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1	Disentangling the influence of local and remote anthropogenic aerosols on South
2	Asian Monsoon daily rainfall characteristics
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4	
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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 anthropogenic aerosols have a stronger influence on late-20th century changes in the 27 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^{th} 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^{th} century, the net radiative flux at the surface decreased by -6 to -15 W/m ² and at the
84	top of the atmosphere (TOA) decreased by -3 and -9 W/m^2 , 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.)

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	wat to be amound

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 th 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),

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°x0.25° 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

143

142 2.2. Climate model experiments

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°x2.5° 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 th 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).

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 W/m²) and CSIRO-MK3.6.0 (-1.4 W/m²) are higher than 237 the ERF estimates (-0.45 to -0.93 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 ± 0.68 standard deviations (σ), which approximates the $25^{th}/75^{th}$
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 0.68σ 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

emissions, particularly of sulfates and black carbon, which increased by ~6 times since

the early 20th 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

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

also aligns with the availability of the historical forcing simulations (which, according to

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°-32°N, 68°-90°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-

forcing experiment (3 realizations for each forcing), yielding 15 correlation values for
each pair of forcing experiments. We also report the spatial correlations between the
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

inability to reject the null-hypothesis suggests that an influence of that individual forcingcannot 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 ~0.4-0.8 mm/day over eastern-central 364 India, the climatologically wetter sub-region of South Asia, but increases significantly by 365 $\sim 0.3-0.5$ 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 $\leq 1 \text{ mm/day}$) 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 \sim 5-7 wet events and \sim 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

the decline in mean and increase in rainless day frequency, these changes in wet and dry
event frequency in the Natural-Only ensemble are consistent with the presence of an
active volcanic eruption period, which has an overall weakening effect on the monsoon
(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 PIcontrol simulation are calculated for all pairs of non-overlapping 25-year periods.). For 440 all characteristics (Fig. 2b-e), the PIcontrol correlations are small (25th-75th percentile of 441 442 the correlation distribution $\leq \pm 0.2$) 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 intensity497 of rainfall events.

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).

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 (>1.5K) 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 stability¹ 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

¹ 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 lowlevel 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.
549 550	including their modulation by individual external forcing factors.
	<i>3.3. Impact of aerosols from local and remote sources</i>
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550 551	
550 551 552	3.3. Impact of aerosols from local and remote sources
550 551 552 553	3.3. Impact of aerosols from local and remote sources In addition to aerosols emitted from sources within the domain, rising aerosol emissions
550 551 552 553 554	3.3. Impact of aerosols from local and remote sources In addition to aerosols emitted from sources within the domain, rising aerosol emissions over other parts of the world, particularly East Asia (Fig. 1a), have the potential to
550 551 552 553 554 555	3.3. Impact of aerosols from local and remote sources In addition to aerosols emitted from sources within the domain, rising aerosol emissions over other parts of the world, particularly East Asia (Fig. 1a), have the potential to modulate the circulation and rainfall over South Asia (Bollasina et al. 2014; Guo et al.
550 551 552 553 554 555 556	3.3. Impact of aerosols from local and remote sources In addition to aerosols emitted from sources within the domain, rising aerosol emissions over other parts of the world, particularly East Asia (Fig. 1a), have the potential to modulate the circulation and rainfall over South Asia (Bollasina et al. 2014; Guo et al. 2016). Here, we examine the relative importance of South Asian aerosol emissions
550 551 552 553 554 555 556 556	3.3. Impact of aerosols from local and remote sources In addition to aerosols emitted from sources within the domain, rising aerosol emissions over other parts of the world, particularly East Asia (Fig. 1a), have the potential to modulate the circulation and rainfall over South Asia (Bollasina et al. 2014; Guo et al. 2016). Here, we examine the relative importance of South Asian aerosol emissions compared to aerosols over the rest of the world ("Remote Aerosol Emissions") in shaping

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 tranport 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 coherrent 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,1).

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

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.

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 peninuslar 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

temperature and low-level humidity, the pattern of Aerosol-Only changes in verticalstability 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 similarties 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.

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

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

al. 2016), are also not represented in most models (Rotstayn et al. 2015). Further

analyses, including additional experiments with cloud-resolving models, can improve the

simulation of these effects, and thereby help to elucidate the exact mechanisms by which

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

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

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; Salzmann 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.

