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1       **Disentangling the influence of local and remote anthropogenic aerosols on South**  
2                               **Asian Monsoon daily rainfall characteristics**

3       Deepti Singh<sup>\*,1,2</sup>, Massimo Bollasina<sup>3</sup>, Mingfang Ting<sup>2,4</sup>, Noah S. Diffenbaugh<sup>5,6</sup>

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5       <sup>1</sup> School of the Environment, Washington State University, Vancouver, WA, USA

6               <sup>2</sup> Lamont Doherty Earth Observatory, Columbia University, NY, USA

7                               <sup>3</sup>School of Geosciences, University of Edinburgh, UK

8       <sup>4</sup>Department of Earth and Environmental Sciences, Columbia University, NY, USA

9               <sup>5</sup>Department of Earth System Science, Stanford University, CA, USA

10       <sup>6</sup>Woods Institute for the Environment, Stanford University, CA, USA

11

12       \* corresponding author:

13       Deepti Singh

14       School of the Environment, Washington State University, Vancouver, WA, 98686, USA.

15       email: [deepti.singh@wsu.edu](mailto:deepti.singh@wsu.edu)

16       Phone: +1 650-468-8791

17

18       Keywords: South Asian Monsoon; anthropogenic aerosols; local and remote aerosols;

19       daily-scale precipitation

20 **Abstract (250 words):**

21 Wet and dry periods within the South Asian summer monsoon season can have acute  
22 societal impacts. Recent studies have identified changes in daily rainfall characteristics of  
23 the monsoon, but the underlying causes are poorly understood. In particular, although the  
24 dominant role of anthropogenic aerosols in shaping historical changes in seasonal-mean  
25 monsoon rainfall has been documented, their influence on daily-scale rainfall remains  
26 unconstrained. Using an ensemble of single-forcing climate simulations, we find that  
27 anthropogenic aerosols have a stronger influence on late-20<sup>th</sup> century changes in the  
28 frequency of wet events, dry events and rainless days, compared with other climate  
29 forcings. We also investigate the role of aerosol-cloud interactions (“indirect effects”) in  
30 the total aerosol response, and the contribution of aerosols emitted from South Asia  
31 versus from remote sources. Based on additional simulations with the GFDL-CM3  
32 climate model, we find that the simulated aerosol response over South Asia is largely  
33 associated with aerosol-indirect effects. In addition, local aerosols suppress wet-event  
34 frequency and enhance dry-event frequency over eastern-central India, where increases in  
35 aerosol loading are the largest. Remote aerosols cause a north-south dipole pattern of  
36 change in mean rainfall over India and fewer rainless days over western India. However,  
37 the overall spatial response of South Asian rainfall characteristics to total aerosol forcing  
38 is substantially influenced by the non-linear climate response to local and remote  
39 aerosols. Together, our results suggest that understanding the influence of different  
40 aerosol emissions trajectories on the regional climate dynamics is critical for effective  
41 climate-risk management in this populated, vulnerable region.

42

43 **1. Introduction**

44 Variations in the timing, spatial distribution and characteristics of the South Asian  
45 summer monsoon rainfall can affect the economy, agriculture, ecosystems, human health,  
46 and water resources of the world's most densely populated region (Gadgil & Kumar  
47 2006; Gadgil & Gadgil 2006). Subseasonal monsoon variability – which manifests as wet  
48 and dry periods – is a critical factor in determining monsoonal impacts via, for example,  
49 intense rainfall and droughts, which can adversely affect agricultural output and farmer  
50 livelihoods (Gornall et al. 2010). Numerous studies have documented changes in the  
51 historical subseasonal rainfall characteristics over India on a range of spatial scales,  
52 including changes in the frequency of wet and dry spells over different sub-regions  
53 (Guhathakurta & Rajeevan 2008; Dash et al. 2009; Rajeevan et al. 2010; Guhathakurta et  
54 al. 2011; Singh et al. 2014; Vinnarasi & Dhanya 2016; Krishnan et al. 2016; Roxy et al.  
55 2017).

56

57 On a global-scale, studies have found a strong anthropogenic contribution to the observed  
58 changes in daily rainfall extremes (Min et al. 2011; Fischer & Knutti 2015; Diffenbaugh  
59 et al. 2017). On a regional-scale, Lin et al. (2018) suggest that anthropogenic aerosols  
60 have had a substantial influence on the large-scale pattern of historical changes in  
61 extreme heavy rainfall events over Asia. However, the influence of individual  
62 anthropogenic forcings – including greenhouse gases (GHG) and anthropogenic aerosols  
63 – on these historical changes over South Asia have not been distinguished. Studies  
64 suggest that changes in aerosol forcing might have a stronger effect on precipitation than  
65 changes in GHG in coming decades, if the world progresses on a low GHG emissions

66 pathway (Lin et al. 2016). Rainfall extremes have different sensitivities to GHGs and  
67 anthropogenic aerosols (Lin et al. 2016), and different concentrations of aerosols can  
68 either enhance or inhibit rainfall (Rosenfeld et al. 2008; Koren et al. 2014; Fan et al.  
69 2013; Fan et al. 2016). Given that emissions of GHGs and aerosols will likely exhibit  
70 different pathways in the future (van Vuuren et al. 2011), it is important to understand  
71 whether and how changes of each individual forcing have influenced subseasonal rainfall  
72 events during the historical period.

73

74 Unlike GHGs, aerosols concentrations and their historical trends have large regional  
75 variations (Fig. 1). Anthropogenic aerosols from fossil fuel burning – particularly sulfate  
76 aerosols and black carbon – have increased rapidly throughout the late 20<sup>th</sup> century over  
77 South and East Asia (Fig. 1a-b). During the same period, aerosol loadings decreased over  
78 North America and Europe, following strict air-quality regulations (Smith et al. 2011;  
79 Granier et al. 2011; Lu et al. 2011). The increases in aerosol loading over Asia are  
80 associated with large negative radiative forcing over the region relative to the  
81 preindustrial period (e.g., Ramanathan et al. 2001; Bollasina et al. 2011). Simulations  
82 with the GFDL-CM3 model (Donner et al. 2011) suggest that over the second half of the  
83 20<sup>th</sup> century, the net radiative flux at the surface decreased by -6 to -15 W/m<sup>2</sup> and at the  
84 top of the atmosphere (TOA) decreased by -3 and -9 W/m<sup>2</sup>, with strongest values located  
85 over the areas of largest emissions (Fig. 1c-d). (Unfortunately, observational estimates of  
86 long-term radiative flux changes are unavailable and a comparison of these simulated  
87 changes with observations is not straight forward. One would have to rely on shorter  
88 periods and simulations with fixed SSTs to reduce the effects of internal variability.)

89  
90 Increases in anthropogenic aerosol emissions have played a dominant role in driving a  
91 shift to an earlier monsoon onset and a weakening of the seasonal rainfall since the 1950s  
92 (Ramanathan et al. 2005; Lau & Kim 2010; Bollasina et al. 2011; Turner & Annamalai  
93 2012; Bollasina et al. 2014; Salzmann et al. 2014; Li et al. 2015; Krishnan et al. 2016;  
94 Guo et al. 2016; Li et al. 2016). Aerosols from both local (i.e., within South Asia) and  
95 remote sources are important in shaping historical changes in seasonal rainfall, although  
96 their relative contributions are still uncertain (Bollasina et al. 2014; Guo et al. 2016).  
97 Recent observational evidence also suggests that natural and anthropogenic aerosols, can  
98 affect daily-scale rainfall events over South Asia, including dry spells (Vinoj et al. 2014;  
99 Dave et al. 2017). The relative influence of local and remote aerosols on historical  
100 changes in daily-scale rainfall events in the presence of other external climate forcings is  
101 yet to be examined.

102  
103 We therefore seek to better understand the influence of aerosols on mean and daily-scale  
104 rainfall characteristics (wet events, dry events, and rainless day frequency) over South  
105 Asia by addressing three main questions: (1) Do anthropogenic aerosols have a stronger  
106 influence than other external forcings on the spatial pattern of changes in daily rainfall  
107 characteristics during the peak monsoon season? (2) Is the overall aerosol response most  
108 strongly associated with direct radiative effects or aerosol-cloud interactions (“indirect  
109 effects”)? (3) How are these rainfall changes influenced by aerosol emissions from local  
110 and remote regions?

