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5	Systematic Variations of Cloud Top Temperature
6	and Precipitation Rate with Aerosols over the
7	Global Tropics Seen from Space
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10	F. Niu and Z. Li
11	Dept of Atmospheric and Oceanic Sciences & Earth System Science Interdisciplinary Center
12	University of Maryland, College Park, MD 20740
13	
14	
15	Submitted to
16	Atmospheric Chemistry and Physics
17	
18	Correspondence to F. Niu (niufeng@atmos.umd.edu)
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1 Abstract

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Aerosols may modify cloud properties and precipitation via a variety of mechanisms with 3 4 varying and contradicting consequences. Using a large ensemble of satellite data acquired by the Moderate Resolution Imaging Spectroradiometer onboard the Earth Observing System's Aqua 5 platform, the CloudSat cloud profiling radar and the Cloud-Aerosol Lidar and Infrared 6 Pathfinder Satellite Observations (CALIPSO) satellite over the tropical oceans, we identified two 7 distinct correlations of clouds and precipitation with aerosol loading. Cloud-top temperatures are 8 9 significantly negatively correlated with increasing aerosol index (AI) over oceans and aerosol 10 optical depth (AOT) over land for mixed-phase clouds with warm cloud bases; no significant changes were found for liquid clouds. Precipitation rates are positively correlated with the AI for 11 mixed-phase clouds, but negatively correlated for liquid clouds. The distinct correlations might 12 be a manifestation of two potential mechanisms: the invigoration effect and the microphysical 13 effect. The former enhances convection and precipitation, whereas the latter suppresses 14 precipitation. If they are indeed the causes for the observed relationships, these effects may 15 change the overall distribution of precipitation rates, leading to a more extreme yet unfavorable 16 17 rainfall pattern of suppressing light rains but fostering heavy rains.

1 Introduction

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Several studies suggested that suppression of warm rain by aerosols may allow more cloud 3 particles to ascend above the freezing level, initiating an ice process in which more latent heat is 4 released thus invigorating convection (Andreae et al., 2004; Khain et al., 2005). A further study 5 6 using a parcel model suggests that this effect exists when ice processes are involved. The effect is much stronger for clouds with warm bases because cloud particles have longer distances to 7 grow before freezing (Rosenfeld et al., 2008). The invigoration effect may exhibit as systematic 8 9 increases in cloud-top heights or rain rates (Lin et al., 2006; Bell et al., 2008; Koren et al., 2005). However, the lack of full cloud geometry information in these studies, i.e. cloud top and base 10 heights, makes it hard to investigate the effect, especially if cloud phase and base are concerned. 11 This can be further complicated by warm rain processes for which aerosols are generally known 12 to suppress precipitation (Gunn and Phillips, 1957; Albrecht, 1989; Rosenfeld, 1999). 13

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The difficulties and complications may be alleviated considerably with the availability of more 15 extensive and variety of measurements made both from ground-based and space-borne sensors. 16 17 The former is illustrated clearly with 10 years of Atmospheric Radiation Measurement (ARM) that provides far more and richer information pertaining meteorology and aerosols than any other 18 ground observations (Li et al. 2011). On the other hand, the A-Train satellite sensors also 19 20 provide unprecedented amount of data that may also help tackle the complicated problems, especially cloud geometry and rainfall from its active sensors, and aerosol attributes from a 21 22 passive sensor.

In this study, we perform statistical analysis by classifying clouds according to their top and base heights as detected by space-borne active sensors, in combination with the aerosol index (AI) or aerosol optical thickness (AOT) retrievals from a passive sensor aboard the A-Train constellation. We show that clouds and precipitation are correlated with aerosol loading differently for different types of clouds. The rain distribution is also different under relatively clean and dirty conditions.

