1 Large-Scale Covariability between Aerosol and Precipitation Over the 7-SEAS

2 **Region: Observations and Simulations**

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21 Abstract

22 One of the seven scientific areas of interests of the 7-SEAS field campaign is to

evaluate the impact of aerosol on cloud and precipitation (<u>http://7-</u>

24 seas.gsfc.nasa.gov/). However, large-scale covariability between aerosol, cloud and 25 precipitation is complicated not only by ambient environment and a variety of 26 aerosol effects, but also by effects from rain washout and climate factors. This study 27 characterizes large-scale aerosol-cloud-precipitation covariability through synergy 28 of long-term multi-sensor satellite observations with model simulations over the 7-29 SEAS region [10S-30N, 95E-130E]. Results show that climate factors such as ENSO 30 significantly modulate aerosol and precipitation over the region simultaneously. 31 After removal of climate factor effects, aerosol and precipitation are significantly 32 anti-correlated over the southern part of the region, where high aerosols loading is 33 associated with overall reduced total precipitation with intensified rain rates and 34 decreased rain frequency, decreased tropospheric latent heating, suppressed cloud 35 top height and increased outgoing longwave radiation, enhanced clear-sky 36 shortwave TOA flux but reduced all-sky shortwave TOA flux in deep convective 37 regimes; but such covariability becomes less notable over the northern counterpart 38 of the region where low-level stratus are found. Using CO as a proxy of biomass 39 burning aerosols to minimize the washout effect, large-scale covariability between 40 CO and precipitation was also investigated and similar large-scale covariability 41 observed. Model simulations with NCAR CAM5 were found to show similar effects to 42 observations in the spatio-temporal patterns. Results from both observations and

43 simulations are valuable for improving our understanding of this region's

44 meteorological system and the roles of aerosol within it.

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Key words: aerosol; precipitation; large-scale covariability; aerosol effects; washout;
climate factors; 7-SEAS; CO; CAM5

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49 **1. Introduction**

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51 In recent decades, atmospheric aerosols have attracted increasing attention, in part 52 owing to their relations to human activities and anthropogenic sources, air quality 53 and environment sustainability, and corresponding climatic impact. However, a 54 quantitative evaluation of global aerosol climatic effects still faces many hurdles and 55 have large uncertainties (IPCC, 2007). Conceivably, a better understanding of large-56 scale covariability between aerosol and precipitation in aerosol 'hotspot' regions is 57 needed as an important step towards their global evaluation. 58 Large-scale covariability between aerosol and precipitation consists of three 59 components. The first component is the effect of precipitation on aerosol (wet 60 deposition, a.k.a. the 'washout effect'). Washout always results in a negative linkage 61 between aerosol loading and precipitation, because stronger or longer raining 62 removes more aerosols from in the air. However, few studies have focused on the

63 effectiveness of washout processes to answer questions such as what is the effective

rain rate range that can wash out aerosols most efficiently. Imaging satellite sensors
are still having difficulties in measuring aerosols in the presence of clouds, whereas
ground measurements of aerosol depositions are still very limited and sparse. These
all reinforce the need for studies to start documenting the observational evidences
on aerosol-precipitation covariability.

69 The second component in the large-scale aerosol-precipitation covariability is 70 aerosol effects on precipitation. It includes direct, semi-direct, and microphysical 71 ('indirect') effects. The direct aerosol radiative effect is so-called because aerosol 72 reflects or absorbs solar radiation and therefore directly perturbs the radiation 73 budget, regional dynamics as well as large-scale climate system (Carlson and 74 Benjamin 1980; Miller and Tegen 1998; Diaz et al. 2001; Ramanathan et al. 2001; 75 Yoshioka et al. 2007). The semi-direct effect links aerosol absorption to cloud 76 amount, through excess radiation absorbed by aerosols within clouds leading to 77 faster evaporation of cloud water and in turn reducing cloud amount (Ackerman et 78 al., 2000; Feingold et al., 2005). On the other hand, microphysical or 'indirect' effect 79 emphasizes changes in cloud microphysics due to aerosols acting as cloud 80 condensation nuclei (CCN) or ice nuclei (IN) and changing cloud droplet number 81 and effective radius, cloud lifetime and precipitation efficiency (Twomey et al. 1984; 82 Albrecht 1989; Sassen et al. 2003; Lohmann and Feichter 2005). Model simulations 83 from Ackerman et al. (2000) found that mid-tropospheric radiative heating from smoke 84 absorption stabilized the lower troposphere and reduced cloudiness. Koren et al. (2004) 85 and Feingold et al. (2005) through observations and simulations respectively also demonstrated that cloud fraction decreased over Amazon in response to higher aerosol 86

87	optical thickness (AOT) that increased tropospheric solar absorption and radiation
88	warming. It is intriguing to explore how the cloud systems in tropical Asia, as another
89	aerosol hotspot, may respond to aerosol effects.
90	The third component in the aerosol-precipitation covariability is mediated by
91	climate factors such as the El Niño-Southern Oscillation (ENSO) and meteorological
92	factors such as water vapor (Prospero and Nees, 1986; Prospero and Lamb 2003;
93	Huang et al. 2009a-d). Some such factors can influence aerosol and precipitation
94	simultaneously (for example Huang et al., 2009a-d for cases over tropical Atlantic);
95	therefore, their effects should be addressed before examining the direct linkages
96	between aerosol and precipitation.