111

112 Our analysis primarily employs an ensemble of simulations conducted with the NOAA  
113 Geophysical Fluid Dynamics Laboratory CM3 (GFDL-CM3) coupled climate model.  
114 GFDL-CM3 has been previously used to identify key influences of anthropogenic  
115 aerosols in driving the overall weakening trend of the summer monsoon and its earlier  
116 onset during the second half of the 20<sup>th</sup> century (Bollasina et al. 2011; Bollasina et al.  
117 2013), which have subsequently been supported by analysis of multi-model ensembles  
118 (Li et al. 2015; Salzmann et al. 2014; Guo et al. 2015). In this study, we employ a set of  
119 GFDL-CM3 single-forcing experiments to test the influence of anthropogenic aerosols on  
120 daily-scale precipitation characteristics relative to GHGs and natural forcings. In  
121 addition, we use targeted experiments to understand the mechanisms by which aerosols  
122 influence these characteristics over South Asia (including the role of direct and indirect  
123 effects), and isolate the contribution of local aerosols from that of non-South Asian  
124 aerosols. To evaluate inter-model differences in the influence of forcings on historical  
125 changes, we also compare results from the GFDL-CM3 model with a subset of models  
126 from the Coupled Model Intercomparison Project (CMIP5) suite (Taylor et al. 2012).

127

## 128 **2. Data and Methods:**

129

### 130 *2.1. Observations*

131 We analyze two widely-used gridded rainfall datasets derived from rain-gauge  
132 observations: the India Meteorological Department (“IMD”) dataset, which contains  
133 gridded data at 1°x1° horizontal resolution from 1951 to present (Rajeevan et al. 2010),  
134 and the Asian Rainfall Highly-Resolved Observational Data Integration Towards

135 Evaluation of Water Resources (“APHRODITE”) dataset, which contains gridded data at  
136  $0.25^{\circ} \times 0.25^{\circ}$  horizontal resolution from 1951 to 2007 (Yatagai et al. 2012). These are the  
137 only available gridded, long-term, daily rainfall datasets for the region. While the IMD  
138 dataset is restricted to India, APHRODITE covers the entire Asian domain (Fig. S1).  
139 Long-term changes in rainfall characteristics over sub-regions of India show considerable  
140 differences between these datasets (Fig. S2).

141

## 142 *2.2. Climate model experiments*

143 We use a suite of ensemble experiments with the NOAA Geophysical Fluid Dynamics  
144 Laboratory (GFDL-CM3) global coupled chemistry-climate model with a  $2^{\circ} \times 2.5^{\circ}$   
145 horizontal resolution. This is one of the few global climate models to realistically  
146 simulate the observed climatological mean and daily characteristics of peak-monsoon  
147 season rainfall (Fig. S1; Sperber et al. 2013; Ashfaq et al. 2017). GFDL-CM3 simulates  
148 the observed climatological timing and spatial patterns of several summer monsoon  
149 seasonal rainfall characteristics with lower biases than most other CMIP5 models (Fig.  
150 S3; Ashfaq et al. 2017). The model also reasonably represents the overall observed  
151 pattern of changes in monsoon season rainfall characteristics though the finer-resolution  
152 observations have greater spatial heterogeneity (Fig. S2). GFDL-CM3 is also one of the  
153 few models to archive multi-member simulations of daily-scale climate under individual  
154 external forcings, which are needed to account for “internal” variability in decadal-scale  
155 changes (Salzmann & Cherian 2015).

156



157 The GFDL-CM3 simulations use the standard CMIP5 historical anthropogenic emissions  
158 (Lamarque et al. 2010). In addition to the direct radiative effects of aerosols, the model  
159 includes a physically-based representation of aerosol-cloud interactions (commonly  
160 referred to as the aerosol “indirect effects”) (Donner et al. 2011; Levy et al. 2013).  
161 Aerosol indirect effects are simulated for liquid clouds and are parameterized for  
162 stratiform cloud microphysics (Ming et al. 2007; Golaz et al. 2011; Levy et al. 2013). In  
163 GFDL-CM3, water soluble aerosols (i.e. sulfate, sea-salt and organic carbon) act as cloud  
164 condensation nuclei (CCN) following the parameterizations of Ming et al. (2006) and  
165 Ming et al. (2007). Black carbon is assumed to be insoluble. Anthropogenic sulfate  
166 aerosols, which are more efficient CCN than the other aerosol species, are the major  
167 driver of changes in CCN and, therefore, of aerosol indirect effects in the model (Levy et  
168 al. 2013). Further, aerosols are considered as prognostic variables, and sulfate and black  
169 carbon are internally mixed using a uniform mixing scheme for radiative transfer  
170 calculations (Persad et al. 2017). These aerosol species are, however, assumed to be  
171 externally mixed for the estimation of aerosol indirect effects in the stratiform cloud  
172 microphysics scheme (Salzmann et al. 2010). Dust concentrations show negligible  
173 changes during the 20<sup>th</sup> century as GFDL-CM3 dust emission changes are modulated  
174 only by modest variations in the wind speed (i.e., the model does not simulate dust  
175 emission changes associated with land use/land cover change) (Pu & Ginoux 2016). One  
176 limitation of the representation of aerosols that has implications for this study is that  
177 aerosols in GFDL-CM3 do not interact with deep convection (Donner et al. 2011), which  
178 is also a limitation of most other global climate models (Rotstayn et al. 2014). More  
179 information on the GFDL-CM3 model formulation can be found in Donner et al. (2011).

180

181 In this study, we use three sets of GFDL-CM3 ensemble experiments. The first set of  
182 simulations includes experiments that are part of the public CMIP5 archive. This set  
183 consists of a 5-member ensemble with all historical forcings (“ALL-Forcing”), and three  
184 3-member individual-forcing ensembles forced by changes in greenhouse gases (“GHG-  
185 Only”), anthropogenic aerosols (“Aerosol-Only”), and solar and volcanic activity  
186 (“Natural-Only”) in isolation. The individual members within each respective ensemble  
187 differ only in their initial conditions and, therefore, the spread between them results from  
188 internal variability in the presence of the forcing. An additional 600-year preindustrial  
189 control simulation (“PIcontrol”) with forcings fixed at preindustrial levels is used to  
190 quantify the range of unforced internal variability. This suite of experiments allows us to  
191 study the relative influence of individual forcings on simulated historical changes, while  
192 simultaneously considering internal variability and removing model differences that may  
193 confound interpretation in a multi-model framework (e.g., differences in model  
194 resolutions, parameterizations, and aerosol representations).

195

196 The second set of simulations includes an additional 3-member ensemble in which  
197 aerosols interact only with clouds but not with radiation (i.e., the aerosol direct effect is  
198 switched off; Levy et al. 2013). This ensemble is designed to isolate the role of aerosol  
199 indirect effects in driving the overall changes in the Aerosol-Only ensemble.

200

201 The third set of simulations includes two complementary 3-member ensembles, designed  
202 to examine the relative influence of local and remote aerosols. The first has varying

203 aerosol emissions over South Asia and constant preindustrial levels over all other regions  
204 (South Asian Aerosol Emissions). The second has varying aerosol emissions over all  
205 remote regions and constant preindustrial levels over South Asia (Remote Aerosol  
206 Emissions). In both cases, other external forcing factors are kept constant at preindustrial  
207 levels.

208

209 Given the GFDL-CM3 model's overall performance, the availability of multiple  
210 realizations of single-forcing experiments, and the availability of experiments that  
211 isolated the direct and indirect effects and the roles of local and remote aerosols, we  
212 determine that GFDL-CM3 is a unique tool for exploring the influence of aerosols on  
213 historical changes in the South Asian monsoon rainfall characteristics.

