aqua (MYD08) datasets. We use the following cloud properties: Cloud fraction of Successful Retrievals (CF, total and by phase: liquid, ice), Cloud Optical Thickness (COT, total), Cloud Effective Radius (CER, by phase: liquid, ice), and Cloud Top Pressure (CTP, no phase discrimination). Most importantly, MODIS cloud data are used to define Cloud Regimes (CRs). Specifically, we use International Satellite Cloud Climatology Project (ISCCP)-like joint histograms of CTP and COT. The joint histograms from both Terra and Aqua are treated as a single ensemble subjected to *k-means* clustering analysis [Rossow et al. 2005] to define 12 global MODIS CRs. Details can be found in Oreopoulos et al. [2016], hereafter O16, with the regime centroids (mean of alike histograms) and their frequency of occurrence maps displayed in Figs. 3 and 4. Descriptions of the regimes and a summary of the process used to derive them can be found in O16. Their average properties within the 50°S - 50°N area of study are summarized in Table 1 which also includes a short description of each CR. It is important to clarify that even though we restrict ourselves here to the 50°S - 50°N zone, the CRs are not specifically derived from the joint histograms encountered only in that zone (this is why Figs. 3 and 4 are identical to those in O16) ; rather, we only use the subset of the O16 dataset within our area of interest. Furthermore, we draw attention to Fig. 4 of O16 showing the mean cloud fraction by water phase of each CR. This figure is subsequently used as a guide to form groups of CRs based on their dominant phase.

In addition to cloud and precipitation data, we also use Clouds and the Earth's Radiant Energy System (CERES) SYN1deg-daily data at 1° resolution to examine the overall apparent radiative signatures on clouds of aerosol changes. The variable we composite by CR is the Top-of-the-Atmosphere (TOA) Cloud Radiative Effect (CRE), namely the flux difference between all-sky and cloudless-sky conditions. We examine CRE changes for both

the solar/shortwave (SW) and thermal infrared/longwave (LW) part of the spectrum. Since for a specific grid cell the Terra and Aqua CRs on a particular day can differ, the same daily CRE value may be assigned to two distinct CRs.

Finally, we also investigate whether the meteorological environment and AOD exhibit signs of coupling. It is prudent to at least attempt to do so since it may reveal whether meteorological variability may be partly responsible for systematic apparent dependences of cloud-affected quantities on aerosol. We consider three meteorological variables thought to be closely connected to cloud variations and structure: vertical pressure velocity, air temperature, and relative humidity. Full profiles for these three variables are obtained from the MERRA-2 re-analysis dataset [Bosilovich et al. 2015] and are spatiotemporally matched with the Terra and Aqua CR occurrences by appropriate interpolation and sampling.

2.2 Analysis Methodology

Cloud, precipitation, radiative, and meteorological variability are examined in terms of relative aerosol variations, similar to Gryspeerdt et al. [2014a]. This is a critical choice of our analysis because when contrasting values for cloud-affected quantities at “low” and “high” values of AOD, what constitutes low and high aerosol loading is specific to the grid cell and season (and eventually to the CR as well, since all results are composited by CR). Had we used absolute AOD values, meteorology would have been less constrained since absolute AOD values confined within certain ranges may have systematically originated from distant and therefore meteorologically distinct locations (despite CR compositing provides a degree of meteorological constraint). As Fig. 4 clearly shows, CR occurrences do not necessarily aggregate in regions of geographical proximity. Despite our choice to emphasize relative AOD changes, we recognize that absolute AOD values may still matter. For instance, small aerosol changes in an absolute sense may limit cloud and precipitation responses; on the flipside, large absolute changes in AOD occurring at the upper range of values may also be of limited efficacy in triggering substantial cloud and precipitation responses because of saturation effects (this is analogous to different behaviors of cloud albedo susceptibility under absolute versus relative droplet density perturbations as explained by Platnick and Oreopoulos 2008).

To create relative AOD variations we construct multi-annual seasonal histograms of daily AOD for each grid cell using 5% percentile increments (i.e., 20-quantiles or vigintiles). For all grid cells that contain a specific CR member, we average (composite) all precipitation, cloud, and radiative quantities that coincide with AOD values that fall within the same seasonal vigintile. The end result is variability curves for each CR that represent how cloud or cloud-affected properties vary vs AOD vigintile. This compositing is always performed separately for land and ocean grid cells. The MODIS-based and MERRA-based curves will in general be dissimilar because, besides the overall sampling discrepancy (MODIS has missing values while MERRA-2 has none), the differences between the two datasets depend on location, so even for the same CR the ensemble of grid cells belonging to a certain AOD vigintile will be different. Since we have 12 CRs, many of the figures that follow consist of two sets of 12 panels, one for land and one for ocean. As will become evident shortly, such a

separation proves indispensable for capturing the full diversity of responses to AOD changes.

3. Precipitation variability vs AOD

Because of the aforementioned availability of two sets of AOD values, one from MODIS DT and one from MERRA-2, the investigation of aerosol-precipitation links, and actually all analysis that follows, is conducted along two distinct paths. The comparison of the two classes of results is actually of great value for assessing the reliability of the apparent interaction signatures. Let us state upfront that each aerosol dataset has its own shortcomings and imperfections. As mentioned in the introduction, observed MODIS AOD suffers from reduced sampling and greater than usual retrieval uncertainties because our analysis requires aerosol in the vicinity of clouds. The re-analysis AOD has perfect sampling, but suffers from other drawbacks, such as the assimilation drawing toward the forecast from the GEOS-5/GOCART model in the absence of observations to assimilate, changes with time in the observing systems providing the assimilation data, and forecasts being influenced by prescribed aerosol emissions inventories [Randles et al. 2016]. In the end, we elected to keep MODIS DT as the default aerosol dataset to be used for the figures in the main body of the paper, and delegated all figures based on MERRA-2 aerosols to the supplementary material. The results contained in the latter figures are nevertheless still discussed and compared with their MODIS-based AOD counterparts. Our decision to split the figures is guided therefore by both practical considerations and a preference to pay greater attention to what is possible deduced solely from observations, the more traditional route followed in most previous studies.

In Figure 5, TMPA precipitation rate (PR, mm/h), excluding zero precipitation grid cells, has been composited as a function of relative aerosol burden, namely AOD percentile (5% increments) from distributions built in each 1° grid cell, per season. The counterpart results based on MERRA-2 AOD are shown in Fig. S1. These, and the other figures of this type that follow, display averages coming from grid cells that have a particular CR (Terra or Aqua) occurrence, along with daily AOD values (per procedure depicted in Fig. 2) that belong to the same vigintile of the AOD distribution (but with substantially different absolute AOD values possible). In other words, we create composites/averages of the variable of interest (in this case non-zero precipitation) from all grid cells where the same CR and AOD vigintile occurs. Land and ocean grid cells are considered separately in this procedure, as stated earlier.

The color symbols in Fig. 5 and figures that follow indicate where in the distribution particular AOD values, from 0.1 to 0.5 in increments of 0.1, occur (their position is different for each CR and depend on the sample used to create the composite curve). For example, in the CR1 ocean panel of Fig. 5, AOD=0.2 corresponds to the 55% percentile and AOD=0.3 to the 90% percentile. In this representation, even if the detailed nature of the dependence on absolute AOD is often not seen, the direction of change can nevertheless be discerned.

When Fig. 5 is compared to Fig. S1 we see that the same relative burden means different absolute AOD values for MERRA-2 and MODIS (MERRA-2 absolute AOD is smaller).