98 2. Study Area and Methodology

99

100 The Seven SouthEast Asian Studies (7-SEAS, http://7-seas.gsfc.nasa.gov/) region covers 101 a wide area from Java through the Malay Peninsula and Southeast Asia to Taiwan, where 102 biomass burning smoke is the prevalent aerosol type. One of the seven scientific areas of 103 interests of the 7-SEAS field campaign is to evaluate the impact of aerosol on cloud and 104 precipitation. The region from the tropics to subtropics has significant gradients in air 105 pollution varying from near pristine to heavily polluted atmospheric conditions. It 106 therefore provides a unique natural laboratory for atmospheric measurements and 107 aerosol-cloud-precipitation-climate interaction research. The cloud system in the region is so unique that they allow us to investigate not only the interactions of smoke and stratus
clouds in the north but also the relations of smoke and deep convective cloud in the
south.

111 In this paper, oriented by aerosol sources and cloud types, we will focus on two regions 112 of interests (ROI) within the 7-SEAS region. The northern ROI (N-ROI) is from 5N to 113 30N, where biomass burning is active during early boreal spring, and smoke plumes are 114 transported above low-level stratus over the South China Sea. An example from March 8, 115 2009 can be seen in Figure 1(a) where a heavy smoke layer was transported from 116 biomass burning sources in Thailand to northern Vietnam, southern China and the South 117 China Sea. Smoke layers appeared to be above the low-level stratus and significantly 118 darkened the clouds. The southern ROI (S-ROI) is centered over Borneo and has a peak 119 in aerosol loading in boreal fall. Figure 1(b) shows biomass burning over southern 120 Borneo on October 5th 2006. The smoke was then transported westward and mixed with 121 deep convective clouds over the Indian Ocean. Anthropogenic fires in equatorial Asia are 122 one of the major contributors to global fire aerosol emissions along with Africa and South 123 America (van der Werf et al., 2006, 2010). One focus of the upcoming SouthEast Asia 124 Composition, Cloud, Climate Coupling Regional Study (SEAC4RS) campaign during 125 August and September 2012 is on fire activity over equatorial Asia. The differences and 126 similarities between the two ROI in the large-scale aerosol-precipitation covariability are 127 of great interest to explore.

128 The main objective of this study is to explore the significance of the large-scale

129 covariability between aerosol and precipitation in the 7-SEAS region from observational

130 evidence perspective. The analysis was carried out in the following steps. Step 1: we first

131 examine the large-scale covariability between aerosol and precipitation from long-term 132 satellite datasets. ENSO effects are illustrated and removed. The response of rain 133 characteristics to aerosols is also investigated. Step 2: to minimize the washout effect, we 134 use carbon monoxide (CO) as a proxy for aerosol to reexamine the large-scale 135 covariability between CO and precipitation in the same region. Step 3: to compare 136 observational evidence to model simulations, we apply the similar data analysis 137 approach to model simulations with both aerosol radiative and microphysical 138 effects. Similarities and discrepancies from observations and simulations are 139 compared. Step 4: To illustrate aerosol effects on clouds, we further investigate 140 changes in cloud top height and outgoing longwave radiation in response to high 141 aerosol scenarios. Step 5: We further explore the corresponding changes in the top 142 of atmosphere (TOA) shortwave radiation flux to different aerosol anomalies. Step 143 6: The major ambient factor that influences aerosol effect on precipitation is liquid 144 water path. We therefore further stratify liquid water path to reexamine aerosol-145 precipitation covariability. Step 7: we will repeat the data analysis in Step 1, 4 and 5 146 by using datasets from aerosol prevalent seasons only, to further evidence the 147 seasonal significance of aerosol-precipitation covariability. The outcomes from the 148 above analysis would help us to improve our current understanding of the aerosol-149 cloud-precipitation-climate interaction in this region.

Section 3 describes all the datasets we used in this study, followed by Section 4,
which introduces all the findings. Section 5 concludes the study with summaries and
discussions.

3. Datasets and Data Analysis

156	The major datasets used in this study are tabulated in Table 1. In Step 1 analysis, for
157	aerosol, we used the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) version 3
158	aerosol optical thickness (AOT, <u>http://disc.sci.gsfc.nasa.gov/dust/</u> ; Sayer et al.,
159	2011) as primary dataset, complemented by the Moderate Resolution Imaging
160	Spectrometer (MODIS) AOT (Kaufman et al., 1997; Remer et al., 2005; Hsu et al.
161	2004, 2006) and the Total Ozone Mapping Spectrometer (TOMS) aerosol index (AI,
162	Herman et al., 1997; Hsu et al., 1999). SeaWiFS AOT was preferred because of it is
163	the longest single-sensor AOT dataset with retrievals over all surface land and
164	ocean. For clouds, we used the International Satellite Cloud Climatology Project
165	(ISCCP) cloud products for cloud type and cloud amount analysis (Schiffer et al.,
166	1983). For precipitation, the Global Precipitation and Climatology Project (GPCP)
167	(Huffman et al., 1997) and the Tropical Rainfall Measuring Mission (TRMM)
168	precipitation products (Fisher, 2004) were used. TRMM was more preferable to
169	match up with concurrent SeaWiFS aerosol retrievals owing to its higher temporal
170	and spatial resolution. For Step 2 analysis, the AIRS carbon monoxide (CO) data
171	(McMillan et al., 2011) were used as proxy of aerosol loading because CO is a direct
172	product from biomass burning, and it is not washed out from the atmosphere by
173	rain as easily as aerosols. For Step 3 analysis, a 5-year NCAR Community
174	Atmosphere Model (CAM5) simulation, that includes both aerosol radiative and