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8 Data and Methodology

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Satellite products employed in this study include one year's worth (2007) of observations from 10 CloudSat and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) as 11 well as the Aqua/MODerate resolution Imaging Spectroradiometer (MODIS) over the tropical 12 region extending from 20°N to 20°S. The MODIS products include Level 3 AOT at 550 nm and 13 the Angstrom exponent (Kaufman et al., 1997a; Remer et al., 2005). The Level 3 products were 14 generated by averaging the daily Level 2 data with a resolution of 10 km to $1^{\circ} \times 1^{\circ}$ grids. The 15 validation of Level 2 aerosol products with ground-based observations shows good agreement 16 17 (Li et al., 2007; Mi et al., 2007; Levy et al., 2007a, 2010). The CloudSat/CALIPSO products include cloud bases and top heights, cloud ice water paths, and precipitation rates (Stephens et 18 al., 2002; Haynes et al., 2009). The ECMWF-AUX data set is also used for atmospheric state 19 20 variables interpolated to each CloudSat pixel and cloud profiling radar (CPR) bin. The variables include atmospheric pressure, temperature, and specific humidity at each level. These data are 21 used to convert cloud top and base heights to cloud top and base temperatures. The column water 22 23 vapor and lower tropospheric static stability (LTSS), defined as the potential temperature

difference between the surface and the 700 hPa pressure level (Klein and Hartmann, 1993), are
calculated from the data set to constrain meteorological conditions. These products are
summarized in Table1.

4

All data from CloudSat/CALIPSO products are averaged over $1^{\circ} \times 1^{\circ}$ grids in order to match the 5 MODIS Level 3 aerosol product. Due to the narrow swath of CloudSat, at daily resolution, only 6 a "curtain" of CloudSat data is available within each $1^{\circ} \times 1^{\circ}$ grid box. Higher resolution data (e.g. 7 level 2) could be employed, but cloud-edge effect would introduce a more serious problem due 8 9 to cloud contamination. For aerosol indirect studies, it is essential to have both cloud and aerosol data parameters obtained within certain distance that cannot be too short to incur cloud 10 contamination nor too far apart for not being correlated. By adopting the 1° data, we essentially 11 make an assumption that AOD retrievals up to 55 km (half grid) remain the same, which would 12 be more sound than an alternative assumption that AOD is not contaminated by the adjacent 13 cloud pixel (Marshak et al., 2008). Data over the tropical region of 20°S-20°N are used. Grids 14 with AOT > 0.6 are excluded in our study to further reduce the possibility of cloud 15 contamination in AOT retrievals. Cloud top and base heights at original resolution are converted 16 17 to temperatures using temperature profiles from the ECMWF-AUX product. This information is used to define different cloud types: mixed-phase clouds with cloud-base temperatures (CBT) > 18 15° C and cloud-top temperatures (CTT) < -4° C, mixed-phase clouds with CBT in the range of 0-19 15° C and CTT <-4°C, and liquid clouds with CBT>0°C and CTT > 0°C. These criteria are 20 consistent with their ground-based study (Li et al. 2011) for the sake of comparison. Only single-21 layer clouds detected by the CloudSat are considered here. Table 2 summarizes the cloud types 22 23 under study.

2 The AI, which is a product of AOT and the aerosol Angstrom exponent, is binned and the dependencies of averaged cloud properties in each bin on the AI are examined over the ocean. AI 3 serves a better proxy for cloud condensation nuclei (CCN) than the AOT (Nakajima et al., 2001; 4 Feingold et al., 2006). How the precipitation rate and its distribution change with AI is also 5 6 examined. Over land, AOT is used instead of AI because the Angstrom exponent retrieved from MODIS over land is not quantitative and much less reliable than over oceans (Levy et al., 2010). 7 Tests on the dependencies of LTSS and column water vapor on AI or AOT are also done over 8 9 both land and ocean.