Closely spaced (horizontally) symbols indicate small samples within the corresponding AOD range. The statistical significance of each curve is judged with respect to the range of values represented by the light gray shade. This gray “envelope” was generated as follows: For each AOD percentile, we selected randomly a set of precipitation values equal in number to that of the proper set of precipitation values corresponding to that vigintile. We made one hundred such random trials to emulate a “bootstrap”-type approach. The envelope was then defined by the range (minimum to maximum) of means calculated from the values within these randomly selected sets. With this empirical confidence interval measure in hand, we can essentially compare the curve derived from proper compositing (that accounts for coincidence of precipitation with specific relative AOD amounts) to the range of possible curves that would result if precipitation was composited using an identical number of samples, but from random instead of proper selection. Essentially, the gray envelope represents therefore the natural variability of precipitation values that are uncorrelated with aerosol. Note that if the composite curve is flat (little variability with AOD), it inevitably tracks very closely the overall mean value of non-zero precipitation for that CR (dashed line), and the gray envelope by construction is also narrow. Such narrow envelopes are actually the norm for many panels of this kind that follow later, an indication that data sampling for these variables is sufficient for robust means at the AOD vigintile level, and therefore yields statistically significant results. But in this particular compositing attempt, the requirement of non-zero precipitation makes the sampling inferior, and gray envelopes of considerable width can be easily discerned in Figs. 5 and S1. When sampling is especially poor, such as for CRs that have small presence over land, the width of the envelope becomes more pronounced, as for CR7 and CR9 in Figs. 5 and S1.

For the most part, in these and many other plots to follow, the curves derived from proper compositing are usually rather smooth with orderly and systematic variations; exceptions that manifest as noisy curves can be seen when sampling is inadequate, i.e., noisy curves and broad gray envelopes typically go together. While not necessarily proving causality, smooth composite curves accompanied by a range of variations outside the bounds of natural variability (gray envelope) are indicative of statistically robust relationships.

With the above in mind, let us examine Figs. 5 and S1 more closely. These figures mean to capture aerosol effects on precipitation in cloud systems that are already precipitating. Except for CR12 (omnipresent small cloud fraction regime that includes most low altitude fair weather clouds, but also non-extensive high clouds), and perhaps CR6 (high frequency of mid-level clouds and perhaps some congestus clouds) there do not seem to be unambiguous signals (i.e., range of variations substantially outside the range defined by the grey envelope) of enhanced precipitation rate due to increased aerosol loading over land. By the same token, there are no signs of precipitation suppression either. These conclusions are not affected by the choice of AOD dataset. However, the picture is somewhat different over ocean, where, incidentally, one can see clearly the smaller absolute values of AOD compared to land, and the smaller values of MERRA-2 compared to MODIS. First, CR12 exhibits again rather clear signs of invigorated precipitation, and while the shape of the curve looks quite different between MODIS and MERRA-2, this is partly because of the choice of relative AOD as a measure of aerosol loading; for both datasets the upward trend starts at similar values of absolute AOD (~ 0.1). Moving on to other CRs, the difference in the shape

of the CR1 (a mostly tropical CR containing a lot of the cirrus associated with convection, and deeper clouds as well) precipitation composite curve between MODIS and MERRA-2 is quite pronounced and cannot be explained by the different range of absolute AOD values: the change in precipitation above 0.2 AOD is much larger and more abrupt for MERRA-2 despite the absence of high absolute AOD values. CR2-CR5 (all regimes that contain storm activity, with CR2 and CR3 being more of the convective variety, while CR4 and CR5 being more closely linked to extratropical storm tracks) in Fig. 5 indicate a suppression of precipitation by aerosol (in contrast to e.g., Koren et al., 2012), but MERRA-2 AOD (Fig. S1) supports this only for CR3. To what extent this behavior, as well as that reflected in the left part of “U”-shape curves is indicative of wet scavenging effects which would make the heaviest precipitation (in a relative sense) be associated with the lowest AOD and vice-versa, is unknown (see Gryspeerd et al. 2015 for an attempt to tackle this). Regimes CR8-CR11 (regimes with large proportions of boundary layer stratus, stratocumulus, and cumulus) produce rather weak precipitation and therefore also weak precipitation change vs AOD. Moreover, over land the sampling and therefore the statistical significance of the curves is poor. Precipitation associated with CR7 (more stratus than the other boundary layer CRs) is somewhat stronger, but variability remains flat, and sampling over land is inadequate. In the end, with the dataset at hand, no evidence of precipitation suppression (SIE) can be seen CRs where low clouds are prevalent. One should keep in mind, however, that for the types of clouds contained in these regimes, no clear threshold distinguishing precipitating from non-precipitating scenes [Stephens and Kummerow 2007] can be set, so that changes in liquid water path (increases are consistent with SIE) can disguise as precipitation changes in the TMPA dataset.

Figures 5 and S1 were an attempt, with the admitted shortcoming of all “snapshot”-based analyses not being able to distinguish aerosol effects on precipitation vs precipitation effects on aerosol, to find evidence of aerosol effects on precipitation already occurring. But it is probably as important to also try to determine whether the likelihood of precipitation occurring also depends on aerosol. To this end, Figure 6 shows the probability of precipitation (POP) as a function of relative MODIS DT AOD, with the counterpart MERRA-2 figure being S2. In these figures we do not devote a separate panel for each curve, but rather collect all curves in two separate panels for ocean and land. The curves were constructed by calculating the ratio of the number of grid cells that are precipitating to the total numbers of grid cells within each AOD vigintile and for each CR. The ratio is expressed as a percentage.

The first thing we notice is that POP is greater over ocean than over land. For land, CR7-CR12 have POP < 10% across the range of AOD for both MODIS and MERRA-2. This puts in context the precipitation variability vs AOD for CR12 in Figs. 5 and S1. That unambiguous signal applies to only a small fraction of CR occurrences. However, because CR12 has by far the largest Relative Frequency of Occurrence (RFO) of all CRs, its precipitation variability and POP still matters a lot. The MODIS AOD dataset indicates some gentle tendency of its POP to increase with relative AOD over land, but the corresponding curve for MERRA-2 is more flat. Oceanic CR12s on the other hand, exhibit a clearer increase of POP with relative AOD. Regardless of the AOD dataset, in general no increase in POP with AOD is seen over land, in contrast with the ocean case where several CRs increase

their POP with AOD. Aside from the aforementioned CR12, we see CR1, CR6, CR7, and CR8 (recall the weak mean precipitation of the latter, however) exhibiting such behavior, with a ~5-10% increase from minimum to maximum POP values in the MODIS AOD plots (Fig. 6). For MERRA-2 (Fig. S2), CR1 no longer belongs to the group of CRs with POP increasing in tandem with AOD, joining almost all boundary layer cloud CRs with weak mean precipitation. Generally, the MERRA-2 curves in Fig. S2 illustrate greater suppression of POP and non-monotonic behavior for the CRs that exhibit a rather flat POP for MODIS AOD. But for the other CRs (i.e., oceanic low-level cloud CRs) neither dataset detects precipitation signals consistent with SIE.

When considered collectively, the results of Figs 5, 6, S1 and S2 do not reveal signs of invigoration if concomitant AOD and rainfall increases is used as the sole criterion (as e.g. in Koren et al. 2012). Such invigoration would be expected in CR1-CR5 which are either mixed-phase or ice-dominated regimes. Instead, the MODIS AOD dataset indicates suppression of rainfall for CR2-CR5 (except for CR3, a less clear picture emerges from MERRA-2). A clear tendency for increased rainfall with AOD was found in our only other regime that is a candidate for invigoration, CR12. In view of the fact that the POP of CR12 also increases with AOD, invigoration can indeed be occurring in these small CF cloud systems.