175 microphysical effects, were used to compare to observational evidence. For Step 4 176 analysis, the MODIS cloud top pressure (CTP, Menzel et al., 2008) and the NCEP-DOE 177 Reanalysis II outgoing longwave radiation (OLR) data (Kanamitsu et al., 2002) were 178 used to illustrate possible aerosol-cloud interaction mechanisms. For Step 5 179 analysis, the Cloud and Earth's Radiant Energy System (CERES) top of atmosphere 180 (TOA) shortwave flux data at both clear sky and all-sky conditions (Wielicki et al, 181 1996) are used to observe the aerosol-related radiation changes. For Step 6, 182 precipitation changes at different stratified liquid water paths retrieved from NCEP-183 DOE reanalysis II will be investigated to elucidate effects from ambient moisture. 184 Lastly for Step 7, investigations are repeated using the above-mentioned datasets but for aerosol prevalent seasons only to further verify the significance of aerosol-185 186 induced cloud, precipitation and radiation changes in high aerosol scenarios. 187 Monthly datasets for aerosol, cloud, precipitation and radiation were used 188 throughout the study, except the TRMM 3B42 daily rainfall data was used to count 189 the number of days without rain ('no-rain days'). Monthly data was used because 190 aerosol events in this region are usually of synoptic scale and therefore the 191 aggregation from daily to monthly minimizes the impact of washout effect and cloud 192 contamination on spatial completeness. Thus the relative comparison between 193 monthly aerosol anomalies is not impacted by these effects as significantly as the 194 relative comparison between daily aerosol anomalies. Raw data were detrended to 195 remove long term trends. This is to avoid complications from variability at climate 196 scale. Seasonal cycles were then removed so that we keep our primary focus on the 197 anomalous changes of these parameters. Linear effects from ENSO were removed

198 from the anomalies through a linear regression to calculate residual anomalies. 199 Normalization was performed by dividing the residual anomalies by the 200 corresponding seasonal cycle of standard deviation calculated from the raw data. In 201 this way, the residual normalized anomalies are fairly comparable from month to 202 month for evaluating effects from anomalous aerosol conditions on anomalous 203 cloud and precipitation changes. To category high and low aerosol scenarios, the 204 aerosol residual normalized anomalies are sorted into three (low, middle, and high) 205 terciles. The corresponding residual anomalies of cloud and precipitation variables 206 between high and low aerosol tercile months are compared and difference 207 calculated to illustrate aerosol-associated variability.

208

209 **4. Results**

210

211 4.1 Aerosol, precipitation and cloud climatology

Figure 2 provides more information about the spatial variability of aerosol and

213 precipitation during the two smoke-laden aerosol seasons: Feb-March-April for the

214 North ROI (N-ROI) and Sep-Oct-Nov for the South ROI (S-ROI).

215 The N-ROI and S-ROI are shown in Figure 2(a) by dashed and solid outlines

respectively. The aerosol domains (N-ROI, [10-30N, 95-130E]; S-ROI, [10S-10N, 95-

217 130E]) are shown in yellow while the precipitation domains (N-ROI, [15-25N, 100-

218 125E]; S-ROI, [5S-5N, 100-125E]) are shown in red. We selected a larger aerosol

domain than precipitation domain to further overcome the potential complications
from cloud contamination on aerosol data uncertainties. The assumption is that
aerosol levels within clouds can be approximated by aerosol levels in closeby cloudfree retrievals because aerosol events are usually of synoptic scales therefore
aerosol levels in the immediate adjacent cloud free vicinity are fairly similar to
aerosol levels within the cloudy areas.

225 TOMS AI, with its capability to detect aerosol above clouds qualitatively, observed 226 significant amount of aerosol in the N-ROI domain in early spring (Figure 2a). 227 Similarly SeaWiFS AOT and MODIS AOT also showed a similar spatial pattern of 228 aerosol in the same season. Such similarity in aerosol spatial distribution between 229 AOT and AI in monthly data further evidences that 1) cloud contamination in 230 monthly data is minimized in the daily-to-monthly data aggregation process, and 2) 231 in monthly data, AOT is as indicative as AI to observe aerosol existence, despite the 232 persistence of clouds in this region. 233 AIRS CO data also shows elevated level of CO over Thailand and the South China Sea 234 in early spring and over Indonesia during fall, owing to biomass burning and

atmospheric transport. However the spatial pattern of AOT and CO are not always

236 consistent particularly during early spring when the CO distribution seems much

237 stronger over South China Sea than over Northern Vietnam and Southern China,

while AOT indicates the opposite patterns. Over S-ROI, on the other hand the spatial

239 patterns of CO and AOT are more consistent with each other. Additionally, the

overall enhanced signal in the AIRS CO data show a more widespread pattern of

biomass burning emissions from the Borneo over the surrounding ocean regions,
during autumn season relative to lower emissions during spring-time. The overall
distribution for the two seasons suggests a shift of the enhanced biomass burning
zone from the Indochina peninsula (during spring) to the equatorial islands (during

autumn).

246 From precipitation data over the N-ROI during the premonsoon season in early 247 spring, the stratus clouds that are often under the influence of smoke transported 248 from the Indochina region do not precipitate much (monthly precipitation over N-249 ROI is less than 3 mm/day, Figure 2i). The deep convective rain in the South ROI, 250 however, can produce 10 mm/day in both seasons (Figure 2i and 2j). It is worth 251 pointing out that the regions are under the influence of different regimes such that 252 the smoke aerosol prevails in a low precipitation efficiency stratus frontal system 253 over the N-ROI, whereas the presence of smoke in the S-ROI co-exists in the deep 254 convective cloud regime i.e., associated with higher precipitation efficiency.

255 The seasonal variation of aerosol and precipitation can also be seen in the time 256 series in Figure 3. In general, both aerosol loading peaks in early spring and autumn 257 during smoke seasons were well captured by TOMS AI, Aqua MODIS and SeaWiFS 258 AOT, and AIRS CO measurements. Aqua MODIS and SeaWiFS AOT showed very 259 comparable regional averaged AOT levels to each other while SeaWiFS has extended 260 data record back to late 1997. It is also noteworthy that generally the AOT/AI peaks 261 over the N-ROI are consistently higher than that over S-ROI from all three disparate 262 satellite measurements (TOMS, SeaWiFS and MODIS). In addition, the CO time series

also indicates a similar inter-annual variation with higher emissions over the N-ROI
compared to S-ROI. Precipitation in N-ROI showed strong contrast between the
boreal summer monsoon season and the rest of the seasons. In S-ROI, however, the
seasonal contrast is not as significant as in N-ROI and the peak season is in boreal
winter.

