

1 **Separating atmospheric and ocean-mediated impacts of time-evolving**
2 **industrial and biomass burning aerosols on historical boreal summer**
3 **climate**

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ABSTRACT

28 The spatial pattern of industrial aerosol emissions has changed markedly over the past
29 century, alongside regional shifts in biomass burning (BMB) sources. While previous studies
30 examined evolving industrial aerosol effects, the relative contributions of atmospheric radiative
31 vs. ocean-mediated responses to combined aerosol sources remain unclear. Using global
32 atmosphere-land simulations driven by time-evolving historical industrial and BMB aerosol
33 emissions and aerosol-induced sea surface temperature (SST) anomalies, we identify two
34 leading modes of multi-decadal aerosol optical depth (AOD) variability in boreal summer,
35 together accounting for 94% of the AOD variance from 1930-2030. Mode 1 depicts a
36 monotonic increase over low latitudes, while Mode 2 describes a sinusoidal trajectory over
37 Northern Hemisphere continents with weaker opposite changes over South and East Asia.
38 These AOD modes drive distinct atmospheric circulation, precipitation and terrestrial air
39 temperature responses. We separate the atmospheric radiative and ocean-mediated pathways
40 of response through a novel design using time-evolving aerosol-induced SST anomalies from
41 the Community Earth System Model version 1 (CESM1) Single-Forcing and All-Forcing
42 Large Ensembles. We find that ocean-mediated effects dominate the large-scale atmospheric
43 circulation and precipitation responses, while atmospheric radiative effects induce robust
44 regional impacts and modulate the ocean-mediated response in remote regions. Our results
45 demonstrate that transient aerosol impacts reflect a dynamic balance between atmospheric
46 radiative and ocean-mediated pathways. These highlight the importance of considering both
47 spatial pattern and response pathway when assessing aerosol-driven climate variations.

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SIGNIFICANCE STATEMENT

50 Aerosols from industrial activities and biomass burning have changed significantly over
51 the 20th century. We conduct a novel set of climate model simulations to investigate how the
52 changing spatial distribution of these aerosol emissions influenced historical climate via
53 atmospheric and oceanic pathways. Using a pattern-based approach, we find that these aerosols
54 influence global climate by modifying ocean surface temperatures, which in turn drive changes
55 in global-scale atmospheric circulation, affecting precipitation and terrestrial air temperature.
56 Aerosols' interaction with radiation in the atmosphere drives local and regional impacts and
57 modulates oceanic-driven responses in remote regions. Our results highlight the importance of

58 evolving aerosol patterns in shaping historical anthropogenic climate change and underscore
59 the complexity of atmospheric and oceanic pathways governing the responses.

60 **1. Introduction**

61 Aerosols from anthropogenic sources are one of the dominant drivers of climate change
62 over the industrial era (IPCC AR6, 2021). Aerosols from industrial sources have played a key
63 role in shaping historical patterns of forced climate change in the atmosphere and ocean, with
64 impacts on regional temperature and precipitation (e.g., Dong et al. 2024; Shi et al. 2022; Kang
65 et al. 2021; Deser et al. 2020; Xie et al. 2013). For instance, Northern Hemisphere (NH)
66 industrial aerosol emissions have been linked to the observed southward shift of the tropical
67 Pacific and Atlantic rain belts in the late 20th century (Ming and Ramaswamy, 2011; Hwang
68 et al., 2013; Hill et al., 2015; Allen et al., 2015). Aerosols from industrial sources, including
69 those at lower latitudes, have also contributed to the weakening of South and East Asian
70 Summer Monsoon circulations and the associated decrease in monsoon precipitation (Bollasina
71 et al., 2011, 2014), as well as Sahel drought and subsequent recovery (Hua et al. 2019;
72 Hirasawa et al., 2020). Industrial aerosol emissions have also been implicated in the recent
73 weakening of the NH summertime atmospheric circulation, including the upper-level jet stream
74 (Dong et al., 2022) and storm tracks (Kang et al., 2024; Chemke and Coumou, 2024). In
75 addition, the historical NH mid-latitude aerosol increase has been suggested to delay the
76 emergence of Arctic amplification until the past few decades (Mueller et al., 2018; England et
77 al., 2021) while their reductions after the 1980s are linked to recent enhanced summer warming
78 in western Europe (Schumacher et al., 2024; Roesch et al., 2025). Furthermore, industrial
79 aerosol emissions, by modulating atmospheric radiation reaching the surface, have affected
80 oceanic properties such as sea surface temperature (SST), sea surface salinity and heat content
81 (Shi et al., 2023; Dong et al., 2024), which in turn influence ocean heat uptake and heat
82 exchange between basins (Li et al., 2023), and have delayed the formation of the “North
83 Atlantic Warming Hole” (Dagan et al., 2020).