214

215 However, there are substantial uncertainties associated with the representation of aerosols  
216 and aerosol-cloud interactions in the current generation of climate models (Boucher et al.  
217 2013; Rotstayn et al. 2015), and GFDL-CM3 is known to have an overly-strong aerosol  
218 effect (Levy et al. 2013). Therefore, we complement our analysis with three other CMIP5  
219 models (Table 1). These models are selected based on the availability of multiple  
220 realizations with individual forcings (e.g., aerosols and GHGs) at a daily resolution. They  
221 have varying degrees of biases in representing the climatology, and changes in surface  
222 radiative forcing and precipitation over the Indian subcontinent. Figure S3 compares the  
223 simulated rainfall mean and variability in these models with the CMIP5 suite and their  
224 representation of historical trends in several rainfall characteristics. Among the 4 models  
225 analyzed in this study, GFDL-CM3 and CCSM4 have relatively low biases in

226 representing the monsoon precipitation and circulation characteristics, while CSIRO-  
227 MK3.6.0 and CanESM2 have larger biases (Fig. S3; Ashfaq et al. 2017).  
228  
229 Critically for our analysis, the 4 models have varying aerosol effective radiative forcings  
230 (Rotstayn et al. 2015) and representations of aerosol effects (Salzmann et al. 2014) (Table  
231 1). GFDL-CM3 and CSIRO-MK3.6.0 are amongst the few CMIP5 models that include  
232 aerosol indirect effects, whereas CanESM2 only includes one indirect effect (cloud-  
233 albedo) and CCSM4 does not include either (Salzmann et al. 2014). Further, GFDL-CM3  
234 has the strongest aerosol effective radiative forcing (ERF) of all CMIP5 models, followed  
235 by CSIRO-MK3.6.0 within our subset of models (Rotstayn et al. 2015). The aerosol  
236 ERFs of both GFDL-CM3 ( $-1.6 \text{ W/m}^2$ ) and CSIRO-MK3.6.0 ( $-1.4 \text{ W/m}^2$ ) are higher than  
237 the ERF estimates ( $-0.45$  to  $-0.93 \text{ W/m}^2$ ) derived from satellite observations (Boucher et  
238 al. 2013). Aerosol indirect effects are a major contributor to the aerosol ERF, and are  
239 particularly sensitive to the model's cloud tuning parameters, as demonstrated by Golaz  
240 et al. (2011) and Golaz et al. (2013) specifically for GFDL-CM3 (but also true for other  
241 models). These pervasive uncertainties in aerosols and their interactions with clouds have  
242 important implications for our understanding of the influence of aerosols on climate  
243 processes, including precipitation. Although we are able to conduct an initial  
244 quantification of the influence of these uncertainties on our results, that quantification is  
245 limited by the number of models that both incorporate such effects and have multiple  
246 realizations of individual-forcing simulations.  
247

248        *2.3. Characteristics of the daily rainfall distribution*

249        We focus our analysis on rainfall characteristics during the peak-monsoon (July-August)  
250        months for three main reasons. First, at this time, the monsoon is fully established over  
251        the Indian Subcontinent. Second, monsoonal rainfall and the occurrence of wet/dry  
252        events are highest during July-August (Pai et al. 2015; Rajeevan et al. 2010). Third, the  
253        peak months coincide with the growth period of the Kharif (“monsoon”) crops, meaning  
254        that identifying the drivers of monsoon changes during these months has direct  
255        implications for agriculture.

256

257        We analyze four metrics of the peak-season daily rainfall distribution: mean rainfall,  
258        frequency of rainless days, frequency of deficit rainfall events (dry events), and  
259        frequency of excess rainfall events (wet events). Following Salinger and Griffiths (2001),  
260        we define rainless days as days with rainfall  $<1$  mm/day. In accordance with previous  
261        studies (Annamalai & Slingo 2001; Mandke et al. 2007; Rajeevan et al. 2010; Singh et al.  
262        2014), we define wet and dry events based on daily rainfall anomalies exceeding a certain  
263        standardized threshold. Standardized rainfall anomalies are calculated based on the mean  
264        and standard deviation calculated for the baseline period (1951-1975). Protracted  
265        anomalies with consecutive days meeting this criterion are considered a single event. We  
266        use a threshold of  $\pm 0.68$  standard deviations ( $\sigma$ ), which approximates the 25<sup>th</sup>/75<sup>th</sup>  
267        percentile of a normal distribution. The wet event frequency is defined as the number of  
268        events with daily rainfall anomalies exceeding the  $+0.68\sigma$  threshold in a season, while the

269 dry event frequency is the number of events with daily rainfall anomalies exceeding the -  
270  $0.68\sigma$  threshold.

271

#### 272 *2.4. Statistical Analysis*

273 We examine long-term changes in these characteristics during the 1951-2000 period,  
274 when the South Asian monsoon rainfall underwent a noticeable linear decrease of  $\sim 10\%$   
275 (e.g., Bollasina et al. 2011; Turner & Annamalai 2012; Singh et al. 2014). This  
276 weakening occurred simultaneously with an increase in regional anthropogenic aerosol  
277 emissions, particularly of sulfates and black carbon, which increased by  $\sim 6$  times since  
278 the early 20<sup>th</sup> century (e.g., Ramanathan et al. 2001; Ramanathan et al. 2005; Lau & Kim  
279 2010), and a change in phase of the Pacific Decadal Oscillation (PDO) - a mode of  
280 multidecadal internal variability - from negative to positive (Salzmann & Cherian 2015).  
281 Note, however, that even if the processes driving such internal modes of variability are  
282 accurately simulated, the exact timing of particular historical transitions should not be  
283 expected to be reproduced in individual coupled climate model realizations. This period  
284 also aligns with the availability of the historical forcing simulations (which, according to  
285 the CMIP5 protocols, run through 2005).

286

287 We compute differences in rainfall characteristics between two 25-year periods (1951-  
288 1975) and (1976-2000), which equally divide this 50-year period. We use a non-  
289 parametric permutation test to quantify the significance of changes in the mean of the  
290 distribution of different rainfall characteristics between the two periods, at each grid point  
291 (Stanberry 2013). The permutation test involves calculating changes between these 25-

292 year periods by randomly reorganizing the original timeseries several times. The p-value  
293 of this test is the proportion of absolute changes from these resampled timeseries that  
294 exceed the absolute magnitude of change between these time periods in the original time  
295 series. This significance test makes no assumptions about the underlying distribution,  
296 thereby accommodating the non-normality of the distributions of the various rainfall  
297 characteristics. To account for internal variability in the model ensembles, we first  
298 calculate changes for each ensemble member, and then average the changes across the  
299 ensemble to calculate the “forced response” to each forcing factor. The robustness of the  
300 model results at each grid point is measured by the agreement on the direction and  
301 statistical significance across the changes in the individual ensemble members.

302

### 303 *2.5. Approach for identifying spatial similarity*

304 To provide a quantitative estimate of the relative influence of individual forcing factors in  
305 driving South Asian monsoon rainfall changes, we use the pairwise Pearson’s correlation  
306 method to assess the similarity between the spatial pattern of changes in the ALL-Forcing  
307 ensemble and those in each individual-forcing ensembles. Given the spatial  
308 inhomogeneity of the aerosol distribution, we calculate the pattern correlations for the  
309 region  $6^{\circ}$ - $32^{\circ}$ N,  $68^{\circ}$ - $90^{\circ}$ E (shown in Fig. 1c), which encompasses the area of strong  
310 increase in aerosol emissions and forcing (Fig. 1a-d). Additionally, this domain accounts  
311 for the competition between rainfall changes over land and nearby ocean, which  
312 ultimately represent two facets of the response of the overall coupled monsoon system.  
313 The pattern correlations are calculated between each ensemble member of the ALL-  
314 Forcing experiment (5 realizations) and each ensemble member of each individual-

315 forcing experiment (3 realizations for each forcing), yielding 15 correlation values for  
316 each pair of forcing experiments. We also report the spatial correlations between the  
317 ensemble-mean changes in the different forcing experiments.

318

## 319 *2.6. Quantifying the role of internal variability relative to “forced” changes*

320 Changes in rainfall characteristics could result from internal fluctuations of the climate  
321 system that are largely independent of any forced changes (i.e., “internal variability”). To  
322 quantify the range of changes that could arise from internal variability, we calculate the  
323 distribution of changes between all pairs of non-overlapping 25-year periods in the  
324 unforced 600-year GFDL-CM3 PIcontrol simulation. Next, we calculate the distribution  
325 of spatial correlations between the changes calculated in the PI simulation and those  
326 calculated from the 5 members of the ALL-Forcing ensemble (“ALL-PI distribution”).  
327 Then, we calculate the distribution of spatial correlations between the changes calculated  
328 in the 5 members of the ALL-Forcing ensemble and the 3 individual ensemble members  
329 for each single-forcing experiment (respectively). Finally, we use the Kolmogorov-  
330 Smirnov (“K-S”) test to quantify the significance of the difference between the ALL-PI  
331 distribution of correlations and the respective ALL-Forcing/single-forcing distributions of  
332 correlations. The p-value from the K-S test indicates the confidence with which we can  
333 reject the null hypothesis that the ALL-Forcing changes arose from internal variability  
334 alone. Rejection of the null-hypothesis with high confidence implies that the forced  
335 changes are outside of the range expected from internal variability. In contrast, the



336 inability to reject the null-hypothesis suggests that an influence of that individual forcing  
337 cannot be concluded.