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11 **Results**

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While cloud and precipitation are affected by meteorology much more than by aerosols, the 13 influences are generally fast-evolving as seen in the changes of weather patterns/episodes. By 14 analyzing the relationship between cloud and precipitation with aerosol loading for a large 15 ensemble of cases, we intend to average out the influences of meteorological conditions. For the 16 17 large volume of satellite data as employed here, the meteorological conditions corresponding to a fixed range of AOT for all the data analyzed would have little difference from those for a 18 different range of AOT. As such, any dependence on AOT is more manifestation of the 19 20 influence of aerosols than that of meteorology. Using rich ARM data, Li et al. (2011) tested the dependence of aerosols on a variety of meteorological variables. No significant dependence is 21 found except for surface wind for it can sweep away aerosol particles. Precipitation can washout 22 23 aerosols beneath, but AOT is retrieved over nearby clear regions.

2 Fig. 1 shows the correlations of the cloud-top temperature and ice water path with AI over oceans and AOT over land for different cloud types as defined in Table 2. The CTT for mixed-3 phase clouds with warm bases is highly negatively correlated with AI, whereas the CTT for 4 liquid cloud does not show significant correlation. The results for mixed-phase clouds with cold 5 6 bases lie somewhere in-between. The sample sizes for each point in Fig.1a and 1c are shown in Table 3 and 4. The ice water path (IWP) is also positively correlated with AI for mixed-phase 7 clouds. Similar results are obtained over land using AOT instead of AI (bottom panels of Fig. 1). 8 9 Note that a logarithmic scale is used on the x-axis in the top panels. These satellite-based 10 observational findings are consistent very well with those from long-term ground data (Li et al. 2011). We cannot think of any mechanism except the aerosol invigoration effect (Andreae et al., 11 2004). Per the theory of Rosenfeld et al. (2008), the aerosol invigoration effect is more 12 significant for mixed-phase clouds with warm bases than those with cold bases because the 13 14 former generate more latent heat which can fuel cloud convection into higher altitudes.

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Previous studies (Khain et al., 2005; Lin et al., 2006) suggest that deeper clouds and enhanced
ice processes in polluted conditions could lead to enhanced rainfall. Therefore, the correlation
between AI and the precipitation rates from the CloudSat radar is also examined.

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The precipitation rates from mixed-phase and liquid clouds show very different responses to increasing AI (Fig.2). Note that only clouds with precipitation rates greater than 1mm/h are studied here. If the aerosol invigoration effect exists, it is significant chiefly for deep clouds which favor the production of heavy rain (Rosenfeld et al., 2008). To increase the sample size, only two types of clouds are differentiated here: mixed-phase and liquid clouds, regardless of cloud-base heights. Results show a positive correlation between precipitation rate and AI for
 mixed-phase clouds, but a negative correlation for liquid clouds.

3

In reality, precipitation may remove aerosols due to the scavenging effect, which could lead to a 4 5 false aerosol effect on the precipitation rate. The scavenging effect depends on precipitation 6 intensity, frequency, raindrop size distribution, aerosol and cloud properties, and also relative locations between aerosols and clouds (Jennings, 1998; Radke et al., 1980; Andronache, 2003), 7 which make it difficult to assess its effect on our results here. However, both theoretical and 8 observational studies (Andronache, 2003) showed that the scavenging effect generally increase 9 with the precipitation rate. If this is the case, we would expect to see a decrease in precipitation 10 11 rate with the AI. However, the precipitation rate for warm base mixed-phase clouds increases with increasing AI, which cannot be simply explained by the scavenging effect of rain. 12 Nevertheless, we cannot completely rule out any influences of the scavenging effect in our 13 14 analysis, as it is inherently a part of the aerosol-cloud-precipitation interaction process.

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As shown in Fig. 2, precipitation rate is positively correlated with the AI for mixed-phase clouds, 16 but negatively correlated with the AI for liquid clouds. Apparently, the opposite finding cannot 17 be explained by the scavenging effect, which would exhibit a ubiquitous negative correlation. 18 The opposite behaviors do, however, agree with the two dominant mechanisms that have been 19 proposed in many previous studies, namely, the suppression of warm cloud/rain (Albrecht, 1989; 20 Andreae et al., 2004), even though we cannot prove they are the causal effects with the limited 21 22 satellite data available. If the correlations are the manifestation of the causal relationships, they would explain the overall distribution of precipitation rate, as shown in Fig.3. It implies that 23 under polluted environments (AI>0.3), the frequency of occurrence of heavy rains (high 24

precipitation rates) is greater than that under clean conditions (AI<0.3). Conversely, light
rains (low precipitation rates) are more likely to occur under cleaner conditions than under
polluted ones.