84 Biomass burning (BMB) emissions, another major partially anthropogenic source of
85 aerosols, have co-evolved with industrial emissions. However, the large-scale climatic impacts
86 of BMB aerosols have received little attention until recently, due in part to incomplete
87 inventories of historical BMB emissions (Hua et al., 2024) and BMB’s relatively minor effect
88 on the global mean energy balance (IPCC AR6, 2021). In addition, large uncertainties in BMB-
89 driven aerosol-cloud interactions, and their dependence on aerosol types has hampered

90 progress (IPCC AR6, 2021). The recent acceleration of human-related wildfire worldwide has
91 prompted renewed scrutiny of the role of BMB emissions in anthropogenic climate change
92 (Cunningham et al., 2024). In particular, it has been demonstrated that BMB emissions reduce
93 time-mean total aerosol radiative forcing (Heyblom et al., 2023), partially mitigating the effect
94 of industrial aerosol emissions on the strength of the Atlantic Meridional Overturning
95 Circulation (AMOC; Allen et al., 2024; Liu et al., 2024) and accelerating externally-forced 20th
96 century warming of the tropical Indian Ocean at a faster speed relative to other tropical ocean
97 basins (Tian et al., 2023). In addition, interannual variations in BMB emissions have been
98 shown to produce a sizeable effect on NH climate via non-linear aerosol-cloud interactions,
99 contributing to high-latitude warming and Arctic sea ice decline in recent decades (Fasullo et
100 al., 2022; DeRepentigny et al. 2022).

101 Industrial aerosol and BMB emissions are spatially heterogenous and have evolved
102 substantially over time though with different spatiotemporal patterns. Industrial aerosol
103 emissions are primarily located in the NH subtropics and middle latitudes, while BMB
104 emissions mainly occur in the deep tropics and boreal regions (Van Der Werf et al., 2017).
105 From the 1950s-1970s, industrial aerosol emissions increased rapidly over North America and
106 western Europe, followed by marked declines resulting from clean air legislation (e.g., Deser
107 et al., 2020). After the late 1970s, the locus of industrial aerosol emissions shifted to East Asia
108 and South Asia. Following the passage of China’s Air Pollution Prevention and Control Action
109 Plan (Zhang et al., 2021), local industrial emissions dropped rapidly particularly after 2013
110 (Wang et al., 2021), shaping a dipole pattern between East and South Asia (Xiang et al., 2023).
111 Meanwhile, tropical BMB emissions increased substantially over the second half of the 20th
112 century in central Africa, South America and Southeast Asia due to agricultural activities and
113 land use changes (Van Der Werf et al., 2017). Boreal BMB emissions have also risen over the
114 20th century, particularly in Siberia and parts of Canada (Van Der Werf et al., 2017).

115 The effect of radiative forcings such as aerosols can be decomposed into atmospheric
116 radiative and ocean-mediated components. These are commonly referred to as “fast” and
117 “slow” responses, respectively, reflecting the timescales over which they appear, where “fast”
118 denotes the response in the absence of significant surface temperature changes and “slow”
119 denotes the surface temperature feedback response (Gregory et al., 2004; Bala et al., 2009).
120 Distinguishing them is important because their drivers emerge on different timescales, and thus
121 the timescale of corresponding responses varies. This decomposition is typically assessed using

122 a parallel set of idealized experiments in which aerosol emissions averaged over a particular
123 time interval of interest are prescribed in both the fully-coupled model and its atmosphere-only
124 component (e.g., Samset et al., 2016; Liu et al., 2018).

125 While such a “time-slice” approach serves as a valuable tool for studying the fundamental
126 physical processes underlying the response to a fixed aerosol emission target, it becomes
127 limited under transient conditions. Given the distinct spatial distributions and temporal
128 evolutions of industrial and BMB aerosols, their varied aerosol types (primarily absorbing
129 carbonous particles for BMB and predominantly scattering particles for industrial aerosols) and
130 location-dependent efficacies (Persad and Caldeira 2018), it is crucial to account for their
131 evolving trajectories when assessing their climate impacts. Under the transient framework, the
132 separation between “fast” and “slow” responses becomes blurred. In particular, the response to
133 time-evolving aerosol emissions in the absence of SST changes (e.g., the radiatively forced
134 contribution) will include a “slow” component by virtue of the fact that aerosol emissions vary
135 slowly through time. Thus, both the atmospheric radiative and ocean-mediated effects of
136 aerosols must be considered on decadal and longer time scales. To date, the climate response
137 to time-evolving patterns of historical industrial, and to a lesser extent BMB, aerosol emissions
138 has been investigated using fully coupled models (e.g., Deser et al., 2020; Wang and Wen,
139 2022; Dong et al., 2024). While such studies highlight the importance of accounting for the full
140 spatiotemporal evolution of aerosol emissions, they do not allow for insights into the relative
141 contributions of atmospheric radiative *vs.* ocean-mediated components of response. Moreover,
142 the role of BMB aerosols and their interplay with industrial aerosols has been largely ignored.