338

### 339 **3. Results and Discussions**

340

#### 341 *3.1. Influence of individual forcings on daily rainfall characteristics*

342 For all four rainfall characteristics, the spatial pattern of changes in the ALL-Forcing  
343 ensemble mean shows the closest similarity to the Aerosol-Only ensemble, with the  
344 GHG-Only and Natural-Only ensembles exhibiting little correspondence (Fig. 2a). The  
345 spatial correlation between the ensemble mean ALL-Forcing and Aerosol-Only changes  
346 is weaker for mean rainfall (0.4,  $p$ -value $<0.05$ ) than for the daily rainfall characteristics,  
347 particularly for rainless day frequency (0.7,  $p$ -value $<0.05$ ) and dry event frequency (0.6,  
348  $p$ -value $<0.05$ ). In contrast, the spatial correlations between the ALL-Forcing and GHG-  
349 Only changes are significantly negative for all characteristics, suggesting a consistent  
350 opposing effect of aerosols and GHGs. Changes in the Natural-Only ensemble are  
351 uncorrelated with changes in the ALL-Forcing ensemble for all metrics, with the  
352 exception of a significantly negative correlation for rainless day frequency. These results  
353 are robust across the various ensemble members (Fig. 2b-e), though in the case of wet  
354 event frequency, the individual members have lower spatial correlations than the  
355 ensemble means, likely due to dampening of the internal variability.

356

357 Observations exhibit robust declines in mean peak-season rainfall over eastern-central  
358 India, and moderate increases over parts of western and northwestern India between the

359 1951-75 and 1976-2000 (Fig. S2a-b). Changes in mean peak-season rainfall in the ALL-  
360 Forcing ensemble display a coherent large-scale east-west dipole pattern across South  
361 Asia, largely similar to the observed pattern of changes, albeit of slightly weaker  
362 magnitude and with sub-regional biases (e.g., over the Western Ghats) (Fig. 3b). Mean  
363 rainfall in GFDL-CM3 decreases significantly by  $\sim 0.4\text{-}0.8$  mm/day over eastern-central  
364 India, the climatologically wetter sub-region of South Asia, but increases significantly by  
365  $\sim 0.3\text{-}0.5$  mm/day over northwestern India and Pakistan, the climatologically drier sub-  
366 region of South Asia (Fig. 3a-b). A very similar pattern, though of larger magnitude, is  
367 recognizable in the Aerosol-Only ensemble (Fig. 3c). In contrast, changes induced by  
368 GHGs are largely opposite to those induced by aerosol forcing, including a wetting of  
369 eastern-central India and a drying to the west (Fig. 3d). In the Natural-Only ensemble,  
370 rainfall is suppressed over the entire domain (Fig. 3e). This indicates that the overall  
371 ALL-Forcing response of mean peak-season rainfall in the GFDL-CM3 model is largely  
372 driven by aerosol forcing.

373

374 The simulated climatology of rainless day frequency (days with  $< 1$  mm/day) during the  
375 peak-monsoon season features the highest occurrence over northwestern India, Pakistan,  
376 and parts of peninsular India, and fewer than 6 days over the rest of the domain (Fig. 3f).  
377 Changes in rainless day frequency have considerable uncertainties in observations, with  
378 widespread increases in the IMD dataset and spatially variable and contrasting trends in  
379 the APRHODITE dataset (Fig. S2d-e). The pattern of changes in rainless day frequency  
380 in the GFDL-CM3 ALL-Forcing ensemble is more consistent with the declines over  
381 northwestern India and slight increases over eastern-central India in the APHRODITE

382 dataset (Fig. S2e-f). The simulated pattern of changes in rainless day frequency closely  
383 resembles the corresponding changes in mean rainfall (Fig. 3b,g). The most robust  
384 anomalies in rainless days occur over the climatologically dry northwestern sub-region,  
385 where both the ALL-Forcing and Aerosol-Only ensembles simulate decreases of up to 3-  
386 4 days (Fig. 3g-h). In contrast, there are relatively small and largely insignificant changes  
387 in the frequency of rainless days over eastern-central India in both the ALL-Forcing and  
388 Aerosol-Only ensembles (Fig. 3g-h). The GHG-Only and Natural-Only ensembles show  
389 an overall weak increase in rainless day frequency across much of the domain, with the  
390 only significant changes being increases over parts of the western sub-domain in the  
391 GHG-Only ensemble (Fig. 3i-j).

392

393 Together, these results suggest that the simulated increases in mean rainfall over the  
394 northwestern sector of the domain in the ALL-Forcing and Aerosol-Only ensembles (Fig.  
395 3b) are driven at least in part by aerosol-induced increases in the number of days with  
396 rainfall (converse of rainless day frequency), while the strong declines in mean rainfall  
397 over central India in the ALL-Forcing and Aerosol-Only ensembles (Fig. 3b-c) are driven  
398 primarily by decreases in the intensity of rainfall (average precipitation on rainy days)  
399 rather than decreases in the number of days with rainfall (Fig. S2i). This decline in  
400 rainfall intensity over much of central India simulated in the ALL-Forcing ensemble is  
401 consistent with IMD and APHRODITE, though there are slight differences in the location  
402 of peak changes (Fig. S2g-h).

403

404 The highest climatological frequency of wet and dry events generally occurs over the  
405 areas that experience the heaviest mean climatological rainfall (Fig. 4a,f). Eastern-central  
406 India typically averages ~5-7 wet events and ~6-8 dry events during the peak-monsoon  
407 season in GFDL-CM3 (Fig. 4a,f). Observed changes in wet and dry event frequency in  
408 the two observational datasets are broadly consistent. However, there are discrepancies in  
409 the magnitude and spatial pattern of changes, again emphasizing the observational  
410 uncertainties in these measures of rainfall extremes (Fig. S2i-o). The ALL-Forcing  
411 ensemble broadly simulates the observed patterns of reduced wet event frequency in  
412 eastern central India and increased dry event frequency in the same region, albeit with  
413 less heterogeneity. Wet event frequency significantly decreases by over 0.6 events/season  
414 – and dry event frequency significantly increases by over 0.8 events/season – over  
415 eastern-central India during the 1976-2000 period relative to the 1951-1975 period in  
416 GFDL-CM3 (Fig. 4b,g). In addition, the ALL-Forcing ensemble shows significant  
417 increases in wet event frequency of approximately the same magnitude over the  
418 climatologically drier regions of Pakistan and northwestern India (Fig. 4g). Among the  
419 single-forcing ensembles, this ALL-Forcing dipole pattern of changes in dry and wet  
420 event frequency is only present in the Aerosol-Only ensemble (Fig. 4). In contrast, the  
421 GHG-Only ensemble exhibits changes that are largely opposite to the Aerosol-Only  
422 changes, with wet event frequency increasing significantly across northern and eastern  
423 India and decreasing significantly over peninsular India (Fig. 4d,i). The Natural-Only  
424 ensemble shows decreases in wet event frequency and increases in dry event frequency  
425 across most of the domain, but the changes are of smaller magnitude and less  
426 significance, and bear little similarity to those in the ALL-Forcing ensemble. Along with

427 the decline in mean and increase in rainless day frequency, these changes in wet and dry  
428 event frequency in the Natural-Only ensemble are consistent with the presence of an  
429 active volcanic eruption period, which has an overall weakening effect on the monsoon  
430 (Ning et al. 2017).