268 The fractional contributions from high, middle and low clouds as derived from

269 ISCCP datasets over the two ROIs are shown in Figure 4. High and middle clouds

that are strongly associated with deep convection are more often observed in S-ROI

than in N-ROI (Figure 4a,b). In the contrast, low clouds that are usually stratus

clouds in the 7-SEAS region are more prevalent in N-ROI (~25%) than in S-ROI

273 (~15%) (Figure 4c). These observations of contrasting high and low cloud

274 prevalence over the two regions are consistent within the general precipitation

275 regimes of the 7-SEAS region.

276 4.2 Large-scale aerosol and precipitation covariability from satellite

277 observations

Before the direct linkage between aerosol and precipitation are further investigated,
it is crucial to minimize climate factor effects, such as ENSO, as these impact both
aerosol and precipitation simultaneously. Relation between ENSO Nino3 index and
precipitation anomalies is significantly negative at 95% confidence level over S-ROI
but positive over N-ROI, as suggested by the spatial correlation distribution (Figure
5(a)). In contrast, ENSO Nino3 index and aerosol anomalies are positively correlated
with 95% confidence level over S-ROI, suggesting the association of enhanced

285	biomass burning activities with stronger ENSO signals, and a rather weak
286	relationship is found over the N-ROI with a negative correlation at a less confidence
287	level (Figure 5(b); Field et al., 2009). This implies that ENSO, as a climate factor,
288	modulates aerosol and precipitation simultaneously. Over the 7-SEAS region, ENSO
289	partially contributes to a negative large-scale covariability between aerosol and
290	precipitation. In this study, ENSO effects on aerosol and precipitation were linearly
291	removed through multivariate regression to focus on the aerosol-precipitation
292	interactions that are independent of the modulating climate factors.
293	In the Step 1 analysis, we first calculated changes in the precipitation anomalies
294	between high and low aerosol tercile months sorted by the normalized aerosol
295	anomalies in order to identify covariability of aerosol and precipitation fields . Two
296	independent runs were conducted for comparison: 1) SeaWiFS AOT vs. TRMM
297	precipitation, for relatively short-term but high quality observational data (1998-
298	2009 with two missing months, total 142 months); 2) TOMS AI vs. GPCP
299	precipitation, for long-term datasets (1979-2000 with some data gaps, total 235
300	months). AOT difference between high and low aerosol tercile months were first
301	shown in Figure 6(a) for N-ROI and 6(b) for S-ROI with a regional averages of 0.13
302	and 0.09 respectively. The large-scale covariability between aerosol and
303	precipitation are more negative in the S-ROI shown in both long and short-term data
304	analysis (Figure 6d vs. 6f). But the results from short-term and long-term data
305	analysis are not so consistent for the N-ROI cases: it appears to be precipitation
306	reduction in the TOMS AI vs. GPCP runs but precipitation increases in the SeaWiFS
307	AOT vs. TRMM runs (Figure 6c vs. 6e). The low covariability between aerosol

loading and precipitation over the N-ROI could be attributed to the low precipitationefficiency of the stratus clouds during pre-monsoon season.

310 Because the SeaWiFS data has similar overlapping coverage as TRMM, i.e. from 1997 311 onwards, and TRMM provides large variety of precipitation measurements, we were 312 able to examine changes in rain rate and latent heating from TRMM TMI profiling 313 product (3A12), no-rain days counted from TRMM daily precipitation product 314 (3B42). Over the S-ROI, along with the reduction in total precipitation, rain intensity 315 in terms of instantaneous rain rate increased while the number of raining days 316 decreased between high and low aerosol terciles (Figure 7b and 7d). Systematic 317 decreases of latent heating were observed below 10 km during anomalously high 318 aerosol lading periods (Figure 7e and 7f), consistently indicating overall 319 precipitation reduction at these levels. Over the N-ROI, however, the observed 320 changes in rain rate, no-rain days and latent heating profiles were not as significant 321 as over the S-ROI (Figure 7a,c and e). This is because stratus clouds over N-ROI are 322 not usually precipitating. It is therefore difficult to measure marginal changes in 323 precipitation attributable to changes in aerosol. On the other hand, deep convective 324 clouds and precipitation are present in S-ROI most of the year. It was reported that 325 aerosols induce cloud and precipitation suppression (Rosenfeld 1999) as well as 326 invigoration (Koren et al., 2005; Rosenfeld et al., 2008) depending on ambient moisture 327 conditions and sensitivity of cloud condensation nuclei (CCN) changes to aerosols. To 328 further elucidate the mechanisms behind the negative large-scale covariability of aerosol 329 and precipitation over the S-ROI, more observational and simulated evidences in Steps 2 330 to 7 is needed.