We recognize that our analysis method suffers somewhat from the asymmetrical nature of our evidence: while rainfall rate increases with AOD support invigoration, rainfall increases towards the other direction (lower AOD) do not necessarily negate invigoration because we may be simply observing the stage of the storm's lifecycle where stronger rainfall accelerates the wet removal of aerosol (or, on the flipside, weaker rainfall allowing aerosol to accumulate prior to invigoration), i.e., the situation where rainfall is the cause and aerosol AOD is the effect. Wet scavenging likely appears more pronounced in the MERRA-2 analysis because AOD values exist even under the heaviest precipitation, their low values reflecting the advection of depleted aerosol amounts from locations where the storm has already passed.

Along with invigoration signs not materializing, precipitation suppression is also not detected in other cloud systems where such a mechanism would be expected, namely oceanic CRs dominated by boundary layer warm clouds. While the precipitation produced by most of these CRs is weak, we do not see a decrease in their capacity to precipitate as AOD increases. On the contrary, over the ocean we see a small increase in likelihood that they will precipitate. While the precipitation curves for these CRs do not support interpretations that involve wet scavenging, the results should nevertheless be viewed with caution given TMPA's lower skill at the low end of rainfall intensity [Huffman et al. 2007], and the possibility that high concentrations of suspended small hydrometeors (i.e., cloud droplets) are not distinguishable in those CRs from larger, but sparse, falling hydrometeors (i.e., light rain and drizzle).

4. Cloud Property variability vs AOD

We now examine apparent cloud property changes with relative AOD. Specifically, we examine changes in CF, COT, CER and CTP. The presentation is very similar with that for precipitation above. The uncertainty envelope is always included in the plots even if in many cases the natural variability (per our estimation method) is much narrower, because of superior sampling compared to precipitation, and generally cannot be discerned. We should also note that because a CR's joint histograms members have mean values of CF, COT and CTP that by design cannot be greatly different, the variations of these cloud properties with AOD are intrinsically constrained to a certain degree.

4.1. Cloud fraction

We start with CF composites, shown in Fig. 7 for MODIS AOD and Fig. S3 for MERRA-2 AOD. Each panel shows three curves, one for the mean of total CF (black), and one for each of liquid (blue) ice (cyan) CF ensemble means (the phase-specific means include zero values) within AOD percentiles. We also use two distinct CF ordinates, the left corresponding to the total CF and sometimes to one of the phase-specific CFs, and the right corresponding to either liquid or ice CF, or sometimes both, as indicated by the colored arrow(s). The uncertainty "envelope" is very narrow in almost all cases and essentially closely tracks the overall mean (dashed line) across AOD percentiles.

One can immediately see that there are virtually no negative trends, i.e., decreases of total CF as relative MODIS AOD increases (Fig. 7), and this is true for both ocean and land. CF either increases (CR1, CR8, oceanic CR9, CR10-CR12) or remains relatively flat (remaining CRs, although it must be noted that for the virtually overcast CR2 there is no room for neither increases nor decreases). Positive correlations between CF and AOD have been discussed before, and have been given interpretations other than aerosols producing CF increases via the lifetime "Albrecht" (SIE) or the invigoration effect (for a review see Gryspeerdt et al. 2016). One of the interpretations is that as more aerosol retrievals are performed closer to clouds in progressively higher CF situations, the retrieved AOD is biased increasingly higher because of more occurrences of enhanced clear-sky reflectance near cloud edges due to 3D effects [Várnai and Marshak 2009]. Two other possibilities that have received attention are greater aerosol swelling near clouds because of higher humidities [Twohy et al. 2009] and pockets of thicker aerosol layers due to convergence (Mauger and Norris 2007). All the above can certainly bias MODIS DT AOD retrievals, but such imprints certainly do not appear universally in our results. Quite the contrary, the multitude of flat CF composite curves in our results is a notable deviation from the persistent positive CF-AOD correlation of prior works. What may play a role in such a rather flat behavior is the fact that our analysis is performed in terms of relative AOD variations which makes it less likely for systematic biases affecting both the low and high end of the AOD distribution to emerge.

The general absence of negative trends also applies for MERRA-2 (Fig. S3). The curves of composite total mean CF are in general more flat, but increases of CF vs relative AOD can still be seen for CR10-12 for both land and ocean. CR1 over land stands out as the only CR with an undisputable decrease of CF with AOD, in direct contradiction to the counterpart results for MODIS AOD which shows CR1's total CF to increase with AOD. This decrease

is somewhat inconsistent with the rather flat precipitation of Fig. S1, but consistent with the decrease in CR1's POP over land (Fig. S2). In this ice-dominated regime, the smaller liquid CF remains then consistently invariant, but the much larger ice CF exhibits opposite behavior between the two datasets. Comparing the two curves in terms of absolute AOD does nothing to resolve the discrepancy: for MODIS AOD the lowest AOD (0.1) is encountered below the CF mean and the highest AOD (0.5) above, while the situation is reversed for MERRA-2 AOD.

Phase-dependent CF behavior merits closer examination for the mixed-phase regimes CR4, CR5 and CR12. For CR12, the two aerosol datasets yield remarkably consistent results, for both land and ocean, with different rates of increase for the two phases over ocean and similar rates over land. CR4 and CR5 behave distinctly for the two datasets and no reconciliation of results is possible. When MERRA-2 AOD is used, there is much more self-consistency, with ice CF increasing and liquid CF decreasing vs AOD for both CRs, and for both land and ocean. This by itself would suggest phase conversion from liquid to ice as AOD increases and the cloud possibly enters a different phase of its development. On the other hand, when MODIS AOD is used, while marine CR4 and CR5 agree that ice CF decreases and liquid CF increases with AOD, this behavior is opposite of what is seen for the continents within the same dataset, and when MERRA-2 AOD is used. The bottom line is that the oceanic CR4 and CR5 phase-specific CF trends for MODIS AOD (negative for ice CF, positive for liquid CF) are opposite of all other phase-specific CF trends for these two regimes. Despite the rather strong and sometimes contradictory trends of individual phase CFs, the total CF astonishingly exhibits virtually no trend vs relative AOD.

In summary, our results suggest that apparent signatures of cloud “lifetime” effects indeed exist for some regimes dominated by boundary layer clouds such as CR10 and CR11, and to a lesser extent CR8, but these signatures, while still present, are somewhat weaker in the analysis based on MERRA-2 aerosols. The differences between the two datasets may be an indication of AOD retrieval biases in the observations as CF increases, as explored in the literature cited earlier. Increases in CF as signatures of invigoration are harder to discern, and are perhaps most evident in CR12 which is a mixed-phase regime with some vertically developed clouds, albeit of low CF, and not fitting the prevalent picture of a strong storm system being invigorated. But invigoration is actually not necessarily exclusive to such systems [Koren et al. 2014]. CF increases in CR12 are consistent with rainfall and POP increases discussed previously. CR1 exhibits a positive trend of CF with AOD for the MODIS analysis, but since this regime is ice-dominated with a lot of cirrus and an analogous behavior is not seen for MERRA-2 AOD, it would be questionable to consider such a trend evidence of invigoration. The more convective CR2 and CR3 would probably be more appropriate regimes to search for invigoration signatures, but both have flat CFs across the AOD range for both datasets that go together with no increases in precipitation or POP (Figs. 5 and S1; Figs 6 and S2). With regard to CR2, however, one should keep in mind that it is the cloudiest regime with members that are by construction very close to overcast, allowing thus little room for CF variability. The search for invigoration should therefore involve a different signature.