431

432 The similarity of the magnitude and spatial pattern of historical changes in mean rainfall,  
433 rainless day frequency, and wet/dry event frequency between the ALL-Forcing and  
434 Aerosol-Only ensembles (Fig. 2-4) indicates a strong and robust aerosol imprint on the  
435 characteristics of daily rainfall over South Asia in the GFDL-CM3 model. To determine  
436 whether the forced changes are statistically distinguishable from those associated with  
437 internal climate variability, we compare the spatial correlations between the ALL-Forcing  
438 and single-forcing ensembles with the spatial correlations between the ALL-Forcing  
439 ensemble and the PIcontrol simulation (*see Section 2.5*). (Changes in the 600-year  
440 PIcontrol simulation are calculated for all pairs of non-overlapping 25-year periods.). For  
441 all characteristics (Fig. 2b-e), the PIcontrol correlations are small (25<sup>th</sup>-75<sup>th</sup> percentile of  
442 the correlation distribution  $<\pm 0.2$ ) and centered around zero, suggesting a relatively  
443 minor role of internal variability in generating the ALL-Forcing patterns of changes. For  
444 the rainfall characteristics that exhibit the strongest influence of aerosol forcings (mean  
445 rainfall, rainless day frequency, and dry event frequency), the distribution of correlations  
446 between the ALL-Forcing and Aerosol-Only patterns are significantly different ( $p$ -value  
447  $< 0.01$ ) from the patterns arising from unforced variability. A similar result, although  
448 slightly less significant, holds for changes in wet event frequency ( $p$ -value=0.09). For all  
449 characteristics, correlations between the ALL-Forcing and Natural-Only ensembles are

450 statistically indistinguishable from correlations between the ALL-Forcing ensemble and  
451 the PIcontrol simulation, suggesting that the Natural-Only changes are within the range  
452 of internal climate variability.

453

454 Together, these results provide strong evidence for the predominant role of anthropogenic  
455 aerosols in driving the ALL-Forcing pattern of changes in multiple daily rainfall  
456 characteristics in the GFDL-CM3 model. In addition, they highlight the greater similarity  
457 between the ALL-Forcing and Aerosol-Only pattern of changes for rainless day  
458 frequency and dry event frequency than for the seasonal mean, indicating that aerosols  
459 likely have a larger influence on low- to moderate-intensity rainfall events.

460

### 461 *3.2. The role of aerosol-cloud interactions*

462

463 To understand the mechanisms by which aerosols influence daily rainfall characteristics,  
464 we separate the contribution of aerosol-cloud interactions (i.e. indirect effects) from the  
465 overall aerosol effect simulated in the Aerosol-Only ensemble. To do so, we make use of  
466 an additional 3-member ensemble experiment (Aerosol Indirect-Only) in which aerosols  
467 do not interact with radiation (i.e., the aerosol direct effect is not active; *see section 2.1*),  
468 allowing us to isolate the role of aerosol indirect effects (Fig. 5). The similarity in the  
469 spatial pattern and magnitude of mean rainfall changes between the Aerosol-Only and the  
470 Aerosol Indirect-Only ensembles (Fig. 5a) – in particular the dipole pattern of drying  
471 over eastern-central India and the wetting over southern India and the western regions –  
472 suggests that aerosol indirect effects play a predominant role in shaping the response of

473 peak-monsoon rainfall to aerosol forcing. Similarly, the correspondence between changes  
474 in net radiation at the top of the atmosphere in both ensembles also confirm the  
475 predominant role of aerosol indirect effects in driving the overall Aerosol-Only changes,  
476 whilst not precluding a secondary role of aerosol direct effects (Fig. S4). These aerosol  
477 indirect effects are largely associated with changes in anthropogenic sulfate  
478 concentrations as black carbon are not treated as CCN in the model. The stronger and  
479 more expansive rainfall suppression seen in the Aerosol Indirect-Only ensemble  
480 compared with the Aerosol-Only ensemble indicates that aerosol direct effects partly  
481 offset the changes induced by the aerosol indirect effects.

482

483 In addition, aerosol indirect effects appear to be important for the aerosol-forced changes  
484 in dry and wet event frequency (Fig. 5e-h). Both the Aerosol-Only and the Aerosol  
485 Indirect-Only ensembles display key similarities in the above patterns of change,  
486 including increased frequency of dry events and decreased frequency of wet events over  
487 eastern-central India, and changes of opposite sign but smaller magnitude over the rest of  
488 the domain (Fig. 5e-h). However, changes in the frequency of rainless days in these two  
489 ensembles are less similar (Fig. 5c-d). The Aerosol Indirect-Only ensemble largely shows  
490 increases in rainless day frequency (i.e decrease in rainy days) over much of South Asia  
491 in contrast to the robust decreases simulated in the Aerosol-Only ensemble. While the  
492 robust increases in the Aerosol Indirect-Only ensemble occur mainly over eastern India,  
493 the decreases in rainless day frequency (i.e increase in rainy days) in the Aerosol-Only  
494 ensemble are strongest and most significant over northwestern India and Pakistan. This  
495 dissimilarity indicates that indirect effects do not influence the overall Aerosol-Only

496 change in occurrence of rainy days, but instead have a stronger influence on the intensity  
497 of rainfall events.

498

499 The potential for the aerosol indirect-effects to influence the frequency of wet and dry  
500 event is rooted in the aerosol modulation of cloud and rainfall processes. Some  
501 observations and cloud-resolving modeling studies support the idea that aerosols could  
502 invigorate convection, particularly in deep convective clouds, which could support the  
503 intensification of rainfall events (Rosenfeld et al. 2008; Fan et al. 2012; Koren et al.  
504 2014; Fan et al. 2016 and references therein). However, such convection-aerosol  
505 interactions are not included in coarse-resolution models including GFDL-CM3 (Donner  
506 et al. 2011; Rotstayn et al. 2015). A contrasting hypothesis is that enhanced aerosol  
507 concentrations suppress rainfall by increasing the number of CCN. Higher number of  
508 CCN lead to reduced cloud droplet size and smaller droplets are likely to reduce the  
509 efficiency of rainfall formation in the clouds to produce less heavy rain and, to a lesser  
510 extent, increase rainless day frequency (e.g., Ramanathan et al. 2001; Forster et al. 2007;  
511 Rosenfeld et al. 2008; Fan et al. 2012; Li et al. 2016). Consistent with the latter  
512 hypothesis, we find a decline in precipitation intensity across central India in the Aerosol  
513 Indirect-Only ensemble, inferred from the relatively large decreases in mean rainfall and  
514 small changes in rainless day frequency (Fig. 5b,d). This decline in overall precipitation  
515 intensity manifests as a decrease in the frequency of wet events and an increase in the  
516 frequency of dry events (Fig. 5f,h). The largest decreases in rainfall intensity and  
517 associated changes in wet and dry event frequency are located over eastern-central India,  
518 where aerosol loading underwent the strongest increase (Fig. 1b).



519

520 Aerosol-forced rainfall variations are also associated with large-scale dynamic and  
521 thermodynamic changes, which are very similar to those driven by aerosol indirect  
522 effects alone (Fig. 6). The strong surface cooling ( $>1.5\text{K}$ ) in the northwest of the domain,  
523 predominantly driven by aerosol indirect effects, is associated with a reduction of the  
524 meridional pressure gradient over the Indian Subcontinent and, correspondingly, with a  
525 weakening of the low-level circulation (Fig. 6a-c,d-f). The west-east dipole pattern in  
526 mean rainfall and in wet event frequency corresponds closely to changes in moisture  
527 availability, likely associated with these aerosol-driven circulation changes (Fig. 6d-f). In  
528 addition, the patterns of changes in wet and dry event frequency in the Aerosol-Only  
529 ensemble largely follow the patterns of changes in vertical stability<sup>1</sup> associated with  
530 temperature and moisture changes (Fig. 6g-i). Increases in dry event frequency and  
531 decreases in wet event frequency are accompanied by increased vertical stability over  
532 eastern-central India in both ensembles.

533

534 These results suggest an important role of aerosol-cloud interactions in driving the total  
535 aerosol response of wet and dry event frequency over parts of central and eastern India,  
536 the region with largest aerosol increases. Direct radiative effects – through interactions  
537 with the circulation – also appear to be important in shaping changes in daily and mean

---

<sup>1</sup> Vertical stability is calculated by computing the vertical difference in equivalent potential temperature (EPT) between two layers close to the surface (925 hPa minus 2m), calculated using the expression suggested in Bolton (1980). By definition, EPT accounts for both changes in temperature and humidity as the moist parcel of air ascends and its vapor condenses, releasing latent heat. Warmer low-level temperatures and higher low-level humidity tend to increase instability.