While recent literature examining daily-scale rainfall has primarily focused on the 743 744 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

Along with previous studies highlighting the impact of local and remote anthropogenic aerosols on seasonal-scale rainfall (Ramanathan et al. 2005; Wang et al. 2009; Bollasina et al. 2011; Guo et al. 2016), our study highlights potential mechanisms by which they can impact daily rainfall characteristics of the South Asian summer monsoon. Given current efforts to manage both global GHG increases and regional air quality, our results have important implications for near-term climate adaptation. Although aerosols are projected to decrease globally in the late 21st 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; Kitoh 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.
801	

References:

805	Ackerman, A.S. et al., 2004. The impact of humidity above stratiform clouds on indirect
806	aerosol climate forcing. Nature, 432, p.1014. Available at:
807	http://dx.doi.org/10.1038/nature03174.
808	Annamalai, H. & Slingo, J.M., 2001. Active / break cycles: diagnosis of the intraseasonal
809	variability of the Asian Summer Monsoon. Climate Dynamics, 18(1), pp.85–102.
810	Available at: http://dx.doi.org/10.1007/s003820100161.
811	Arora, V.K. et al., 2011. Carbon emission limits required to satisfy future representative
812	concentration pathways of greenhouse gases. Geophysical Research Letters, 38(5).
813	Available at: https://doi.org/10.1029/2010GL046270.
814	Ashfaq, M. et al., 2017. Sources of errors in the simulation of south Asian summer
815	monsoon in the CMIP5 GCMs. Climate Dynamics, 49(1-2), pp.193-223. Available
816	at: http://link.springer.com/10.1007/s00382-016-3337-7 [Accessed June 20, 2017].
817	Ashfaq, M. et al., 2009. Suppression of south Asian summer monsoon precipitation in the
818	21st century. Geophys. Res. Lett., 36(1), p.L01704. Available at:
819	http://dx.doi.org/10.1029/2008GL036500.
820	Bollasina, M.A. et al., 2014. Contribution of local and remote anthropogenic aerosols to
821	the twentieth century weakening of the South Asian Monsoon. Geophysical
822	<i>Research Letters</i> , 41(2), pp.680–687.
823	Bollasina, M.A., Ming, Y. & Ramaswamy, V., 2011. Anthropogenic Aerosols and the
824	Weakening of the South Asian Summer Monsoon. Science, 334(6055), pp.502–505.
825	Available at: http://www.sciencemag.org/content/334/6055/502.abstract.
826	Bollasina, M.A., Ming, Y. & Ramaswamy, V., 2013. Earlier onset of the Indian monsoon
827	in the late twentieth century: The role of anthropogenic aerosols. Geophysical
828	<i>Research Letters</i> , 40(14), pp.3715–3720.
829	Bolton, D., 1980. The Computation of Equivalent Potential Temperature. <i>Monthly</i>
830	Weather Review, 108(7), pp.1046–1053. Available at: https://doi.org/10.1175/1520-
831	0493(1980)108%3C1046:TCOEPT%3E2.0.CO.
832	Boucher, O. et al., 2013. Clouds and Aerosols. In: Climate Change 2013: The Physical
833	Science Basis. Contribution of Working Group I to the Fifth Assessment Report of
834	the Intergovernmental Panel on Climate Change. In T. F Stocker et al., eds.
835	Cambridge University Press, Cambridge, United Kingdom and New York, NY,
836	USA.
837	Chakraborty, A., Nanjundiah, R.S. & Srinivasan, J., 2014. Local and remote impacts of
838	direct aerosol forcing on Asian monsoon. International Journal of Climatology,
839	34(6), pp.2108–2121. Available at: http://doi.wiley.com/10.1002/joc.3826.
840	Dash, S.K. et al., 2009. Changes in the characteristics of rain events in India. J. Geophys.
841	Res., 114(D10), p.D10109. Available at: http://dx.doi.org/10.1029/2008JD010572.
842	Dave, P., Bhushan, M. & Venkataraman, C., 2017. Aerosols cause intraseasonal short-
843	term suppression of Indian monsoon rainfall. Scientific Reports, 7(1), p.17347.
844	Available at: https://doi.org/10.1038/s41598-017-17599-1.
845	Day, J.A., Fung, I. & Risi, C., 2015. Coupling of South and East Asian Monsoon
846	Precipitation in July–August. Journal of Climate, 28(11), pp.4330–4356. Available