331 4.3 Large-scale CO and precipitation covariability

332	One of the largest complications to studying aerosol-precipitation interaction in the
333	large-scale aerosol and precipitation covariability is the washout effect. Washout
334	effect contributes to a negative relationship between aerosol loading and
335	precipitation. As previously shown in Figure 2(d), (h) and Figure 3(a), CO, as
336	produced from biomass burning, appears to resemble the aerosol variability pattern
337	fairly well. However, as an atmospheric trace gas species, CO is not washed out by
338	precipitation as effectively as aerosols are. Thus using CO as a proxy to approximate
339	aerosols that modulate cloud and precipitation processes helps minimize the
340	washout effect to some extent in the aerosol-precipitation studies.
341	Therefore in the Step 2 analysis, following the same data analysis procedure as in
342	Step 1, large-scale covariability between CO and precipitation over N-ROI and S-ROI
343	are shown in Figure 8. Over the S-ROI, higher CO concentration in the air is shown to
344	be associated with significant precipitation reductions, while it is not significant in
345	the N-ROI. These observations involving co-variability of CO and precipitation are
346	similar to that previously shown using aerosol loading and precipitation. The
347	implication is that, with less influence from wet removal, the aerosol-induced
348	precipitation changes that are more attributable to aerosol radiative and
349	microphysical effects, are more observable in the deep convective clouds than over
350	the stratus with less precipitation. Over the equatorial Asia, the net aerosol radiative
351	and microphysical effects are more likely to induce precipitation reduction.

352 4.4 Large-scale aerosol and precipitation covariability from model simulations

353 While observational evidence provides valuable inputs for model parameterization 354 improvement, model simulation in turn helps identify aerosol effects from the 355 observed aerosol and precipitation covariability. This requires comparisons 356 between observational evidence and model simulations. 357 In the Step 3 analysis, 60-month CAM5 model simulations, which include both 358 aerosol radiative and microphysical effects (Liu et al., 2011; Ghan et al., 2011), were 359 used for this research. The large-scale covariability between model-simulated 360 aerosol and precipitation was negative over S-ROI, in agreement with the results 361 from observational evidences in Figure 6 (b, d) and 8(b). Over N-ROI however, 362 precipitation changes between high and low aerosol terciles were less organized. 363 similar to Figure 6(a) and 8(a). Therefore, model simulations of reduced 364 precipitation over the equatorial Asia region support the observed covariability. 365 From the point of view of model development, it is encouraging that the simulations 366 matched observational patterns reasonably well. It is noteworthy however that the 367 60-month simulation is too short to draw any conclusive remarks. Longer model 368 runs with only aerosol radiative forcing or only aerosol microphysical effect should 369 be conducted in comparison to a reference run with only washout effect. In doing so, 370 relative contributions from aerosol radiative and microphysical effects can thus be 371 quantified with more confidence. Further investigations on those perspectives will 372 follow this study.

373 4.5 Aerosols versus CTP and OLR

374 As discussed in the introduction, because of the coherent relationship between cloud and precipitation, thus aerosol-related cloud changes would subsequently 375 376 induce precipitation changes. Therefore it is also of great interest to see how 377 aerosols may impact cloud through aerosol-cloud interactions. In the Step 4 analysis 378 of this study we explored the changes in cloud top pressure (CTP), cloud amount 379 (CA) and outgoing longwave radiation (OLR) associated with aerosol loading 380 anomalies, i.e. from aerosol low tercile to high tercile months. Figure 10 shows that 381 over both N-ROI and S-ROI, there were clear increases of CTP but small decrease of 382 CA, indicating less cloudiness with lower cloud top height in response to high AOT 383 anomalies that also leads to increases of OLR. Interestingly, N-ROI also showed 384 significant aerosol-induced changes in clouds although its changes in precipitation 385 were not systematically observed in previous steps. The observed increase of CTP 386 over the N-ROI as induced by smoke aerosols above stratus is consistent with the 387 results of Wilcox (2010) that South African smoke over South Atlantic marine 388 stratus leads to cloud layer subsidence, attributed to solar absorption by smoke 389 above marine stratocumulus clouds increasing the buoyancy of free-tropospheric 390 air above the temperature inversion capping the boundary layer. The observed CTP 391 increase over the S-ROI is also in line with Huang et al. (2010a-d) that African 392 aerosols were associated with suppression of deep convective cloud and 393 precipitation over Atlantic Marine ITCZ and West African Monsoon, indicating cloud 394 responses to more aerosol radiative forcing or semi-direct effects that overturns 395 aerosol microphysical effects when aerosol anomalies are high (> 0.25). This is 396 because very thick aerosol layers would reduce surface latent and sensible heating.

warm the mid-troposphere, stabilize the atmosphere, results in less convection and
convective rainfall (Ramanathan et al., 2001; Koren et al., 2008; Rosenfeld et al.,
2008; Huang et al., 2009).

400 4.6 Aerosols versus Shortwave Radiation Flux

In clear sky conditions, aerosol radiative forcing can change the top of atmosphere
(TOA) shortwave radiation fluxes via aerosol absorption and scattering. In aerosolladen clouds, aerosols can also alter cloud albedo or cloudiness and consequently
influence TOA shortwave radiation flux indirectly. To elucidate such aerosol effects,
the differences in all-sky and clear sky shortwave flux between high and low aerosol

406 tercile months are plotted in Figure 11.

407 For clear sky cases, both N-ROI and S-ROI indicated that high aerosol leads to higher

408 TOA shorwave flux, which implies stronger aerosol scattering in the heavy aerosol

409 laden atmosphere. This aerosol feedback signal seems rather stronger over S-ROI

than over N-ROI, probably because the S-ROI is mostly ocean with darker surface so

the aerosol-induced TOA shortwave flux increase is relatively larger and more

412 observable.