538 rainfall characteristics over northwestern India and Pakistan, where aerosol loading  
539 shows little change. Aerosol direct effects appear to have contrasting effects on  
540 temperature and precipitation over this part of the domain, given the enhanced rainfall  
541 and weaker cooling in the Aerosol-Only ensemble relative to the Aerosol Indirect-Only  
542 ensemble (Fig. 5a-b, 6b-c). Although the response of daily-scale rainfall characteristics to  
543 individual forcing factors can be explained in part by seasonal-mean changes in the large-  
544 scale atmospheric environment, a wide range of processes acting across spatial and  
545 temporal scales affect the monsoon rainfall and its daily-scale characteristics (e.g., Hurley  
546 & Boos 2014; Krishnamurthy & Shukla 2008; Rajeevan et al. 2010). Further research is  
547 needed to improve current understanding of the multitude of processes and features (e.g.,  
548 monsoon depressions) governing sub-seasonal-scale rainfall variability of the region,  
549 including their modulation by individual external forcing factors.

550

### 551 *3.3. Impact of aerosols from local and remote sources*

552

553 In addition to aerosols emitted from sources within the domain, rising aerosol emissions  
554 over other parts of the world, particularly East Asia (Fig. 1a), have the potential to  
555 modulate the circulation and rainfall over South Asia (Bollasina et al. 2014; Guo et al.  
556 2016). Here, we examine the relative importance of South Asian aerosol emissions  
557 compared to aerosols over the rest of the world (“Remote Aerosol Emissions”) in shaping  
558 the regional response of rainfall characteristics to aerosols (Fig. 7). Note that changes in  
559 non-South Asian aerosols are mostly due to East Asian aerosol emissions, as emissions  
560 over North America and Europe show only small changes between the two historical

561 periods considered in this analysis (not shown; see Fig. 1 in Bollasina et al. (2014)). It is  
562 also worth noting that, despite multiple aerosol transport and removal processes, the  
563 largest AOD changes are closely located over areas with the largest variations in aerosol  
564 emissions.

565

566 In the simulations with aerosols varying only over South Asia (“South Asian  
567 Emissions”), there is widespread decline in rainfall across much of India, with the largest  
568 changes over northern and eastern India (Fig. 7a), which is also the sub-region of largest  
569 forcing (Fig. 1b-d). This sub-region also experiences the strongest decreases in wet event  
570 frequency and increases in dry event frequency (Fig. 7g,j), similar to the total aerosol  
571 response (Fig. 4c,h). In contrast, in the Remote Aerosol Emissions simulations, seasonal  
572 rainfall exhibits a north-south dipole pattern of changes with increases over the northern  
573 India and decreases over peninsular India (Fig. 7b). There are few robust and coherent  
574 changes in the frequency of wet and dry events in these simulations that only include the  
575 remote aerosol emissions (Fig. 7h, k). However, rainless day frequency decreases over  
576 much of India, especially over the western half of the domain, closely resembling the  
577 total aerosol response (Fig. 3h,7e). These results highlight the importance and distinct  
578 roles of aerosols from both sources in shaping the Aerosol-Only response in seasonal and  
579 daily rainfall characteristics.

580

581 The non-linearity in the combined response to local and remote aerosols for these  
582 characteristics is notable, which likely results from feedbacks within the coupled climate  
583 system. To quantify the degree of nonlinearity between the climate impacts of emissions

584 from local and remote sources, we calculate the difference between the ensemble-mean  
585 changes in the Aerosol-Only simulations and the arithmetic sum of changes in the South  
586 Asian and Remote Aerosol Emissions simulations (Fig. 7c,f,i,l). For mean rainfall, the  
587 Aerosol-Only changes (Fig. 3c) cannot be explained by the response to either South  
588 Asian or remote emissions, but closely resemble the pattern of nonlinear changes (Fig.  
589 7a-c). While the response to remote aerosol emissions largely explains the Aerosol-Only  
590 changes in rainless day frequency over India (Fig. 3h), the overall changes over Pakistan  
591 resemble the nonlinear effects of combined remote and South Asian aerosols (Fig. 7d-f).  
592 For both the mean rainfall and rainless day frequency, the pattern of the nonlinear term  
593 suggests that the presence of local emissions acts to shift the region of wetting westward  
594 over northwestern India and Pakistan (Fig. 7c,f). In contrast, the Aerosol-Only changes in  
595 wet and dry event frequency over eastern-central India are mainly driven by local aerosol  
596 emissions (Fig. 7g, j). Similar to changes in other characteristics, the main nonlinear  
597 effect of combined local and remote emissions is the relative wetting over the  
598 northwestern sub-region of the domain, which acts to increase wet event frequency (Fig.  
599 7i,l).

600

601 The substantial nonlinearity in the rainfall response to local and remote aerosols are  
602 associated with non-additive responses of the monsoon circulation and other  
603 thermodynamic variables (Fig. 8). The strong cooling in the Aerosol-Only ensemble over  
604 the northern and northwestern sub-regions of the domain results largely from the  
605 influence of remote aerosols (Fig. 8a-c). Although surface temperature is unaltered in the  
606 simulations with varying South Asian aerosol emissions alone, historical changes in

607 remote aerosol emissions cause a cooling over the northwestern sub-region, which is  
608 further strongly amplified by the combined presence of local and remote aerosol forcings.  
609 The occurrence of cooling over regions of enhanced precipitation is suggestive of the  
610 modulation of temperature by feedbacks with precipitation rather than due to direct  
611 radiative forcing.

612

613 Consistent with the relatively large effect of remote aerosols on surface temperature, the  
614 weakening of the 850-mb circulation in the Aerosol-Only ensemble appears to occur  
615 largely as a response to remote aerosol emissions (Fig. 8d-f). In the Remote Aerosol  
616 Emissions simulations, the anomalous easterly winds are shifted relatively south, leading  
617 to drier conditions over peninsular India (Fig. 6e, 8e). In addition, southerly flow  
618 associated with the anticyclonic circulation over the eastern part of the domain, leads to  
619 wetter conditions over northern India (Fig. 7b). In comparison, the effect of local  
620 emissions on the circulation is relatively small (Fig. 8d). However, combined local and  
621 remote aerosols have the non-linear effect of amplifying the cooling in the northwest that  
622 is dominated by remote emissions, resulting in a sharper decrease in the meridional  
623 temperature and pressure gradients. The nonlinear circulation response includes an  
624 anomalous anticyclonic circulation over central India and an anomalous cyclonic  
625 circulation over the Arabian Sea (Fig. 8f). These anomalies shift the remotely-forced  
626 anomalous easterlies over peninsular India northwards, causing drying over central India,  
627 and convergence and enhanced rainfall in the southern and western sub-regions of the  
628 domain (Fig. 7c). Consistent with the relatively large nonlinear effects on surface

629 temperature and low-level humidity, the pattern of Aerosol-Only changes in vertical  
630 stability also closely resemble the pattern of the nonlinear term (Fig. 8g-i).  
631  
632 The closer similarity of anomalies in surface temperature and lower-tropospheric  
633 circulation between the Aerosol-Only ensemble and the Remote Aerosol Emissions  
634 ensemble (compared with the South Asia Aerosol Emissions ensemble) indicates a  
635 stronger impact of remote aerosols on the regional circulation and thermodynamics.  
636 However, the substantial magnitude of the nonlinear temperature and circulation  
637 anomalies resulting from the presence of local and remote aerosols suggest that the total  
638 Aerosol-Only response in rainfall characteristics is strongly modulated by the non-linear  
639 climate response to regional aerosol emissions. These non-linearities could be associated  
640 with local feedbacks (such as between temperature and precipitation) and/or large-scale  
641 feedbacks (such as that of the coupled Asian Monsoon circulations). Given the  
642 comparably higher emission rates over East Asia (Fig. 1), and the large-scale coupling  
643 between the South Asian and East Asian monsoons (Day et al. 2015; Ha et al. 2017;  
644 Preethi et al. 2017), nonlinearity in the climate response to local and remote aerosols  
645 could arise via circulation-precipitation feedbacks between these monsoon systems. For  
646 instance, deep tropospheric heating anomalies associated with precipitation increases in  
647 one region could influence the upper-tropospheric circulation, which can propagate  
648 downstream via, for example, Rossby waves and in turn affect climate in remote regions.  
649 Another factor contributing to the non-linearity could be the non-additive effects of  
650 different aerosols species over different regions. Such non-linearity was reported by Guo  
651 et al. (2016), in particular in the response to black carbon. Given the feedbacks within the

652 climate system, the role of different aerosol species in creating these non-linearities are  
653 not straightforward to identify. The magnitude of the nonlinearities highlights the need  
654 for simulations similar to those of Guo et al. (2016) to distinguish the effects of  
655 individual anthropogenic aerosol species – particularly separating absorbing and  
656 scattering aerosols – and allow for a deeper investigation of the sources of these  
657 nonlinearities.