847 at: https://doi.org/10.1175/JCLI-D-14-00393.1. 848 Deser, C. et al., 2012. Communication of the role of natural variability in future North 849 American climate. *Nature Clim. Change*, 2(11), pp.775–779. 850 Diffenbaugh, N.S. et al., 2005. Fine-scale processes regulate the response of extreme 851 events to global climate change. Proc Natl Acad Sci USA, 102(44), p.8. 852 Diffenbaugh, N.S. et al., 2017. Quantifying the influence of global warming on 853 unprecedented extreme climate events. Proceedings of the National Academy of 854 Sciences, 114(19), pp.4881–4886. Available at: 855 http://www.pnas.org/content/114/19/4881.abstract. 856 Donner, L.J. et al., 2011. The Dynamical Core, Physical Parameterizations, and Basic 857 Simulation Characteristics of the Atmospheric Component AM3 of the GFDL 858 Global Coupled Model CM3. Journal of Climate, 24(13), pp.3484-3519. Available 859 at: http://dx.doi.org/10.1175/2011JCLI3955.1. 860 Ekman, A.M.L., 2014. Do sophisticated parameterizations of aerosol-cloud interactions 861 in CMIP5 models improve the representation of recent observed temperature 862 trends?, (119), pp.817–832. 863 Eyring, V. et al., 2016. Overview of the Coupled Model Intercomparison Project Phase 6 864 (CMIP6) experimental design and organization., pp.1937-1958. 865 Fan, J. et al., 2012. Aerosol impacts on clouds and precipitation in eastern China: Results 866 from bin and bulk microphysics. Journal of Geophysical Research: Atmospheres, 867 117(D16). Available at: https://doi.org/10.1029/2011JD016537. 868 Fan, J. et al., 2013. Microphysical effects determine macrophysical response for aerosol 869 impacts on deep convective clouds. Proceedings of the National Academy of 870 Sciences, 110(48), pp.E4581–E4590. Available at: 871 http://www.pnas.org/cgi/doi/10.1073/pnas.1316830110. 872 Fan, J. et al., 2016. Review of Aerosol-Cloud Interactions: Mechanisms, Significance, 873 and Challenges. Journal of the Atmospheric Sciences, 73(11), pp.4221–4252. 874 Available at: http://journals.ametsoc.org/doi/10.1175/JAS-D-16-0037.1. 875 Fischer, E.M. & Knutti, R., 2015. Anthropogenic contribution to global occurrence of 876 heavy-precipitation and high-temperature extremes. Nature Clim. Change, 5(6), 877 pp.560–564. 878 Forster, P., et al., 2007. Changes in Atmospheric Constituents and in Radiative Forcing. 879 In: Climate Change 2007: The Physical Science Basis. Contribution of Working 880 Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate 881 Change. In M. T. and H. L. M. [Solomon, S., D. Qin, M. Manning, Z. Chen, M. 882 Marquis, K.B. Averyt, ed. Cambridge University Press, Cambridge, United 883 Kingdom and New York, NY, USA. 884 Gadgil, S. & Gadgil, S., 2006. The Indian Monsoon, GDP and Agriculture, Available at: 885 http://www.epw.in/special-articles/indian-monsoon-gdp-and-agriculture.html. 886 Gadgil, S. & Kumar, K.R., 2006. The Asian Monsoon - agriculture and economy. In B. 887 Wang, ed. The Asian Monsoon. Berlin Heidelberg New York: Springer/Praxis 888 Publishing Co... 889 Gent, P.R. et al., 2011. The Community Climate System Model Version 4. Journal of 890 *Climate*, 24(19), pp.4973–4991. Available at: 891 https://doi.org/10.1175/2011JCLI4083.1. 892 Golaz, J.C. et al., 2011. Sensitivity of the aerosol indirect effect to subgrid variability in