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Constrained by available meteorological data, we examined the dependencies of several 5 meteorological variables on AI or AOT to see if they are the proxies of any meteorological 6 7 variables. Cloud formation is greatly influenced by changes in atmospheric stability and cloudscale relative humidity. However, cloud-scale relative humidity is hard to obtain. Therefore, two 8 atmospheric conditions: column water vapor and LTSS are used to check if AI or AOT are 9 correlated. The results are presented in Fig. 4. For mixed-phase clouds over oceans (red and blue 10 curves), column water vapor and LTSS are generally invariant with respect to the AI and the 11 AOT, so meteorological variable cannot explain either the changes in cloud-top temperature or 12 precipitation rate shown in Fig. 2. For mixed-phase clouds over land, the LTSS is positively 13 correlated with AOT, which means that the atmosphere becomes more stable as AOT increases. 14 Clouds tend to develop higher in the atmosphere under unstable conditions, therefore, the positive 15 16 correlation between LTSS and AOT cannot explain the decrease of cloud-top temperature with increasing 17 AOT. This is not surprising given that the LTSS is computed based on large-scale variables that seem 18 not be affected by the aerosol invigoration effect.

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Tests on column water vapor and LTSS under clean and polluted conditions were also conducted but no systematic differences were found. These tests do not support the premise that the change in the precipitation rate distribution is caused by systematic differences in meteorological conditions.

1 Concluding Remarks

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Strong but conditional correlations of cloud-top temperature, cloud ice water path, and 3 precipitation rate with the AI and AOT are observed from one year worth of satellite data 4 acquired by multiple sensors aboard the EOS Aqua platforms over the global tropical regions. 5 6 Results show that cloud-top temperatures are negatively correlated with AI over the ocean (or AOT over land) for both warm and cold base mixed-phase clouds, but the decreasing rate of 7 cloud-top temperatures with increasing AI/AOT is higher for the former. Cloud ice water path is 8 9 positively correlated with AI/AOT for mixed-phase clouds. Precipitation rate also has different correlations with AI for different types of clouds. It is positively correlated with the AI for 10 mixed-phase clouds, but negatively correlated with the AI for liquid clouds. 11

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The above findings are consistent with two prominent mechanisms: the aerosol invigoration 13 effect and microphysical effect, although it is beyond the scope and ability to establish their 14 causal relations due to the limits of observation data. It has been known that the invigoration 15 effect is stronger for mixed-phase clouds with warm bases than with cold bases due to the fact 16 17 that cloud particles in clouds with warmer bases have more chances to grow before freezing. Therefore, more liquid water is frozen to release more latent heat. This effect may lead to faster 18 decreasing rates of cloud-top temperatures with AI/AOT for clouds with warm bases than those 19 20 with cold bases as observed in Fig. 1. For liquid clouds, the dearth of ice processes does not incur any significant invigoration, and precipitation from these clouds is most likely suppressed 21 22 due to the aerosol microphysical effect. Tests on the dependencies of column water vapor and 23 LTSS on AI/AOT show that the above results cannot be explained by any changes in

meteorological conditions. The correlation study as presented here alone, however, cannot lead to an affirmative conclusion that these findings are the evidence of these two aerosol effects. The fact that all the findings presented here are in good agreement with those from ground-based long-term data, and the fact that they can be simulated with a cloud resolving model, lend us more confidence on the aforementioned explanations.