658

#### 659 *3.4. Comparison of the influence of aerosols in CMIP5 models*

660 Among the available CMIP5 models, there is disagreement about the influence of  
661 aerosols on the ALL-Forcing trends (Fig. 9). CSIRO-MK3.6.0, the only other model  
662 (along with GFDL-CM3) that includes both aerosol indirect effects, consistently exhibits  
663 a stronger influence of aerosols on the ALL-Forcing changes in all 4 rainfall  
664 characteristics (relative to other individual forcings; Fig. 9a). In contrast, CanESM2,  
665 which only includes the cloud-albedo effect, exhibits negative correlations between the  
666 ALL-Forcing and Aerosol-Only changes, and stronger positive correlations between the  
667 ALL-Forcing and Natural-Only changes for mean rainfall, dry event frequency and  
668 rainless day frequency (relative to the ALL-Forcing and GHG-Only changes; Fig. 9b).  
669 CCSM4, which does not include either indirect effect, does not show substantial and  
670 consistent similarities between the ALL-Forcing pattern of changes and either individual  
671 forcing pattern of changes (Fig. 9c).

672

673 These inter-model differences can be understood in terms of their treatment of aerosol-  
674 cloud interactions. Aerosol-cloud interactions are known to be critical for representing

675 historical patterns and trends in surface temperature and precipitation (e.g., Wilcox et al.  
676 2013; Golaz et al. 2013; Levy et al. 2013; Ekman 2014; Wang 2015; Lin et al. 2018). The  
677 two models that include a comprehensive treatment of aerosol effects – GFDL-CM3 and  
678 CSIRO-MK3.6.0 – agree on the relatively larger influence of aerosols on historical  
679 changes in these rainfall characteristics (Fig. 9). A recent analysis by Lin et al. (2018)  
680 using CMIP5 models (grouped according to their complexity of aerosol treatment) also  
681 shows disagreements on the sign of aerosol-induced changes in extreme heavy rainfall  
682 over Asia between models that include only direct effects (i.e CCSM4) and those that  
683 include both indirect effects (i.e GFDL-CM3 and CSIRO-MK3.6.0). They also find that  
684 models that include only the first direct effect (i.e CanESM2) differ considerably from  
685 the models that include explicit representations of the cloud-lifetime effect.

686

687 Although our analyses of a limited set of models preclude a quantification of the full  
688 range of uncertainties, they do highlight the importance of the representation of aerosol  
689 effects. While there are still uncertainties in the magnitude of direct radiative effects,  
690 aerosol-cloud interactions still represent the largest source of uncertainty in climate  
691 models (Boucher et al. 2013). Even among the models that include explicit  
692 representations of aerosol-cloud interactions, the representation of various effects is  
693 incomplete, and several important processes are not accounted for in coarse resolution  
694 models. For instance, the coarse resolution global climate models cannot simulate the  
695 effect of increased CCN on mixing and entrainment (Salzmann et al. 2010) – which has  
696 contrasting effects on cloud lifetimes compared to the effect of increased CCN alone  
697 (e.g., Ackerman et al. 2004; Xue & Feingold 2006; Zhou & Penner 2017) – potentially



698 leading to an overestimation of aerosol indirect effects (Levy et al. 2013). The  
699 interactions between aerosols and deep convection, which can have substantial and  
700 potentially contrasting effects on the precipitation distribution in certain regions (Fan et  
701 al. 2016), are also not represented in most models (Rotstayn et al. 2015). Further  
702 analyses, including additional experiments with cloud-resolving models, can improve the  
703 simulation of these effects, and thereby help to elucidate the exact mechanisms by which  
704 aerosols can influence daily rainfall events.

705

#### 706 **4. Concluding Remarks**

707 In addition to the total seasonal rainfall, changes in such daily-scale rainfall events have  
708 implications for agricultural and hydrological systems. For instance, more multi-day  
709 anomalously low rainfall events or rainless days during the peak growing season can  
710 affect the rain-fed agricultural systems prevalent across much of India, which depend on  
711 timely and reliable rainfall. Further, multi-day anomalously heavy rainfall events can also  
712 damage crops, increase the flooding risk in poorly planned urban systems, strain water  
713 management infrastructure, and affect ground water storage (Field et al., 2012, Mondal  
714 and Mujumdar, 2015).

715 Using a suite of ensemble experiments with the GFDL-CM3 climate model, we examine  
716 the influence of anthropogenic aerosols and other external climate forcings on peak-  
717 season (July-August) mean and daily rainfall characteristics over South Asia. Our results  
718 suggest a predominant role of anthropogenic aerosols in weakening mean rainfall over  
719 India, largely associated with aerosol-cloud interactions, which play a fundamental role  
720 during July and August when aerosols and clouds are collocated over the region and

721 when increases in aerosol loading are the strongest in the GFDL-CM3 model. These  
722 findings extend previous work on rainfall changes during the summer (June-September)  
723 monsoon over India (Bollasina et al. 2011; Salzmman et al. 2014; Li et al. 2016; Zhang &  
724 Li 2016).

725

726 We note three new insights about the drivers of change in daily-scale rainfall events  
727 provided by our study:

- 728 • Anthropogenic aerosols have a stronger influence on historical changes in wet event  
729 frequency, dry event frequency, and rainless days frequency, relative to other external  
730 forcings. This influence of anthropogenic aerosols on the dry event and rainless days  
731 frequency is larger than their influence on the seasonal mean rainfall.
- 732 • Aerosol indirect effects have a substantial influence on changes in dry event and wet  
733 event frequency over the areas with the strongest aerosol loading. Despite striking  
734 similarity in the response of the large-scale circulation and thermodynamics to  
735 changes driven by aerosol indirect effects, direct effects appear to be important in  
736 shaping the overall aerosol response of wet events and rainless days over the  
737 climatologically drier parts of the subcontinent.
- 738 • South Asian aerosols lead to an increase in dry event frequency and decrease in wet  
739 frequency, while remote aerosols increase the number of rainy days in the  
740 northwestern sub-region. However, the overall response of several rainfall  
741 characteristics and their atmospheric environment to aerosols is governed to a large  
742 extent by the nonlinear climatic effects of local and remote aerosols.

743 While recent literature examining daily-scale rainfall has primarily focused on the  
744 response to GHG forcing, the potential for anthropogenic aerosols to also play an  
745 important role has been mostly overlooked. A few studies have examined the effect of  
746 different future aerosol trajectories on certain metrics of rainfall extremes at global and  
747 regional scales (Sillmann et al. 2013; Lin et al. 2016; Lin et al. 2018). Our study offers  
748 new insights by distinguishing the influence of historical aerosol and GHG emissions on  
749 daily-scale rainfall characteristics over the historical period, including the roles of direct  
750 and indirect aerosol effects, and the roles of local and remote aerosol emissions. Given  
751 recent findings on the importance of aerosols for the region’s climate, understanding the  
752 mechanisms by which aerosols can influence rainfall variability on daily timescales  
753 warrants further attention. Further insights will require an expanded archive of single-  
754 forcing climate model ensembles, additional simulations with cloud-resolving models,  
755 and further development of long-term observations of daily-scale rainfall and of aerosol  
756 processes.

757

758 We acknowledge a number of caveats in our analysis. First, our analysis of a limited  
759 number of climate models does not account for the large inter-model differences in the  
760 monsoon response to climate forcings (e.g., Sperber et al. 2013; Sharmila et al. 2015).  
761 Additional multi-member ensembles of individual forcing simulations using other climate  
762 models that include advanced representations of aerosol physical and chemical processes  
763 are required to quantify the full range of uncertainties in the role of historical aerosol  
764 emissions. Second, the limited ensemble size might not capture the full range of internal  
765 climate variability, which clearly has a substantial influence on the direction and

766 magnitude of historical trends (Deser et al. 2012; Kay et al. 2015). The large spread in the  
767 PIcontrol ensemble highlights the potential for internal variability to have a substantial  
768 influence on historical trends (Salzmann et al. 2014; Salzmann & Cherian 2015).  
769 Although we have compared our single-forcing results with the range of internal  
770 variability in the GFDL-CM3 model using the long preindustrial control run, larger  
771 ensembles of individual forcing experiments will help to more robustly ascertain this role  
772 of internal variability, especially for higher-frequency rainfall variability (Diffenbaugh et  
773 al. 2017). Third, the relatively coarse spatial resolution of the model might miss  
774 important fine-scale processes that shape the response of such extreme rainfall to forcings  
775 (Diffenbaugh et al. 2005; Ashfaq et al. 2009). Fourth, we have not accounted for the  
776 influence of changes in natural aerosols such as continental dust, which might modulate  
777 short-term rainfall over central India (Vinoj et al. 2014). The CMIP6 experiments (Eyring  
778 et al. 2016) could address some of these caveats through the availability of higher  
779 resolution models that have improved atmospheric chemistry and physics, as well as  
780 larger ensemble sizes.