893 the cloud parameterization of the GFDL atmosphere general circulation model 894 AM3. Journal of Climate, 24(13), pp.3145–3160. 895 Golaz, J.C., Golaz, J.C. & Levy, H., 2013. Cloud tuning in a coupled climate model: 896 Impact on 20th century warming. Geophysical Research Letters, 40(10), pp.2246-897 2251. 898 Gornall, J. et al., 2010. Implications of climate change for agricultural productivity in the 899 early twenty-first century. Philos Trans R Soc Lond B Biol Sci, 365(1554), pp.2973– 900 2989. Available at: http://www.ncbi.nlm.nih.gov/pubmed/20713397. 901 Granier, C. et al., 2011. Evolution of anthropogenic and biomass burning emissions of air 902 pollutants at global and regional scales during the 1980-2010 period. Climatic 903 Change, 109(1), p.163. Available at: https://doi.org/10.1007/s10584-011-0154-1. 904 Guhathakurta, P. & Rajeevan, M., 2008. Trends in the rainfall pattern over India. 905 International Journal of Climatology, 28(11), pp.1453–1469. 906 Guhathakurta, P., Sreejith, O.P. & Menon, P.A., 2011. Impact of climate change on 907 extreme rainfall events and flood risk in India. Journal of Earth System Science, 908 120(3), p.359. Available at: https://doi.org/10.1007/s12040-011-0082-5. 909 Guo, L., Turner, A.G. & Highwood, E.J., 2015. Impacts of 20th century aerosol 910 emissions on the South Asian monsoon in the CMIP5 models. Atmospheric 911 Chemistry and Physics, 15(11), pp.6367–6378. Available at: http://www.atmos-912 chem-phys.net/15/6367/2015/. 913 Guo, L., Turner, A.G. & Highwood, E.J., 2016. Local and Remote Impacts of Aerosol 914 Species on Indian Summer Monsoon Rainfall in a GCM. Journal of Climate, 29(19), 915 pp.6937–6955. Available at: https://doi.org/10.1175/JCLI-D-15-0728.1. 916 Ha, K.-J. et al., 2017. Linkages between the South and East Asian summer monsoons: a 917 review and revisit. Climate Dynamics. Available at: https://doi.org/10.1007/s00382-918 017-3773-z. 919 Hurley, J. V & Boos, W.R., 2014. A global climatology of monsoon low-pressure 920 systems., (April), pp.1049–1064. 921 Kay, J.E. et al., 2015. The Community Earth System Model (CESM) Large Ensemble 922 Project: A Community Resource for Studying Climate Change in the Presence of 923 Internal Climate Variability. Bulletin of the American Meteorological Society, 96(8), 924 pp.1333-1349. Available at: http://journals.ametsoc.org/doi/10.1175/BAMS-D-13-925 00255.1. 926 Kitoh, A., 2017. The Asian Monsoon and its Future Change in Climate Models : A 927 Review., 95. 928 Koren, I., Dagan, G. & Altaratz, O., 2014. From aerosol-limited to invigoration of warm 929 convective clouds., 344(6188), pp.1143–1147. 930 Krishnamurthy, V. & Shukla, J., 2008. Seasonal persistence and propagation of 931 intraseasonal patterns over the Indian summer monsoon region. *Climate Dynamics*, 932 30, pp.353-369. 933 Krishnan, R. et al., 2016. Deciphering the desiccation trend of the South Asian monsoon 934 hydroclimate in a warming world. *Climate Dynamics*, 47(3–4), pp.1007–1027. 935 Available at: http://link.springer.com/10.1007/s00382-015-2886-5. 936 Lamarque, J.-F. et al., 2010. Historical (1850–2000) gridded anthropogenic and biomass 937 burning emissions of reactive gases and aerosols: methodology and application. 938 Atmos. Chem. Phys., 10(15), pp.7017-7039.