413 For all-sky cases, both N-ROI and S-ROI also consistently showed negative changes

414 in the TOA shortwave flux. Over N-ROI, there are two reasons for explanation:

415 Firstly, aerosols are frequently transported above stratus clouds, darken the clouds

- 416 by reducing cloud albedo and thus reduce TOA shortwave flux. Secondly, aerosol
- 417 reduces cloud top height as seen in the increased cloud top pressure in Figure 10.
- 418 Consequently, such aerosol-induced changes in cloud optical properties also impact

419 TOA shortwave flux. For example, lower cloud optical depth would have less cloud 420 scattering and thus result in decreased TOA shortwave flux. Over S-ROI however, 421 aerosols are normally mixed with deep convective clouds. Aerosol can also change 422 cloud albedo because aerosols increase number of cloud condensation nuclei (CCN) 423 and result in more absorbing cloud particle (cloud albedo effect). Secondly, aerosol 424 is also related to suppression deep convection, seen as the increased cloud top 425 pressure and outgoing long wave radiation in Figure 10. Therefore the aerosol-426 induced negative changes of cloud scattering significantly decreases TOA shortwave 427 radiation flux (Figure 11).

428 4.7 Stratifications of cloud liquid water path

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One factor is the ambient moisture or water vapor that also modulates cloud
formation and precipitation processes. To further elucidate our observed aerosol
and precipitation covariability is not sensitive to water vapor in this region, in Step
5 of our analysis, we stratified concurrent liquid water path (LWP) into three
terciles, and calculated precipitation changes between high and low aerosol tercile
months same as in Figure 6 but under at high and low LWP terciles respectively.

Figure 12 compares the results from low and high LWP conditions over S-ROI only.

436 In general, the large-scale negative covariability between aerosol and precipitation

437 persisted under both low and high LWP conditions, which means water vapor effect

alone cannot explain the covariability observed in Figures 6-9 and thus evident the

439 observability of large-scale aerosol effects on precipitation. However the reduction

440 in precipitation seems more significant in the high LWP conditions (Figure 12(b)).

441 Because LWP and precipitation anomalies are usually positively correlated, such enhancement of precipitation suppression owing to higher LWP condition actually 442 443 indicates that if aerosol effects on deep convective precipitation are significant and 444 observable from both observations and models as in Figure 6-10, it favors moist 445 conditions more than dry conditions. Such LWP preference of precipitation 446 suppression is consistent with Fan et al. (2009) for deep convective cases that the 447 decreasing rate of convective strength is greater in humid air than that in dry air 448 when wind shear is strong.

449 4.8 Precipitation-Cloud-Radiation Changes in Aerosol Prevalent Seasons

450 As discussed in the previous steps, aerosol effects on cloud, precipitation and 451 radiation are seemingly significant. More convincing evidence should be seen in 452 aerosol prevalent seasons when changes in aerosol anomalies are larger. Thus, we 453 focus our analysis on boreal early spring season (Feb-Mar-Apr) for N-ROI case and 454 boreal fall season (Sep-Oct-Nov) for S-ROI case and calculate the difference 455 composites of precipitation, all-sky shortwave TOA flux, and cloud top pressure 456 (Figure 13). In comparison to the figures with all season datasets (Figure 6 for 457 precipitation, Figure 10 for CTP, Figure 11 for SW flux), the changes in all three 458 parameters were much larger in comparison to the all season cases. In general, 459 precipitation inhibition, cloud top suppression, and reduction of all-sky shortwave 460 TOA flux were significantly observed in the aerosol prevalent seasons over both N-461 ROI and S-ROI. This further evidences the sensitivity of these meteorological

462 parameters to aerosol changes because aerosol is a more predominant factor in the463 weather system during these aerosol prevalent seasons.

464

465 **5. Summary and Discussions**

466

467 We investigated the large-scale covariability between aerosol, cloud, precipitation, 468 and radiation over the 7-SEAS region by using both satellite observations and model 469 simulations. The study was conducted in seven major steps of analysis: 1) 470 observational evidence of large-scale aerosol and precipitation covariability; 2) 471 observational evidence of large-scale CO and precipitation covariability; 3) model 472 simulations of large-scale aerosol and precipitation covariability; 4) observational 473 evidence of large-scale aerosol and cloud covariability; 5) observational evidence of 474 large-scale aerosol and shortwave radiation; 6) stratification of cloud liquid water 475 path in the large-scale aerosol and precipitation covariability; 7) observational 476 evidence in aerosol prevalent seasons. 477 Main results are summarized in Table 2: 478 Over the deep convective regime in the S-ROI, high aerosols loading is associated 479 with overall reduced total precipitation (-1.23 mm/day and -1.53 mm/day from two 480 independent analysis) with intensified rain rates (+0.029 mm/day) and decreased 481 rain frequency (+4 no-rain days), decreased tropospheric latent heating, suppressed

482 cloud top height (+26.8hPa in CTP) and cloud amount (-1.8%), increased outgoing

483 longwave radiation (+4.41W/m²), enhanced clear-sky shortwave TOA flux

484 (+2.59W/m²), but reduced all-sky shortwave TOA flux (-4.20W/m²).

485 In contrast over the stratus cloud regime in the N-ROI, the overall changes in the

486 cloud, precipitation, and radiation variables between high and low aerosol scenarios

487 are less significant. In anomalously high aerosol loading scenario, precipitation

488 changes are not consistent in two independent analysis (-0.30 mm/day vs. +0.31

489 mm/day) but rain rates decrease (-0.11 mm/day) with slight higher rain frequency

490 (-1 no-rain days). High aerosol loadings are also associated with decreased

tropospheric latent heating, slightly suppressed cloud top height (+4.36 hPa in CTP)

and very marginal change of cloud amount (-0.12%), increased outgoing longwave

493 radiation (+3.21 W/m²), enhanced clear-sky shortwave TOA flux (+1.36 W/m²), but

494 reduced all-sky shortwave TOA flux (-0.41 W/m^2).