4.2. Cloud Optical Thickness

We now turn our attention to COT. We are showing composites of the total COT, i.e., the combined COT of both phases. The immediate impression is that COT variability curves exhibit more monotonic behavior over land than over ocean, especially for the MODIS AOD analysis (Fig. 8). Actually, for this dataset all land COT curves show a consistent and significant (i.e., changes from minimum AOD to maximum AOD residing outside the uncertainty envelope) increase of COT with AOD for all CRs, something not seen when MERRA-2 AODs are used (Fig. S4), with CR1 being the most striking exception. We previously saw no such consistent increases for CF, but it is still conceivable that the purely observational analysis comprises biases in land AOD retrievals when optically thicker clouds occur in the vicinity. While this is certainly a possibility, the fact that no such systematic effect appears over the oceans alleviates somewhat the concern. In the oceanic MODIS case there is actually an assortment of clear significant decreases (CR2, CR4, CR5) of COT with AOD, quite large increases (CR12), but also more complicated shapes (like CR6). When marine cloud COT variability is examined in conjunction with MERRA-2 AOD (Fig. S4), increases of COT prevail slightly over situations where curves are relatively flat (CR5, CR10, CR11), decreasing (CR3), or conspicuously non-monotonic (CR12).

Trying to make sense of the results of Figs. 8 and S4 is a reminder of the complexity of aerosol-cloud interactions, and the fact that they are neither conveniently classifiable according to the major ACPI paradigms, nor easily detectable in apparent relationships emerging in coincident data. Land-ocean, inter-regime, and AOD dataset dependences abound. Are COT increases accompanied by CER decreases (see next subsection) for the temperate (higher index) CRs evidence of FIE/SIE, while those for stormier regimes (lower indices) evidence of invigoration? Do decreases of COT with AOD signal aerosol radiative effects on the environment that favor optically thinner clouds (what has previously been called the “semi-direct” effect)? Admittedly, our analysis is unable to definitively support or rule out any scenario.

MERRA-2 AOD analysis for oceanic CRs with many low clouds (CR7 to CR12), but also CR6, is generally supportive of FIE/SIE when COT changes are solely used as criterion; alas, there is less support when the underlying aerosol dataset comes from MODIS. That dataset on the other hand, supports FIE/SIE for the same regimes over land more than over ocean. It is also instructive to juxtapose the COT figures with their total CF counterparts of the previous subsection which were often flat for low index CRs. One should perhaps look for increases in both CF and COT to make a case for invigoration; and see simultaneous increases of both CF and COT with AOD in warm CRs to have increased confidence that FIE and SIE occur hand-in-hand (as has been documented in Goren and Rosenfeld 2014). CR1, the regime with most ice and cirrus is an interesting case over land: the two AOD datasets disagree about the direction of CF change and they also disagree about the direction of COT change. But the COT and CF change within each dataset is in the same direction, indicating in one case (MODIS AOD) invigoration and in the other case (MERRA-2 AOD) suppression of the cloud system. Coherent increases of marine CR9 CF and COT support FIE/SIE, in the MODIS dataset, but something analogous is not seen for CR11; for MERRA-2 there are no consistent increases for neither of these two regimes.

4.3. Cloud Effective Radius

Other than the broadly accepted droplet effective radius reduction in warm clouds under conditions of increased aerosol loading according to the FIE, there is no consensus on how particle sizes are affected by aerosols in mixed and ice phase clouds [Massie et al. 2007]. With our regime-based analysis we can offer the most comprehensive picture of apparent particle size dependence on aerosol. This dependence is summarized in Figs. 9 (MODIS AOD) and S5 (MERRA-2 AOD). For the three CRs considered to have a greater parity between the two water phases (CR4, CR5, CR12) we are showing both liquid and ice CER variability; for the other CRs we only show the effective radius of the dominant phase.

The well-established land-ocean contrast in mean CER (larger over ocean, smaller over land), is the first prominent feature that immediately springs out. Our results indicate that systematic CER increases with relative AOD are very rare and occur only for the ice particles of CR5 (more clearly over land, regardless of the aerosol dataset). In all other cases CER either decreases or remains relatively flat. When MERRA-2 AOD is used (Fig. S5) one can see a clear reduction of CER for the three ice-dominated regimes CR1-3 for both ocean and land; for the MODIS AOD case (Fig. 9) the negative trend is more clear over land. For the regimes that are mostly liquid, CR6-CR11, the plentiful examples of what appear as manifestations of FIE on CER (with the most consistent across the board being CR9), are intertwined with flat or near-flat curves indicating no or little average change of droplet sizes (for example oceanic CR8 and CR11, for both aerosol datasets).

Taken as a whole, our results suggest observational support for a negative correlation between aerosol loading and particle size as predicted by the mechanisms described by FIE theory. While this negative correlation has always been mostly discussed within the realm of warm clouds, we find such a correlation to also often exist for ice clouds, something not as well established or accepted, with only a few studies [Sherwood et al. 2002; Jiang et al. 2009] making the case for the existence of systematic effects.

4.4. Cloud Top Pressure

CTP (not partitioned by water phase in the MODIS dataset) is another cloud variable intrinsically constrained by the CR breakdown of our analysis; namely, variations of CTP with AOD are limited by the fact that members of the same CR by definition will be restricted to within a certain range of CTPs. Despite this, clear significant variations of CTP can be seen (Figs. 10 and S6). Note that we have reversed the ordinate so that pressure decreases are shown as curves with upward trends to indicate either cloud deepening or clouds ascending towards higher altitudes.

When MODIS AOD is used (Fig. 10) we see a universal decrease in CTP, i.e., an increase in cloud top height (CTH) over oceans. This is a remarkable result because our CRs seem to all respond alike despite their diversity. Gryspeerdt et al. [2014c] attributed a large portion of this positive correlation between AOD and CTH (negative correlation between AOD and CTP or cloud top temperature) also seen in prior work [e.g., Koren et al. 2010; Niu and Li 2012] to the combined effects of positive correlations between AOD-CF and CTH-CF. But this cannot be true in our case because our analysis, which, once again, is based on relative

AOD changes, finds no consistent positive AOD-CF correlations. Yet, this universal CTP decrease with MODIS AOD does not repeat for marine CRs when MERRA-2 AOD is used in the analysis (Fig. S6). Rather, decreases of CTH (increases in CTP) are seen for large portions of the AOD range for CR1-6, an antithesis to expectations which may be (as discussed earlier) due to more potent wet scavenging by clouds of high CTH precipitating more vigorously. For CR7-CR12, however, which are subject to weaker dynamical forcing (O16), CTH increases (CTP decreases) remain.

Mean CTPs stay lower (higher clouds) overall within each CR when we move to land. For MERRA-2 AOD (Fig. S6) the CTP variations over land resemble their oceanic counterparts (keep in mind, however, that absolute AOD values are larger over land). But for MODIS AOD, land and ocean look quite different, with many near-flat curves (CR2-6; CR9) for land, and CTP decreases when occurring (higher or deeper clouds) being gentle compared to ocean.

Collectively our results show a pretty coherent picture of CTP decrease for regimes CR7-CR12, consistent with e.g., Yuan et al. [2011]. The interpretation of this as deepening is consistent with COT increases we see for the same CRs, namely, increases of cloud geometrical thickness with AOD appear to contribute to both COT increases and CTP decreases (as in Li et al. 2011). One would also interpret the CTP decrease as evidence of invigoration in regimes CR1-CR5 if only the evidence had extended beyond the oceans in the MODIS AOD analysis and was consistent with ice (surrogate for anvils) CF increases. But such consistency is not seen and the tendency for CTP to decrease does no longer exist for MERRA-2 AOD over land. We have therefore not found indisputable evidence for omnipresent invigoration, in accordance with our inconclusive findings when other variables (rainfall, CF, COT) were previously examined.