6

The findings reported here may have great implications for studying both the Earth's radiation 7 budget and the global hydrological cycle, if they are truly caused by aerosols. Smaller cloud 8 9 particles caused by higher aerosol loading reflect more solar radiation and result in negative forcing at the top of the atmosphere (the Twomey effect). However, this effect can be 10 compensated by changes in the overall distribution of cloud geometries. Clouds with higher tops, 11 which occur more frequently under polluted conditions, emit less longwave radiation than do 12 clouds with lower tops, leading to a warming instead of a cooling effect (Koren et al. 2010). The 13 suppression and enhancement of precipitation from shallow and deep clouds change the overall 14 distribution of precipitation rates. Heavy rain becomes more frequent and light rain becomes less 15 frequent under polluted than under cleaner conditions. This change could have a very large 16 17 impact on the hydrological cycle, leading to more frequent flooding or drought events under a polluted environment. 18

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Tables

Geophysical Parameter	Product	Sensor	Spatial Resolution
AOT	MYD08	MODIS	1×1 degree
Cloud Geometry	2B-GEOPROF-	CloudSat	Horizontal: 1.4 km×2.5 km
	LIDAR	and CALIPSO	Vertical: ~250m
Cloud Ice Water	2B-CWC	CloudSat	1.4 km×2.5 km
Column Water Vapor	ECMWF-AUX	N/A	1.4 km×2.5 km
Atmospheric	ECMWF-AUX	N/A	1.4 km×2.5 km
Temperature Profiles		~	
Precipitation Rate	2C-PRECIP-	CloudSat	1.4 km×2.5 km
	COLUMN		

2 Table 1. Summary of satellite and model datasets employed in this study.

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4 Table 2. Definitions of warm and cold base mixed-phase clouds and liquid clouds in this study.

	Mixed-phase clouds with warm bases	Mixed-phase clouds with cold bases	Liquid clouds
Cloud base temperature	>15°C	0-15°C	$>0^{\circ}C$
Cloud top temperature	<-4°C	<-4°C	>0°C

5

6 Table 3. Sample sizes for each data point in Fig.1.

AI	Warm base mixed- phase clouds	Cold base mixed-phase clouds	Liquid cloud
0-0.0152	26	63	3198
0.0152-0.0231	72	132	5908
0.0231-0.0351	139	257	10544
0.0351-0.0534	232	467	17137
0.0534-0.0811	399	716	23829
0.0811-0.1233	526	915	25081
0.1233-0.1874	411	756	15290
0.1874-0.2848	171	371	6030
0.2848-0.4329	65	159	2497
0.4329-0.6579	17	45	1075

АОТ	Warm base mixed- phase clouds	Cold base mixed-phase clouds	Liquid cloud
0.0-0.1	353	599	5586
0.1-0.2	814	1513	7277
0.2-0.3	612	1294	4283
0.3-0.4	340	806	2464
0.4-0.5	186	499	1443
0.5-0.6	81	298	951

1	Table 4.	Sample	sizes	for	each	data	point	in	Fig.	3.
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Figure Captions

Figure 1. Cloud-top temperature (A and C) and ice water path (B and D) as functions of AI/AOT for warm (blue dots) and cold (red dots) base mixed-phase clouds and liquid clouds (green dots) over ocean (upper panels) and land (lower panels). The right-hand axes of (A) and (C) are for liquid clouds. The AI is plotted using a logarithmical scale. R² and P are standard errors and P

- 6 values for each type of clouds, respectively.
- 7

Figure 2. Precipitation rate (A) and corresponding cloud-top temperature (B) as functions of AI
for mixed-phase (blue dots) and liquid clouds (red dots) over the ocean. Note that only clouds
with precipitation rates greater than 1 mm/h are included here. The right-hand y-axis of (B)
represents the cloud-top temperatures of liquid clouds.

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Figure 3. Frequency of occurrence of different precipitation rates under relatively clean and polluted conditions.

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16 Figure 4. Column water vapor (A and C) and LTSS (B and D) as a function of AI over the ocean

(upper panels) and AOT over land (lower panels) for warm (blue dots) and cold (red dots) base
 mixed-phase clouds and liquid clouds (green dots). LTSS is plotted in descending order; smaller

values (top part of the y-axis) indicate a more unstable atmosphere.