495 More detailed summary and discussions are organized in the following key points:

1) The 7-SEAS region provides us with a unique testbed for observing the climatic

497 effects of biomass burning aerosols on cloud and precipitation due to its active

biomass burning activities. Moreover, the different cloud systems in the Northern

and Southern parts of the region allow us to directly compare different characteristics

500 of large-scale aerosol and precipitation covariability in different cloud regimes. The

501 upcoming SouthEast Asia Composition, Cloud, Climate Coupling Regional Study

502 (SEAC4RS) campaign, with its focus of aerosol-cloud-precipitation interaction in tropical

503 Asia in August and September 2012, will provide valuable in-situ radiometric

504 measurements to further support or verify the satellite and model results we show in this

study. Furthermore, this study provides more valuable information on the regional scalecovariability of aerosol-cloud-precipitation interactions.

507 2) Large-scale aerosol and precipitation covariability consists of three major components: 508 aerosol effects on precipitation, washout effect, and climate factor effects. Observational 509 evidence has to be investigated along with model simulations to truly separate the 510 components explicitly. However, observational evidence can still provide useful insights 511 to better understand the overall climatic effects of aerosol. For example, the consistency 512 between aerosol-precipitation covariability and CO-precipitation covariability can help us 513 better understand the significance of aerosol effects on precipitation, by minimizing the 514 washout effect. In addition, ENSO can modulate aerosol and precipitation variability in 515 this region simultaneously. Therefore it is important to separate the three components, 516 with the assistance from observations and simulations, before any quantitative evaluation 517 of global or regional scale aerosol climatic effects are conducted. 518 3) Large-scale covariability between aerosol and precipitation are different over the 519 stratus region in N-ROI and the deep convective cloud region in S-ROI. From the 520 sequential Step 1 to Step 5 analysis, both satellite observations and model simulations 521 observed systematic negative covariability between aerosol and precipitation. Although 522 eventually we have to rely on model simulations to demonstrate the exact dominant 523 aerosol effect in this region, it is encouraging that the large-scale aerosol and 524 precipitation covariability from observations and model simulations bear some notable 525 similarities. We now know that a negative aerosol-precipitation interaction more likely 526 occurs in the deep convective cloud system in the 7-SEAS region. More model runs are 527 needed to further pinpoint different aerosol effects on precipitation and quantify their

relative contributions. Over the N-ROI however, although aerosol-stratus interaction can still be active, due to the generally non-precipitating nature of stratus clouds, it is harder to observe aerosol-precipitation covariability there. However it is noteworthy that although precipitation change is less certain, the aerosol associated cloud and radiation changes are as significant over the N-ROI as over the S-ROI.

533 4) There remain uncertain factors that can influence the aerosol-precipitation interactions. 534 For example, although the negative aerosol and precipitation covariability were observed 535 for both high and low cloud liquid water path conditions, moist conditions seem to 536 enhance the precipitation reductions that are attributable to aerosol increases. Moreover, 537 in the vicinity of the western tropical Pacific, the large-scale dynamics over the 7-SEAS 538 region is very strong. It is still unknown but intriguing that how the large-scale aerosol 539 and precipitation covariability would be at different Madden-Julian Oscillation (MJO) 540 phases, a dominant rainfall feature in this region. Moreover, because aerosol emissions 541 usually occur in the pre-monsoon seasons in this area, it is also worthwhile exploring 542 whether anomalous aerosol loadings would impact large-scale dynamic fields through its 543 radiative forcing and, subsequently, affect monsoon rainfall. For example, does the 544 aerosol elevated heat pump (EHP) effect significantly impact precipitation in this region? 545 5) For deep convective clouds in the S-ROI, theoretically, aerosol microphysical effects 546 lead to smaller cloud particle sizes, delay warm rain precipitation processes and 547 invigorate deep convection (Koren et al., 2005; Rosenfeld et al., 2008). The aerosol semi-548 direct effect on other hand can reduce cloud by increasing tropospheric heating in the 549 clouds. Aerosol radiative forcing also suppresses convective clouds by the increased 550 environmental stability in respond to aerosol absorption (Cook and Highwood, 2003).

551 Rosenfeld et al. (2008) suggest an aerosol concentration saturation point at which the 552 fractional contributions from aerosol radiative and microphysical effects will vary to limit 553 convective potential. From the shown evidence in this study, the net aerosol effect on 554 precipitation and cloud deep convection is more negative, in line with aerosol radiative 555 forcing described above but not the invigoration of deep convection as suggested by 556 aerosol microphysical effect described in Rosenfeld et al. (2008) and model simulations 557 from Lebo and Seinfeld (2011). More climate model runs with different aerosol effect in 558 place will help to elucidate the mechanisms better.

6) Data uncertainty could still be a significant issue for this study or similar ones,

560 particularly aerosol observations. It is still challenging for us to completely understand 561 the complicated climate systems over this region, particularly when cloud coverage is so 562 prevalent to prevent extensive aerosol observations. It is not yet possible for satellites to

retrieve aerosol optical properties within or beneath clouds yet, although the UV based

aerosol index is able to detect aerosol qualitatively above clouds. In this study we used

statistical techniques to minimize data uncertainties, for example, using monthly data

other than daily data and selecting larger aerosol domain than precipitation domain.