5. Cloud Radiative Impact vs AOD

Given the apparent cloud property dependence on relative AOD amount shown above, one feels compelled to examine how the observed cloud property changes ultimately affect the radiative strength of clouds. As a measure of the latter we use CRE, the difference in radiative flux between all-sky and cloudless sky conditions. Since we focus here only on TOA, we can define CRE in terms of the upward fluxes, so that LW CRE is usually positive and SW CRE negative when the all-sky flux is subtracted from the cloudless sky flux. Both flux quantities needed to calculate CRE are available from the CERES SYN1deg-daily product which provides gridded values at 1° spatial resolution. Compositing is performed as with all other variables, but we note again that due to the daily-averaged nature of the product, two different CRs (from Terra and Aqua) can be assigned the same CRE. Also important to point out is that when examining CRE variations vs AOD, not only can the cloudy (all-sky) flux change because of the effects of aerosol on clouds (indirect aerosol effect), but so can also the cloudless sky flux because aerosol alters the radiative properties of clear skies (direct aerosol effect). This latter effect is far more important in the SW than the LW. The change in cloudless sky flux tends to suppress any (absolute) increases and enhance any absolute decreases (although as we will see shortly, the latter is rarer) in negative SW CRE. In general SW and LW CRE changes with aerosol are related to changes

in CF, COT, CTP, and (far less) CER, previously shown, but because changes in these cloud variables can produce competing radiative effects, it is difficult to predict the eventual CRE outcomes. Both SW and LW CREs depend linearly on CF (with all other cloud variables constant), which makes CF likely the biggest driver of CRE changes [e.g., Goren and Rosenfeld 2014]; COT and CER changes (which may not be independent) affect mostly SW CRE, while CTP changes affect mostly LW CRE.

Fig. 11 shows SW CRE vs relative MODIS AOD for each CR and with our standard ocean and land breakdown as in all previous figures of this type. There is an unequivocal across the board increase in SW CRE (negative values increase in an absolute sense) with relative AOD for all CRs. The increases are most dramatic over ocean which provides a dark surface background that makes all cloud changes radiatively more impactful. The increases are consistent with the general positive tendency of total CF shown in Fig. 7, but occur also for CRs with flat CF curves, e.g., CR2 and CR7. Any notable CER effect on SW CRE would come through COT changes which are more complex over ocean (Fig. 8) than the SW CRE changes themselves; the land COT increases of Fig. 8, on the other hand, would inevitably, all else being equal, yield the SW CRE changes seen in the land panels of Fig. 11. The results based on MERRA-2 AOD (Fig. S7) resemble those from MODIS only for the high index CRs, CR7-12, i.e., stronger SW CRE with relative AOD increase, also associated with CF and COT increases. For CR2, trends of weakening or flat SW CRE are associated with increasing COT (e.g., CR2) which is puzzling since the total CF curves are inescapably flat (Fig. S3); for CR1, the COT and SW CRE curves are much more consistent. It is possible that the less rigorous compositing we are forced to apply for SW CRE (the same 24-hour value assigned to both the morning and afternoon CR) may be a factor. Still, the exact same type of compositing does not seem to create such issues for the MODIS AOD case, so we suspect that the MERRA-2 AOD vigintiles may have systematic geographical dependences which affect the incoming solar flux.

LW CRE also becomes generally stronger with relative AOD which by itself can be considered evidence of invigoration for our low index CRs whereby more ice clouds with lower CTP and colder top temperatures form under heavier aerosol loading, but has less dramatic increases, as expected, for the CRs dominated by low cloud tops (CR7-CR11) and also for land CR occurrences overall (Fig. 12). Note that, as with SW CRE variability, LW CRE changes are almost always outside the bounds of natural variability defined by the gray envelopes. The less notable LW CRE increases over land are consistent with CTP changes that are also more modest than over ocean (Fig. 10). Keeping in mind that CTP is not the only variable affecting LW CRE, we see slight LW CRE increases associated with flat or declining CTHs (e.g., CR2, CR5), but also near-flat LW CRE curves corresponding to increasing CTHs (CR10). For CR1-3, the CRs that matter the most for LW CRE, the counterpart analysis using MERRA-2 AODs (Fig. S8) shows LW CRE weakening with increasing relative AOD for a large part of the AOD range. While this does not agree with the MODIS AOD results, it also comes as no surprise given the consistency with the corresponding CTP variations (Fig. S6) which differed from those for MODIS AOD (Fig. 10).

The overall impression from the CRE results is that the MODIS AOD results yield a clearer and more conventional picture of SW and LW CRE becoming stronger with aerosol loading, as in another recent study [Peng et al. 2016]. When viewed in isolation these results alone could then be taken as solid evidence that FIE, SIE, and invigoration take place. However, the fact that MERRA-2 AOD results are more complex and a less satisfying match to expectations is reason for a more cautious approach and for not rejecting the possibility that retrieval biases are conspiring to create correlations that may at first look indisputable, but ultimately do not signify underlying physical processes.

6. Meteorological variability vs AOD

We consider the results presented so far often showing striking cloud and precipitation changes as relative AOD varies an important addition to previous efforts that also attacked the ACPI problem from an observational angle. Nevertheless, we do not claim that results fitting the classic paradigms constitute unequivocal evidence of cloud modification by aerosol. In other words, we recognize that both aerosols and hydrometeors may also be modulated separately by environmental factors, so that their co-variations seen in this study are not exclusively manifestations of direct interactions. Rather, the diagnostics presented so far may reflect the combined effects of both independent co-variations and processes where interactions between aerosols and hydrometeors do indeed take place.

In this section we make a modest attempt to investigate whether aerosol variations are also associated with systematic meteorological variations. We focus on large-scale vertical motion (vertical pressure velocity OMEGA), air temperature and relative humidity variations. Information about these variables comes once again from MERRA-2. In accordance to the practice adopted throughout this paper, we composite these meteorological variables by either relative MODIS AOD or MERRA-2 AOD. In the latter case, we therefore have internally consistent compositing, with all data coming from the same source.

OMEGA at 500 hPa (OMEGA500) is a commonly used meteorological indicator of the dynamic state of the atmosphere, and given precedence receives due attention. For our low index CRs it may also be a good proxy of the averaged convective vertical mass flux, propagating from the cloud scale to the regional scale as simulated by *Fan et al.* [2012]. But MERRA-2 actually also provides full profiles of OMEGA, so it makes sense to expand the analysis to profiles for a more complete picture. We do the same for full-profile composites for two other atmospheric variables as well, namely air temperature and humidity. For all three variables we do not actually composite the values themselves, but standardized anomaly profiles defined at each level by the anomaly of the variable relative to its long-term mean, normalized by the standard deviation of the distribution of values at that level. In the figures that follow, positive anomalies (above average ascending motion and below average descending motion; higher than average temperature and humidity) are indicated in red, and negative anomalies (below average ascending motion and above average descending motion; lower than average temperature and humidity) in blue. These normalized anomalies give a better sense of the magnitude of systematic deviations relative to natural variability.

6.1 Vertical Velocity

OMEGA500 is composited in the same manner as all previous cloud or cloud-affected variables and the results are shown in Fig. 13 (MODIS AOD) and Fig. S9 (MERRA-2 AOD) following the form and conventions of previous figures. Most OMEGA500 variations of Fig. 13 appear weak, with prevalent, but not universal, tendencies of more ascent (stronger negative values) or less descent (weaker positive values) as relative AOD increases. Deviations from flatness (no changes in OMEGA500) can be discerned by comparing with the narrow (because of small natural variability) gray envelope which tracks closely the dashed line representing the overall mean, and seem more pronounced over land. CR12 over land is probably the most characteristic example of substantial and systematic OMEGA500 variability where downward motion at one end of the relative AOD range becomes ascending motion at the other. The shape of CR12's OMEGA500 composite curve is similar over both ocean and land when MERRA-2 AOD is used (Fig. S9), but for many other CRs the curves have different shapes with greater deviations from flatness compared to their MODIS AOD counterparts. Overall, vertical motion variability looks more substantial when matched to MERRA-2 relative AOD variability, and in addition the changes are not as monotonic. For the stormier CR2, CR4 and CR5, more abrupt changes in OMEGA500 seem to take place at the upper vigintiles of AOD. While the results are presented in a way consistent with what has preceded, we recognize that what we see is likely not a case of aerosol direct radiative effects affecting vertical velocity (as, for example, in Wang et al. 2015), but rather of vertical motion driving aerosol (or the two variables being uncoupled and co-varying through an external influence). Overall, the MERRA-2 results indicate that the dynamic state of the atmosphere has a rather strong association with aerosol, and can therefore be linked to both (re-analysis) aerosol and observed hydrometeor variability, which weakens to some extent the stature of our MERRA AOD results as evidence of direct influences of aerosol on hydrometeors.