939 Lau, W.K.M. & Kim, K.-M., 2010. Fingerprinting the impacts of aerosols on long-term 940 trends of the Indian summer monsoon regional rainfall. Geophysical Research 941 Letters, 37(16), p.L16705. Available at: http://dx.doi.org/10.1029/2010GL043255. 942 Levy, H. et al., 2013. The roles of aerosol direct and indirect effects in past and future 943 climate change. Journal of Geophysical Research: Atmospheres, 118(10), pp.4521– 944 4532. Available at: http://dx.doi.org/10.1002/jgrd.50192. 945 Li, X. et al., 2015. Mechanisms of Asian Summer Monsoon Changes in Response to 946 Anthropogenic Forcing in CMIP5 Models. Journal of Climate, 28(10), pp.4107-947 4125. Available at: https://doi.org/10.1175/JCLI-D-14-00559.1. 948 Li, Z. et al., 2016. Aerosol and monsoon climate interactions over Asia. Reviews of 949 Geophysics, 54(4), pp.866–929. Available at: 950 http://dx.doi.org/10.1002/2015RG000500. 951 Lin, L. et al., 2018. Changes in Extreme Rainfall Over India and China Attributed to 952 Regional Aerosol-Cloud Interaction During the Late 20th Century Rapid 953 Industrialization. *Geophysical Research Letters*, pp.1–9. Available at: 954 http://doi.wiley.com/10.1029/2018GL078308. 955 Lin, L. et al., 2016. Sensitivity of precipitation extremes to radiative forcing of 956 greenhouse gases and aerosols. Geophysical Research Letters, 43(18), pp.9860-957 9868. Available at: http://dx.doi.org/10.1002/2016GL070869. 958 Lu, Z., Zhang, Q. & Streets, D.G., 2011. Sulfur dioxide and primary carbonaceous 959 aerosol emissions in China and India, 1996–2010. Atmos. Chem. Phys., 11(18), 960 pp.9839–9864. Available at: https://www.atmos-chem-phys.net/11/9839/2011/. 961 Ma, X., von Salzen, K. & Cole, J., 2010. Constraints on interactions between aerosols 962 and clouds on a global scale from a combination of MODIS-CERES satellite data 963 and climate simulations. Atmos. Chem. Phys., 10(20), pp.9851-9861. Available at: 964 https://www.atmos-chem-phys.net/10/9851/2010/. 965 Mandke, S.K. et al., 2007. Simulated changes in active/break spells during the Indian 966 summer monsoon due to enhanced CO2 concentrations: assessment from selected 967 coupled atmosphere-ocean global climate models. International Journal of 968 *Climatology*, 27(7), pp.837–859. Available at: http://dx.doi.org/10.1002/joc.1440. 969 Min, S.-K. et al., 2011. Human contribution to more-intense precipitation extremes. 970 Nature, 470(7334), pp.378-381. 971 Ming, Y. et al., 2006. A New Parameterization of Cloud Droplet Activation Applicable to 972 General Circulation Models. Journal of the Atmospheric Sciences, 63(4), pp.1348-973 1356. Available at: https://doi.org/10.1175/JAS3686.1. 974 Ming, Y. et al., 2007. Modeling the Interactions between Aerosols and Liquid Water 975 Clouds with a Self-Consistent Cloud Scheme in a General Circulation Model. 976 Journal of the Atmospheric Sciences, 64(4), pp.1189–1209. Available at: 977 https://doi.org/10.1175/JAS3874.1. 978 Moss, R.H. et al., 2010. The next generation of scenarios for climate change research and 979 assessment. Nature, 463(7282), pp.747–756. Available at: 980 http://dx.doi.org/10.1038/nature08823. 981 Ning, L., Liu, J. & Sun, W., 2017. Influences of volcano eruptions on Asian Summer 982 Monsoon over the last 110 years. Scientific Reports, 7, pp.3–8. Available at: 983 http://dx.doi.org/10.1038/srep42626. 984 Pai, D.S. et al., 2015. Analysis of the daily rainfall events over India using a new long