567 However, we cannot completely rule out data uncertainty issues to make the results more

568 conclusive. For example, the discrepancy between the TOMS AI vs. GPCP run and the

569 SeaWiFS AOT vs. TRMM run (Figure 6c vs. Figure 6e) could be partially because that

570 TOMS AI can observe aerosols above clouds but SeaWiFS AOT cannot.

571

572 Overall, the study provides us a big picture of the characterizations of the large-scale

573 covariability between aerosol, cloud and precipitation over the 7-SEAS region. More

- 574 systematic investigations will continue to explore more fundamental mechanisms that are
- 575 modulating the weather and climate systems in the region.
- 576

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- 587
- 588

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Data Source	Parameter	Coverage	Length (months)	Unit	
SeaWiFS	АОТ	1997.09-2010.12,	158	Unitless	
		2 missing months			
MODIS	АОТ	2002.07-2010.04	94	Unitless	
MODIS	СТР	2002.07-2010.04	94	hPa	
TRMM	Monthly total rain	1998.01-2009.12	144	mm/day	
3B43					
TRMM	Daily total rain	1998.01-2009.12	144	mm/day	
3B42					
TRMM	Monthly rain rate	1998.01-2009.12	144	mm/day	
3A12					
TRMM	Monthly latent	1998.01-2009.12	144	K/hour	
3A12	heating				
NOAA OI	SST	1981.12-2010.04	341	К	
GPCP	Precipitation	1979.01-2009.09	369	mm/day	
TOMS	AI	1978.11-1993.04;	237	Unitless	
		1996.08-2000.12			
AIRS	СО	2002.09-2011.08	108	1018	
				molecular/cm ²	
NCEP-DOE	OLR	1974.06-2010.04	431	W/m ²	
reanalysis II					
NCEP-DOE	LWP	1979.01-2011.07	391	kg/m ²	
reanalysis II					
ISCCP	Cloud Amount	1983.01-2007.12	300	Unitless	
CAM5	AOT, Precipitation		60	AOT-Unitless	
				Precipitation-	
				mm/day	
CERES	TOA SW Flux	2000.03-2010.12	130	W/m ²	

769 Table 1. The major satellite observed and model simulated datasets used in this770 study

- Table 2. The domain-averaged changes in the anomalies of aerosol, cloud,
- precipitation, radiation variables between high and low tercile months of aerosol

normalized anomalies. Negative changes are in **bold** and positive changes in *italic*.

	SeaWiFS	GPCP	TRMM	TRMM	TRMM	MODIS	NCEP	CERES	CERES	ISCCP
	AOT	Precip.	Precip.	Rain Rate	no-rain	СТР	OLR	TOA SW	TOA SW	Cloud
					Days			Clear-Sky	All-Sky	Amount
unit	unitless	mm/day	mm/day	mm/day	days	hPa	W/m ²	W/m ²	W/m ²	%
S-ROI	+0.13	-1.23	-1.53	+0.029	+4	+26.8	+4.41	+2.59	-4.20	-1.80
N-ROI	+0.089	-0.30	+0.31	-0.11	-1	+4.36	+3.21	+1.36	-0.41	-0.12

777

779 (a) RGB, 03/08/2009

(b) RGB, 10/05/2006



- Figure 1. (a) Terra MODIS RGB Images over the Northern ROI on March 8, 2009,
- showing a smoke plume was transported by westerly jet from western source
- 783 regions to above stratus clouds over Northern Vietnam and Northern South China
- Sea; (b) Aqua MODIS RGB image over the Southern ROI on October 5, 2006, showing
- biomass burning over Borneo island, Indonesia, and the smoke plume was
- transported by easterly jet from Borneo to Indian Ocean.

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Figure 2. Spatial pattern of seasonal aerosol and precipitation. (a) to (d) are the FebMar-Apr seasonal mean of (a) TOMS AI; (b) SeaWiFS AOT; (c) MODIS AOT; (d) AIRS
CO (10¹⁸ molecular/cm²). (e) to (h) are the Sep-Oct-Nov seasonal mean of (e) TOMS
AI; (f) SeaWiFS AOT; (g) MODIS AOT; (h) AIRS CO. The corresponding seasonal GPCP
precipitation are: (i) Feb-Mar-Apr; (j) Sep-Oct-Nov. The N-ROI and S-ROI were
defined in Figure 2(a) where yellow outlines for aerosol domains and red outlines

- defined in Figure 2(a) where yellow outlines for aerosol domains and red outlines
 for precipitation domains for the N-ROI (in dashed lines) and the S-ROI (in solid
- 804 lines) respectively.
- 805







816 Figure 4. High, middle and low cloud amounts in the northern and southern ROIs:

817 (a) high cloud; (b) middle cloud; (c) low cloud.



821 Figure 5. (a) Correlation between ENSO NINO3 index and GPCP precipitation

- anomalies; and (b) correlation between ENSO NINO3 index and TOMS AI anomalies.
- The white contours mark 95% confidence level on the correlation significance.





Aerosol Index; TP: TRMM Precipitation; GP: GPCP precipitation). Square boxes wereused to highlight the areas of interests.







Figure 8. Difference composites of total rain anomalies between high and low tercile months of CO normalized anomalies



856 Longitude Longitude Longitude S57 Figure 9. Model simulations from CAM5: Difference composites of total rain







868 Figure 10. Difference composites of CTP, Cloud Amount and OLR anomalies between high and low aerosol tercile months: (a) CTP over the N-ROI; (b) CTP over the S-ROI;



- the N-ROI, and (f) OLOR over the S-ROI.



Figure 11. Difference composites of Terra CERES retrieved TOA shortwave flux
anomalies between high and low aerosol tercile months in aerosol normalized
anomalies: (a) clear sky SW flux over N-ROI; (a) clear sky SW flux over S-ROI; (a) all-

sky SW flux over N-ROI; and (d) all-sky SW flux over S-ROI. The unit of the flux is
W/m².



887 Figure 12. Difference composites of precipitation anomalies between high and low aerosol tercile months over the S-ROI: (a) only for low LWP tercile months; (b) only for high LWP tercile months.



892 (a) TP (N-ROI, FMA) (b) All-sky SW Flux (N-ROI, FMA) (c) CTP (N-ROI, FMA)



902 Top Panel, and over the S-ROI domain between high and low aerosol normalized

903 anomalies tercile months when only data from boreal fall season (Sep-Oct-Nov) were

904 used in the Bottom Panel.