Switching now to profiles, we start our presentation of normalized anomaly variability profiles vs AOD with Figures 14 and S10 for OMEGA. Recall that the results for the correspondent panels in these figures come from a different sample of grid cells both because of the more limited MODIS AOD availability, but also because the geographical mapping of relative AOD values of the common grid cells are different between the two datasets. One thing that can be immediately seen in both figures is that anomalies over the ocean are weaker than those over land. Moreover, the anomalies are generally stronger when sampling the more densely populated MERRA-2 AOD space. Nevertheless, even in this case, the area of the CTP-AOD percentile phase space covered by normalized anomalies greater than 15% is relatively small. This is especially true for MODIS AOD (Fig. 14), and for the CRs with more high clouds (smaller CR indices). Even if the anomalies are small, the patterns are nonetheless quite organized, with a majority of cases having positive OMEGA anomalies at the high end of AOD and negative anomalies at the low end of AOD. Such patterns suggest that stronger than average ascending motion (suppressed descending motion) is associated with higher AOD and weaker than average ascending motion (enhanced descending motion) with lower AOD. If such OMEGA anomalies also induce cloud property and precipitation changes, these can then appear as systematic, but misleadingly so, direct influences of AOD. CR12 once again shows such a clear correlation

between OMEGA and AOD, that one cannot help but wonder whether previous systematic variations of cloud properties and precipitation with AOD are mediated to some extent by vertical motion. This special case, however, should not distract us from the fact that for a large portion of our dataset (ocean), OMEGA anomalies, albeit apparently systematic, are rather weak.

6.2 Air Temperature

We now turn to air temperature anomalies in Figs. 15 and S11. If we were focusing solely on MODIS AOD, we would probably conclude that temperature profile variations are not involved in cloud property and precipitation variations previously seen. Again, this is because the stronger anomalies are confined to a small portion of the phase space. Yet, we once again do not obtain noisy anomaly patterns indicative of random variations, but rather patterns that are often well-organized in the portion of phase space they occupy. The anomalies have frequently dipole patterns with opposing signs at low and high altitudes. Differences between land and ocean are substantial, with ocean exhibiting more systematic behavior of colder than average low and middle troposphere occurring more often at the low AOD range. CR12 is now virtually featureless over ocean, and this also remains the case for MERRA-2 AOD (Fig. S11). But other than oceanic CR12, temperature profile anomaly patterns corresponding to MERRA-2 AOD are much more dramatic. The anomaly magnitudes are much larger, the dipole patterns remain, and the sign of the anomalies at small and large relative AODs reverses when transitioning from low to high index CRs (with CR6 serving as the transition regime). With AOD variations being relative, and coming from distributions built separately for each grid cell and season, geographical and seasonal influence on the anomalies should be largely suppressed. The fact that organized temperature anomaly patterns do exist, similar to OMEGA, prevents us from rejecting the hypothesis of thermodynamical influences mediating the previously seen co-variations between aerosol and hydrometeors.

We also examined how lower-tropospheric static stability (LTSS), defined as the difference of 700 hPa and 1000 hPa potential temperatures per Klein and Hartmann [1993], varies with relative AOD over the oceans, following Peng et al. [2016]. LTSS is probably relevant only for the CRs containing large amounts of boundary layer clouds, namely CR6-11. We do not show the results here, but note that LTSS either remains flat or decreases with relative AOD for both MODIS and MERRA-2 AOD. Since decreases in LTSS have been linked by Klein and Hartmann [1993] to decreases in CF, the LTSS variations are not mediating any SIE signals seen in our analysis (when using CF increases as criterion of SIE presence).

6.3 Relative Humidity

Lastly, we turn to relative humidity (RH) anomaly variations in Figs. 16 and S12. The amount of atmospheric moisture at the lower troposphere, as measured by RH has a direct influence on AOD because of swelling of aerosols in a moister environment, a physical process that (everything else being equal) increases AOD. Full profiles of RH and associated anomalies, on the other hand, can be considered indicators of meteorological variability. While for some CRs the RH-AOD correlation is seen consistently (MODIS AOD over ocean, except CR1), there are numerous cases where this relationship does not hold,

especially for MERRA-2 AOD composites (Fig. S12), where in grid cells containing CR1-3 cloud systems, the positive RH anomalies coincide with the lowest relative AODs (echoing Engström and Ekman 2010). Otherwise, the RH plots have some of the same characteristics of their OMEGA and temperature counterparts, namely a substantial degree of organization, especially for MERRA-2 AOD compositing, and the largest anomaly values occurring at the upper and lower quartiles of the AOD distribution. Since the anomalies are not flat or random, we cannot reject the hypothesis that RH mediates some of the previously shown systematic variations of cloud and cloud-affected quantities with AOD, as documented in previous studies [Quaas et al. 2010; Chand et al. 2012].

7. Summary

Numerous results depicting how precipitation, cloud properties, and CRE change against relative AOD were shown in this paper. We also showed changes for select meteorological variables vs AOD (which should perhaps be interpreted as changes of AOD with meteorology). Setting aside meteorology for now, does all this information, when viewed collectively, support some of the classic paradigms about how hydrometeors respond to aerosols, as summarized in Table 2? These paradigms of Table 2 reflect the most commonly accepted responses based on mechanisms first suggested by Rosenfeld et al. [2008] (invigoration), Twomey [1977] (FIE) and Albrecht [1989] (SIE). The simplicity and universality of these paradigms has been questioned repeatedly by juxtaposition against the inherent complexity of cloud microphysical mechanisms and of the multitude of possible cloud interactions with the surrounding environment that act as “buffering” mechanisms (a good summary is provided by Stevens and Feingold 2009). We touched on this subject by showing that the dynamical and thermodynamical environment exhibits systematic, albeit frequently weak, relationships with aerosols. The co-variations with meteorological factors, even when not dramatic, may influence cloud and precipitation behavior.

Despite such reservations, we deem useful to provide a concise summary of our findings on cloud, precipitation, and radiation responses to aerosol variations. We have attempted to convey this information in table form. Table 3a summarizes MODIS-based results while Table 3b summarizes MERRA-2-based results. We are showing the direction of change with AOD of seven variables: Precipitation rate PR (of clouds already precipitating, i.e., when only greater than zero precipitation values are considered), probability of precipitation (POP), defined and discussed in section 3, cloud properties CF, CTH (varying inversely to CTP), COT, CER, and radiative impacts SW CRE and LW CRE. Each table also has four columns that represents groups of CRs (ice, mixed, liquid, and CR12 on its own), and these are further broken when needed into land-ocean subcolumns. The tables are populated with upward and downward arrows when a direction of change can be clearly discerned and which are colored only when they are consistent, per Table 2, with FIE/SIE (blue arrows, the two effects considered collectively since they can operate simultaneously on the same types of clouds) or invigoration (red arrows). The following assumptions were used when populating the tables: ice CRs (CR1-CR3) also contain liquid condensate because they are often vertically developed, yet MODIS provides water phase information only near cloud top; invigoration theory does not inform us about particle size changes, but ice particle CER also obeys a form of FIE [Jiang et al. 2008]; no invigoration is sought for mostly liquid CRs

(CR6-CR11), only FIE/SIE; we do not consider liquid water path (increase) as a criterion for SIE occurrence because it is not retrieved independently from MODIS, but COT increases are consistent with both FIE and SIE (despite the fact that the inevitability of water path and COT increases has been questioned, e.g., Ackerman et al. 2004); CR12 which is overall of mixed phase, is often convective in nature, and is therefore a legitimate candidate for identifying invigoration signatures.