985	period (1901–2010) high resolution ($0.25^{\circ} \times 0.25^{\circ}$) gridded rainfall data set. <i>Climate</i>
986	<i>Dynamics</i> , 45(3–4), pp.755–776. Available at:
987	http://link.springer.com/10.1007/s00382-014-2307-1.
988	Persad, G.G. et al., 2017. Competing Atmospheric and Surface-Driven Impacts of
989	Absorbing Aerosols on the East Asian Summertime Climate. Journal of Climate,
990	30(22), pp.8929–8949. Available at: http://journals.ametsoc.org/doi/10.1175/JCLI-
991	D-16-0860.1.
992	Preethi, B. et al., 2017. Variability and teleconnections of South and East Asian summer
993	monsoons in present and future projections of CMIP5 climate models. Asia-Pacific
994	Journal of Atmospheric Sciences, 53(2), pp.305–325. Available at:
995	https://doi.org/10.1007/s13143-017-0034-3.
996	Pu, B. & Ginoux, P., 2016. The impact of the Pacific Decadal Oscillation on springtime
997	dust activity in Syria. Atmos. Chem. Phys., 16(21), pp.13431–13448.
998	Rajeevan, M., Gadgil, S. & Bhate, J., 2010. Active and break spells of the Indian summer
999	monsoon. Journal of Earth System Science, 119(3), pp.229–247. Available at:
1000	http://dx.doi.org/10.1007/s12040-010-0019-4.
1001	Ramanathan, V. et al., 2001. Aerosols, Climate, and the Hydrological Cycle. Science,
1002	294(5549), pp.2119–2124. Available at:
1003	http://www.sciencemag.org/content/294/5549/2119.abstract.
1004	Ramanathan, V. et al., 2005. Atmospheric brown clouds: Impacts on South Asian climate
1005	and hydrological cycle. Proceedings of the National Academy of Sciences of the
1006	United States of America, 102(15), pp.5326–5333. Available at:
1007	http://www.pnas.org/content/102/15/5326.abstract.
1008	Rosenfeld, D. et al., 2008. Flood or Drought: How Do Aerosols Affect Precipitation?
1009	Science, 321(5894), p.1309 LP-1313. Available at:
1010	http://science.sciencemag.org/content/321/5894/1309.abstract.
1011	Rotstayn, L.D. et al., 2012. Aerosol- and greenhouse gas-induced changes in summer
1012	rainfall and circulation in the Australasian region: a study using single-forcing
1013	climate simulations. Atmos. Chem. Phys., 12(14), pp.6377-6404. Available at:
1014	https://www.atmos-chem-phys.net/12/6377/2012/.
1015	Rotstayn, L.D. et al., 2014. Declining aerosols in CMIP5 projections: Effects on
1016	atmospheric temperature structure and midlatitude jets. Journal of Climate, 27(18),
1017	pp.6960–6977.
1018	Rotstayn, L.D., Collier, M.A. & Luo, J.J., 2015. Effects of declining aerosols on
1019	projections of zonally averaged tropical precipitation. Environmental Research
1020	<i>Letters</i> , 10(4).
1021	Roxy, M.K. et al., 2017. A threefold rise in widespread extreme rain events over central
1022	India. <i>Nature Communications</i> , pp.1–11. Available at:
1023	http://dx.doi.org/10.1038/s41467-017-00744-9.
1024	Salinger, M.J. & Griffiths, G.M., 2001. Trends in New Zealand daily temperature and
1025	rainfall extremes. International Journal of Climatology, 21(12), pp.1437–1452.
1026	Available at: http://dx.doi.org/10.1002/joc.694.
1027	Salzmann, M. et al., 2010. Two-moment bulk stratiform cloud microphysics in the GFDL
1028	AM3 GCM: Description, evaluation, and sensitivity tests. Atmospheric Chemistry
1029	and Physics, 10(16), pp.8037–8064.
1030	Salzmann, M. & Cherian, R., 2015. On the enhancement of the Indian summer monsoon