781

782 Along with previous studies highlighting the impact of local and remote anthropogenic  
783 aerosols on seasonal-scale rainfall (Ramanathan et al. 2005; Wang et al. 2009; Bollasina  
784 et al. 2011; Guo et al. 2016), our study highlights potential mechanisms by which they  
785 can impact daily rainfall characteristics of the South Asian summer monsoon. Given  
786 current efforts to manage both global GHG increases and regional air quality, our results  
787 have important implications for near-term climate adaptation. Although aerosols are  
788 projected to decrease globally in the late 21<sup>st</sup> century (Moss et al. 2010; Vuuren et al.

789 2011), near-term local increases over South Asia could continue to negatively impact  
790 societal systems that are strongly dependent on reliable rainfall. In addition, aerosol  
791 changes in remote regions (such as East Asia), which can induce circulation changes  
792 comparable to or larger than those generated by local aerosols (Bollasina et al. 2014;  
793 Chakraborty et al. 2014), may also contribute to future rainfall changes over South Asia.  
794 Further, our analyses of GHG-Only simulations, as well as many previous studies (e.g.,  
795 Ashfaq et al. 2009; Stowasser et al. 2009; Krishnan et al. 2016; Kito 2017), suggest that  
796 continued GHG increases could also result in considerably altered rainfall patterns,  
797 particularly when coupled with decreases in aerosol emissions. Considering the influence  
798 of different aerosol emissions trajectories over South and East Asia on the regional  
799 climate dynamics is therefore critical for effective climate risk management in this highly  
800 populated, highly vulnerable region.

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1114

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1117 **Competing Financial Interest Statement**

1118 All authors declare no competing financial interests.

1119

1120 **Author Contributions:**

1121 D.S., M.B., and N.S.D. conceived the study. All authors designed the analysis. M.B.

1122 provided the data. D.S. performed the analysis and all authors wrote the manuscript.

1123

1124 **Tables**

1125 Table 1: Details of Climate Model Experiments used in the study, partly based on

1126 Salzmann et al. (2014) and Ekman (2014).

| Model         | Ensemble Members<br>(All-Forcing, Individual-Forcing) | Aerosol Effects   | Reference                           |
|---------------|---|---|-------------------------------------|
| GFDL-CM3      | 5,3   | Direct and indirect effects (cloud-albedo and cloud-lifetime) | (Donner et al. 2011)                |
| CSIRO-MK3.6.0 | 10,10   | Direct and indirect effects (cloud-albedo and cloud-lifetime) | (Rotstayn et al. 2012)              |
| CCSM4         | 3,3   | Direct effects only   | (Gent et al. 2011)                  |
| CanESM2       | 5,5   | Direct effects and indirect effects (cloud-albedo only)       | (Ma et al. 2010; Arora et al. 2011) |

1127

1128

1129 **Figure Captions:**

1130

1131 **Figure 1. Historical Emissions and Forcing Changes:** Changes in mean peak-monsoon

1132 season (July-August) (a) anthropogenic aerosol emissions, (b) aerosol optical depth

1133 (AOD) in the ALL-Forcing simulation, (c) net surface radiation, and (d) net top of the

1134 atmosphere (TOA) radiation, between 1951-1975 and 1976-2000, based on the GFDL-

1135 CM3 model. Historical emissions that are input to the model are from the CMIP5

1136 standard gridded dataset (Lamarque et al. (2010)). The black rectangle in panel (c)

1137 encompasses the domain used in the analysis of spatial correlations (6-32°N, 68-90°E).

1138

1139 **Figure 2. Influence of Internal Variability and Individual Forcings on ALL-forcing**

1140 **Changes:** (a) Spatial correlations between ensemble mean changes in the ALL-Forcing

1141 and individual forcing experiments. (b-e) Range of spatial correlations between changes

1142 in all ensemble members of the ALL-Forcing simulations with all ensemble members of

1143 the preindustrial (PI; grey), Aerosol-Only (Aero; blue), GHG-Only (GHG; red), and  
1144 Natural-Only (Nat; green) simulations, over South Asia (box in Fig. 1c). Numbers below  
1145 each boxplot are the p-values for the Kolmogorov Smirnov test between the distribution  
1146 of spatial correlations of ALL-Forcing with PIcontrol changes and ALL-Forcing with  
1147 individual forcing changes. In the text, we refer to all p-values below 0.05 as statistically  
1148 significant.

1149

1150 **Figure 3. Peak-season Rainfall Characteristics:** Climatological mean (1951-1975) and  
1151 ensemble mean changes (1976-2000 relative to 1951-1975) in (a-e) mean rainfall and (f-  
1152 j) frequency of rainless days (precipitation < 1mm/day) during the peak-season (July-  
1153 August) in the ALL-Forcing, Aerosol-Only, GHG-Only, and Natural-Only simulations.  
1154 Grey dots in panels indicate that all ensemble members agree on the direction of change.  
1155 Black dots indicate that all ensemble members agree on the direction of change and the  
1156 change in at least one member is significant at the 5% level.

1157

1158 **Figure 4. Dry and Wet Event Characteristics:** As in Figure 3, but for (a-e) dry event  
1159 frequency, and (f-j) wet event frequency. Here, dry and wet events refer to individual or  
1160 multiple consecutive day events with rainfall anomalies exceeding  $\pm 0.68 \sigma$ .

1161

1162 **Figure 5. Role of Aerosol Indirect Effects:** Ensemble mean changes (1976-2000  
1163 relative to 1951-1975) in July-August (a-b) mean rainfall, (c-d) rainless day frequency,  
1164 (e-f) dry event frequency, and (g-h) wet event frequency in the Aerosol-Only simulations  
1165 and Aerosol Indirect-Only simulations.



1166

1167 **Figure 6. Influence of Indirect Effects on Thermodynamics and Circulation:**

1168 Climatological mean (1951-1975) and ensemble mean changes (1976-2000 relative to  
1169 1951-1975) in the peak-season (July-August) (a-c) surface temperature (K, shading) and  
1170 sea-level pressure (hPa, contours), (d-f) 850mb circulation and moisture (arrows  
1171 represent winds and shading represents moisture), and (g-i) vertical stability (K;  
1172 measured as the difference in equivalent potential temperature between 925mb and 2m)  
1173 in the Aerosol-Only and Aerosol-Indirect Only simulations.

1174

1175 **Figure 7. Local and Remote Aerosols Impacts on Rainfall:** Ensemble mean changes

1176 (1976-2000 relative to 1951-1975) in rainfall characteristics in (left column) simulations  
1177 with anthropogenic aerosols increasing over South Asia and rest of the world emissions  
1178 fixed at preindustrial levels ("South Asian Aerosol Emissions"), and (middle column)  
1179 simulations with anthropogenic aerosols increasing over the rest of the world and aerosol  
1180 emissions over South Asia fixed at preindustrial levels ("Remote Aerosol Emissions").  
1181 (Right column) Difference between the changes in the total aerosol experiment and the  
1182 arithmetic sum of changes in the local and remote aerosol experiment, referred to as  
1183 nonlinear effects.

1184

1185 **Figure 8. Local and Remote Aerosol Impacts on Thermodynamics and Circulation:**

1186 As in Fig. 7 but for (a-c) surface temperature and sea-level pressure, (d-f) 850mb  
1187 circulation and moisture availability, and (g-i) vertical stability.

1188

1189 **Figure 9. Uncertainties in the Effects of Individual Forcings on ALL-forcing**  
1190 **Changes:** Spatial correlations between ensemble mean changes in the ALL-Forcing and  
1191 individual forcing experiments in three additional CMIP5 models with multiple ensemble  
1192 members for the individual forcing experiments. Grey numbers indicate correlations that  
1193 are insignificant at the 5% level.