Absence of arrows in the two tables indicates inconclusive results. A few factors may make the results inconclusive: Different CRs within the group show inconsistent behavior; the curves are not monotonic (increases followed by decreases or vice-versa); in mixed phase clouds, the two different phases move in different directions. Consistency between the two tables is of great importance. When the arrows of corresponding table entries are not the same, then the choice of the specific AOD dataset matters, and this undermines the robustness and attribution of apparent ACPI relationships to physical processes.

Overall, the tables are well-populated and many arrows have been assigned colors, indicating that we were able to reach a conclusion about the nature of the ACPI (aside from meteorological influence, a hypothesis we were not able to reject). Our findings on precipitation are not supporting either invigoration or SIE: whenever the results are not inconclusive, then the direction of change is opposite to expectations. Only CR12 exhibits an increase in both precipitation intensity and probability of precipitation which goes hand-in-hand with increases in CF, COT, and CTH. If one looks for evidence of invigoration in variables other than precipitation and for CRs other than C12, then only the MODIS AOD compositing provides some support: the MERRA-2 results have only one red arrow for both the ice and mixed phase CR column. The results for the two aerosol datasets are much more consistent for liquid CRs where evidence for FIE (COT, CER, CRE) and SIE (CF, COT, CRE) exists in both tables. But precipitation suppression is not seen, possibly because the weak rainfall is at the limit of TMPA detection skills and water path changes may be disguising as precipitation changes. Note that had we conducted only CRE analysis to search for influences of aerosol on hydrometeors, then the MODIS AOD portion of the study would be consistent with all three classic ACPI paradigms occurring, and for MERRA-2 we would still be able to claim occurrence of FIE and SIE.

8. Discussion

Our rather ambitious near-global analysis expands significantly the scope of previous “high volume” aerosol-cloud-precipitation interaction studies [Niu and Li 2012; Chen et al. 2014; Gryespeerdt et al. 2014a;b; Peng et al. 2016], and confirms once again that finding robust evidence of attribution signatures in these interactions is a very challenging endeavor. But even lowering the bar and aiming at extracting only *apparent* consistent relationships between cloud-affected quantities and aerosol is fraught with difficulties. While not proving causal relationships, we believe that our study sets the stage for eventually achieving the more modest latter goal. We were able to demonstrate that when global cloudiness is partitioned by cloud regime, apparent relationships emerge in many instances, and when they do, they are often clear and unambiguous. At the same time, there are numerous examples where the relationships are not straightforward and contradict standard paradigms

or prior modeling results. Our analysis also sheds light on differences between land and ocean, exposes the considerable dependences on aerosol product forming the basis of the analysis, and underlines the additional perspective brought by the use of relative AOD.

The cloud regimes that contain ice and mixed phase clouds proved particularly challenging. They are the most substantial precipitation producers, yet we were unable to find meaningful precipitation responses to aerosols, in contrast to previous studies such as Koren et al. [2005; 2012]. Radiative signatures for these clouds indicate greater cloud radiative effects with more aerosol loading, but only for one of the aerosol datasets (MODIS). Results were much clearer for liquid regimes and largely consistent with first and second indirect effect predictions, regardless of the aerosol dataset, but no precipitation suppression could be discerned, perhaps because of observational limitations at low rain rates. Our most populous cloud regime of low cloud fraction exhibited in most cases dramatic apparent responses (except for particle effective size) to relative AOD increases with unambiguous upturns in precipitation, cloud extent, cloud optical thickness, cloud top height, and radiative effect. The fact that this behavior was also seen when re-analysis AOD was used gives some confidence that observational artifacts and retrieval biases do not dominate the apparent signals. However, we also found that meteorology (represented in a limited way by three variables) is correlated to some extent with AOD, so it may be mediating aerosol-hydrometeor relationships. This finding is a reminder of the challenges of aerosol effect attribution with the available dataset, which also comprise the inability to monitor the temporal evolution of clouds as aerosol conditions change, and the fact that our observations are unable to inform us on whether co-existence of aerosol and cloud translates to intimate coupling (examples of decoupling have been discussed in, e.g., Rosenfeld et al. 2014 and Wood et al. 2015).

In closing, we would like to provide further perspective on the challenges of remote sensing analysis, by bringing up what McComiskey and Feingold [2012] call the “analysis scale problem”. The essence of this problem is that the process scale, i.e., the scale at which the driving mechanisms operate is incompatible with the scale at which the analysis is performed. Our desire to conduct an almost global analysis and our reliance on the cloud regime concept steered us towards large-scale grid cells, much greater in size than the scales in which the relevant physical processes operate, a compromise that is expected to result in dampening of the higher resolution signals that may be present. Yet, we believe that some consistent signal propagation of small-scale processes to larger scales must be occurring and be strong enough for detection by large-scale datasets obtained from space-based remote sensing. Resolving this conundrum will require additional coordinated theoretical, modeling, and properly designed observational efforts, and undoubtedly a multi-year time horizon.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Key points

1. Diagnostic signals of Aerosol-Cloud-Precipitation interactions can be seen in a Cloud Regime analysis
2. Land-Ocean contrasts, dependences on the aerosol dataset, and meteorological ambiguity are found
3. The signals for mostly liquid regimes are more systematic than those for other regimes

Figure 1.

Near-global (50S-50N) histograms of AOD from MODIS Dark Target (top panel) and MERRA-2 (bottom panel). Each panel shows two histograms, in the MODIS case comparing retrievals from the Terra and Aqua satellites, and in the MERRA-2 case comparing the values of the corresponding 3-hour intervals containing the daytime overpasses of the two satellites.

Figure 2. Schematic description of how the daily AOD is derived for each grid cell. Top panels: when both the Terra and Aqua AODs are available within the grid cell of interest, their values are averaged. Middle panels: when one of the two is available, it becomes the daily AOD of the grid cell. Bottom panels: When neither AODs are available within the grid cell, we take the average of all Terra and Aqua AODs of grid cells that are immediate neighbors. When even the immediate neighbors have no AOD values, the AOD is undefined and the grid cell is discarded from the analysis. MERRA-2 always provides an AOD value, so only the top operation is applied.

Figure 3.

The centroids (average joint histograms) that define the MODIS global Cloud Regimes used as a framework for this study. These regimes were introduced by O16. Above each panel the global mean Cloud Fraction (CF) and Relative Frequency of Occurrence (RFO) is provided.

Figure 4.
Geographical distribution of the multi-annual mean RFO of the MODIS CRs of Fig. 3.

Figure 5.

TMPA precipitation rate (PR) composed by CR and relative MODIS DT AOD for only grid cells with non-zero precipitation, shown as curves of precipitation vs AOD vigintile for each CR. The set of twelve panels on the left are composites for ocean grid cells while those on the right for land grid cells. The color symbols on each curve indicate the relative AOD value that corresponds to fixed values of AOD from 0.1 to 0.5 in increments of 0.1 for the data sample used in each panel. The gray envelope represents an estimate of the range of natural (statistical) variability (see text) and the horizontal dashed line is the overall mean precipitation rate derived from the ocean and land samples of the CR's grid cells that have valid AOD values.