1031	drying by Pacific multidecadal variability during the latter half of the twentieth
1032	century., pp.9103–9118.
1033	Salzmann, M., Weser, H. & Cherian, R., 2014. Robust response of Asian summer
1034	monsoon to anthropogenic aerosols in CMIP5 models. Journal of Geophysical
1035	<i>Research: Atmospheres</i> , 119(19), p.11,321-11,337. Available at:
1036	http://doi.wiley.com/10.1002/2014JD021783.
1037	Sharmila, S. et al., 2015. Future projection of Indian summer monsoon variability under
1038	climate change scenario: An assessment from CMIP5 climate models. Global and
1039	Planetary Change, 124(0), pp.62–78.
1040	Sillmann, J. et al., 2013. Aerosol effect on climate extremes in Europe under different
1041	future scenarios. Geophysical Research Letters, 40(10), pp.2290–2295.
1042	Singh, D. et al., 2014. Observed changes in extreme wet and dry spells during the South
1043	Asian summer monsoon season. <i>Nature Climate Change</i> , 4(April), pp.1–6.
1044	Smith, S.J. et al., 2011. Anthropogenic sulfur dioxide emissions: 1850–2005. Atmos.
1045	Chem. Phys., 11(3), pp.1101–1116. Available at: https://www.atmos-chem-
1046	phys.net/11/1101/2011/.
1047	Sperber, K.R. et al., 2013. The Asian summer monsoon: an intercomparison of CMIP5
1048	vs. CMIP3 simulations of the late 20th century. <i>Climate Dynamics</i> , 41(9–10),
1049	pp.2711–2744. Available at: http://dx.doi.org/10.1007/s00382-012-1607-6.
1050	Stanberry, L., 2013. Permutation Test. In W. Dubitzky et al., eds. <i>Encyclopedia of</i>
1051	Systems Biology. New York, NY: Springer New York, p. 1678. Available at:
1052	http://dx.doi.org/10.1007/978-1-4419-9863-7 1186.
1053	Stowasser, M., Annamalai, H. & Hafner, J., 2009. Response of the South Asian Summer
1054	Monsoon to Global Warming: Mean and Synoptic Systems*. Journal of Climate,
1055	22(4), pp.1014–1036. Available at: http://dx.doi.org/10.1175/2008JCLI2218.1.
1056	Taylor, K.E., Stouffer, R.J. & Meehl, G.A., 2012. An Overview of CMIP5 and the
1057	Experiment Design. Bulletin of the American Meteorological Society, 93(4),
1058	pp.485–498. Available at: http://dx.doi.org/10.1175/BAMS-D-11-00094.1.
1059	Turner, A.G. & Annamalai, H., 2012. Climate change and the South Asian summer
1060	monsoon. Nature Clim. Change, 2(August), pp.587–595. Available at:
1061	http://dx.doi.org/10.1038/nclimate1495.
1062	Vinnarasi, R. & Dhanya, C.T., 2016. Changing characteristics of extreme wet and dry
1063	spells of Indian monsoon rainfall. Journal of Geophysical Research: Atmospheres,
1064	121(5), pp.2146–2160.
1065	Vinoj, V. et al., 2014. Short-term modulation of Indian summer monsoon rainfall by
1066	West Asian dust. Nature Geosci, 7(4), pp.308–313.
1067	Vuuren, D.P. et al., 2011. A special issue on the RCPs. Climatic Change, 109(1), pp.1-4.
1068	van Vuuren, D.P. et al., 2011. The representative concentration pathways: an overview.
1069	Climatic Change, 109(1), p.5. Available at: https://doi.org/10.1007/s10584-011-
1070	0148-z.
1071	Wang, C., 2015. Anthropogenic aerosols and the distribution of past large-scale
1072	precipitation change. Geophysical Research Letters, 42(24), pp.10876–10884.
1073	Wang, C. et al., 2009. Impact of anthropogenic aerosols on Indian summer monsoon.
1074	Geophysical Research Letters, 36(21), p.L21704. Available at:
1075	http://dx.doi.org/10.1029/2009GL040114.
1076	Wilcox, L.J., Highwood, E.J. & Dunstone, N.J., 2013. The influence of anthropogenic

- aerosol on multi-decadal variations of historical global climate. *Environmental Research Letters*, 8(2).
- 1079 Xue, H. & Feingold, G., 2006. Large-Eddy Simulations of Trade Wind Cumuli:
 1080 Investigation of Aerosol Indirect Effects. *Journal of the Atmospheric Sciences*,
 1081 63(6), pp.1605–1622. Available at:
- 1082 http://journals.ametsoc.org/doi/abs/10.1175/JAS3706.1.
- Yatagai, A. et al., 2012. APHRODITE: Constructing a Long-Term Daily Gridded
 Precipitation Dataset for Asia Based on a Dense Network of Rain Gauges. *Bulletin of the American Meteorological Society*, 93(9), pp.1401–1415. Available at:
 http://dx.doi.org/10.1175/BAMS-D-11-00122.1.
- 1087 Zhang, L. & Li, T., 2016. Relative Roles of Anthropogenic Aerosols and Greenhouse
 1088 Gases in Land and Oceanic Monsoon Changes during Past 156 years in CMIP5
 1089 Models. *Geophysical Research Letters*, p.n/a-n/a.
- Zhou, C. & Penner, J.E., 2017. Why do general circulation models overestimate theaerosol cloud lifetime effect? A case study comparing CAM5 and a CRM.
- 1092 *Atmospheric Chemistry and Physics*, 17(1), pp.21–29.
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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.

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 (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

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

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
- 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)

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

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
- individual forcing experiments in three additional CMIP5 models with multiple ensemble
- 1192 members for the individual forcing experiments. Grey numbers indicate correlations that
- are insignificant at the 5% level.