Figure 6.

TMPA probability of precipitation (POP), defined as the ratio of number of precipitating grid cells to total number of grid cells composited by CR and relative MODIS DT AOD, and shown as curves of POP vs AOD vigintile for each CR.

Figure 7.

Cloud fraction (CF) composited by CR and relative MODIS DT AOD. As in Fig. 5, the set of twelve panels on the left are composites for ocean grid cells while those on the right for land grid cells. Each panel has three curves, one for total (black), one for liquid (blue) and one for ice (cyan) CF. Each panel also has two CF ordinates, the left corresponding to the total CF and sometimes to a phase-specific CF, while the right can be for either liquid or ice composite CFs, as indicated by the color of the arrow. The color symbols are as in Fig. 5, and are placed only on the total CF curve since the sampling of AOD is the same for all three curves given that we include the zero values in phase-specific CF composites. Gray envelopes as in Fig. 5 are also included, but are often too narrow to discern, and essentially represent the overall mean CF (dashed lines) as discussed in the text.

Figure 8.

Similar to Fig. 7, but for total cloud optical thickness COT, i.e., no water phase discrimination, so that only a single curve of composite COT is shown for each panel.

Figure 9.

Similar to Fig. 7, but for cloud effective particle size REFF. Separate composite curves for each water phase are only shown for the CRs that have substantial proportions of both phases (CR4, CR5, CR12).

Figure 10.

Similar to Fig. 7, but for cloud top pressure CTP. Note that the ordinate scale has been reversed so that ascending curves indicate increases in cloud top height (CTH), and vice-versa.

Figure 11.

Similar to Fig. 10, but for SW CRE from CERES SYN1deg-daily. Ordinate scale has been reversed so that stronger SW CRE (more negative values) with increasing AOD is represented by curves with upward trends.

Figure 12.
As Fig. 11, but for LW CRE.

Figure 13.

As Fig. 12, but for 500 hPa pressure vertical velocity (OMEGA500) from MERRA-2.

Negative values indicate ascent (upward motion), while positive values descent (downward motion).

Figure 14.

Normalized anomalies of OMEGA500 profiles from MERRA-2 vs. MODIS relative AOD. The anomalies are defined with respect to the multi-annual mean of a CR across AODs at a particular vertical level and the normalization is performed by dividing with the standard deviation of the values at that vertical level. Positive anomalies (blue shades) indicate stronger than average descent or weaker than average ascent, while negative values (red shades) indicate stronger than average ascent or weaker than average descent. The mapping of relative AOD values to pre-selected absolute AOD values (from 0.1 to 0.5 in increments of 0.1) is shown with the colored symbols on the abscissa.

Figure 15. Similar to Fig. 14, but for MERRA-2 air temperature profiles. Positive values (red shades) indicate warmer than average, while negative values (blue shades) colder than average.

Figure 16. Similar to Fig. 15, but for relative humidity (RH). Positive means above average RH, and negative below average RH.

Table 1

Brief description of global character and average properties in the 50°S-50°N latitude zone of the 12 MODIS CRs used in this study.

CR	Description	RFO (%)	CF	Mean TAU	Mean CTP (hPa)
CR1	Mostly tropical with a pronounced presence in the Pacific ocean, and elsewhere within the confines of ITCZ. Contains a lot of the tropical cirrus associated with convection, but also deeper convective clouds.	5.31	0.84	9.8	292
CR2	Contains most of the optically thickest and tallest clouds of all CRs; includes storm systems produced by tropical and frontal convection and has the highest CF of all regimes.	3.65	0.97	23.1	344
CR3	Tracks tightly the geographical pattern of CR2, but contains the thinner elements of storm activity.	6.65	0.89	7.4	364
CR4	Extratropical and dominated by alto- and nimbo- type clouds in higher latitude storm systems. More prevalent during the summer months.	2.94	0.92	25.0	461
CR5	Closely associated with CR4, but with fewer optically thick clouds and more prevalence during the winter months.	2.71	0.85	9.4	479
CR6	Contains proportionally the most mid-level clouds and also has some congestus, with strong land presence.	3.97	0.82	20.2	583
CR7	Mainly a high latitude CR of plentiful thick stratus over both land and ocean with small RFO and big CF.	1.32	0.96	24.00	727
CR8	Boundary layer regime with occurrence peaks in known marine stratocumulus areas, but additional presence in far southern oceans and northern lands.	3.91	0.87	11.4	747
CR9	Similar to CR8 in CF, but of a more marine character and with shallower and optically thinner clouds; presence also peaks in known marine stratocumulus areas.	5.32	0.92	12.3	827
CR10	Also as marine as CR9, but with lower CF indicating greater relative presence of broken stratocumulus and shallow cumulus.	8.03	0.68	6.1	821
CR11	Even more broken stratocumulus and shallow cumulus than CR10, with small optical thicknesses and low cloud fractions; almost exclusively oceanic with negligible presence in high latitudes.	13.82	0.50	4.2	840
CR12	Comprises all 2D histograms of small CF with no characteristic shape, or histograms with a dipole pattern where high clouds overlap low clouds; has the highest global RFO and smallest CF, and except the nearly always overcast far southern oceans, is nearly omnipresent.	42.37	0.28	9.3	705

Table 2

Classical paradigms of aerosol-cloud-precipitation interaction and expected changes in cloud-affected quantities. We do not consider liquid water path (increase) as a criterion for SIE occurrence because it is not retrieved independently from MODIS. The direction of expected changes is the criterion used for coloring the arrows of Table 3.

Invigoration (ice and mixed CRs)	FIE (liquid CRs)	SIE (liquid CRs)
<ul style="list-style-type: none"> • Precipitation increase • CF increase • CTP decrease (CTH increase) • COT increase • SW/LW CRE increase 	<ul style="list-style-type: none"> • CER decrease * • COT increase • SW CRE increase 	<ul style="list-style-type: none"> • Precipitation decrease • COT increase • CF increase • SW/LW CRE increase

*
also for ice clouds

Table 3

(a) Direction of changes indicated by arrows (upward arrow=increase; downward arrow=decrease) for precipitation, cloud properties, and CRE, with MODIS AOD. The four columns represent groups of CRs (ice=CR1-3, mixed=CR4-5, liquid=CR6-11, and CR12), and these are further broken, when behaving distinctly, into land-ocean sub-columns. Arrows are colored when consistent with invigoration (red), FIE/SIE (blue), and are black otherwise. Absence of arrows indicates inconclusive results. (b) As in (a), but for MERRA-2 AOD.

(a)	Ice		Liq		Mix		CR12	
	Ocean	Land	Ocean	Land	Ocean	Land	Ocean	Land
PR	-	-	-	↑	-	↓	↑	↑
POP	-	-	↑	-	-	-	↑	↑
CF	↑	-	-	↑	-	-	↑	↑
COT	-	↑	↑	↑	↓	↑	↑	↑
CER	-	↓	↓	↓	-	-	-	-
CTH	↑	-	↑	↑	↑	↓	↑	↑
SW CRE	↑	-	↑	↑	↑	↑	↑	↑
LW CRE	↑	-	↑	↑	↑	↑	↑	↑

(b)	Ice		Liq		Mix		CR12	
	Ocean	Land	Ocean	Land	Ocean	Land	Ocean	Land
PR	↓	-	↑	-	↓	-	↑	↑
POP	-	↓	↑	-	-	-	↑	↑
CF	-	-	↑	-	-	-	↑	↑
COT	-	-	↑	-	↑	-	↑	↑
CER	↓	-	↓	-	-	-	-	-
CTH	↓	-	↑	-	↓	-	↑	↑
SW CRE	↓	-	↑	-	-	-	↑	↑
LW CRE	↓	-	↑	-	-	-	↑	↑