143 We address these gaps by employing a novel atmospheric modeling framework to
144 decompose the combined effects of time-varying industrial and BMB aerosol emissions into
145 atmospheric radiative and ocean-mediated components. Specifically, we conduct a suite of
146 experiments with Community Atmosphere Model version 5 (CAM5) forced with time-varying
147 historical aerosol (industrial plus BMB) emissions and associated SST responses individually
148 and in combination, where the aerosol-induced SST changes are derived from the fully coupled
149 CESM1 Single-Forcing and All-Forcing Large Ensembles (Kay et al., 2015; Deser et al., 2020).
150 This approach allows us to elucidate the individual roles of evolving atmospheric radiative *vs.*
151 ocean-mediated impacts of industrial and BMB aerosols on historical climate change. A similar
152 methodology has been previously used in the context of idealized 1% per year CO₂ forcing (He
153 and Soden, 2015) and quadrupling of CO₂ concentration (Tiffany and Vigot, 2015)

154 experiments, but this is the first time it has been adopted for the historical evolution of industrial
155 and BMB aerosols to the best of our knowledge. If adopted in a model intercomparison effort,
156 this framework could also be beneficial for understanding models' structural diversity in
157 aerosol-induced climate change. In addition, this protocol may prove useful for informing
158 dynamical and statistical regional downscaling applications, as it provides large-scale, aerosol-
159 forced signals that can serve as consistent boundary conditions. This consistency is particularly
160 valuable for intercomparisons across models and methods, where isolating the aerosol
161 influence on regional climate remains a key challenge (Nabat et al., 2025).

162 This study has three aims: 1) To identify the dominant patterns of time-varying aerosol
163 emissions from industrial and BMB sources over the historical period (1920-present); 2) To
164 assess how the dominant patterns of time-evolving aerosol emissions influence precipitation,
165 terrestrial air temperature and the large-scale atmospheric circulation; and 3) To quantify the
166 relative contributions of atmospheric radiative vs. ocean-mediated pathways of response. We
167 focus on boreal summer (June-to-August, JJA), when NH monsoon systems and low-level jets
168 drive strong signals, making it a natural season for assessing aerosol impacts particularly at
169 regional scales. Our experimental design is described in Section 2, results are reported in
170 Section 3, and summarized and discussed in Section 4.

171 **2. Data and Methods**

172 *a. Model Simulations*

173 We use CAM5 (Neale et al., 2010) coupled to Community Land Model version 4 (CLM4;
174 Lawrence et al., 2011) at a spatial resolution of 1° for our AMIP simulations. CAM5 and CLM4
175 are the atmospheric and land model components of CESM1, respectively. CAM5 deploys a
176 three-mode modal aerosol scheme (Liu et al., 2012) with prognostic aerosols and includes both
177 the direct and indirect aerosol radiative effects for liquid and ice phase clouds (Morrison and
178 Gettelman 2008). CLM4 includes a biogeochemical model which can prognostically simulate
179 vegetation changes and a Snow and Ice Aerosol Radiation model which accounts for aerosol's
180 impact on snow cover, as well as its radiative feedback.

181 We conducted three sets of AMIP ensembles, termed RAD-AMIP, SST-AMIP and FULL-
182 AMIP (Table 1). Each set has 10 ensemble members and covers the period 1920-2030. For
183 each set, the ensemble members are initialized with a small (order 10^{-14} K) random atmospheric
184 temperature perturbation to create ensemble spread. RAD-AMIP uses time-varying CMIP5

185 historical (prior to 2006) and Representative Concentration Pathway (RCP) 8.5 (2006 onward)
186 industrial (AER_{indus}) and BMB (from agriculture activities, grass and forest fires) aerosol
187 emissions (Lamarque et al., 2010). All other radiative forcings, as well as SSTs and sea ice
188 concentrations (SICs), are set to their 1920 ensemble-mean seasonal cycles from the 40-
189 member CESM1 “All Forcing” Large Ensemble (ALL). SST-AMIP uses time-varying aerosol-
190 induced SSTs and SICs; all radiative forcings including AER_{indus} and BMB are set to their 1920
191 ensemble-mean seasonal cycles in ALL. The time-varying aerosol-induced SSTs and SICs are
192 computed from the difference between the ensemble-mean of ALL and the sum of the
193 ensemble-means of the fixed AER_{indus} and fixed BMB CESM1 Single Forcing Large
194 Ensembles following Deser et al. (2020). FULL-AMIP uses the same time-varying aerosol
195 emissions as RAD-AMIP plus the same time-varying SSTs and SICs as SST-AMIP. Given that
196 the RAD-AMIP and SST-AMIP responses are largely additive (see below), we conclude that
197 our AMIP experimental design is a valid approach for separating atmospheric radiative and
198 ocean-mediated (SST-driven) components of the aerosol-forced response. Throughout the
199 remainder of the manuscript, we refer to the SST-AMIP signal as the “ocean-mediated” or
200 “SST-driven/induced” response and the RAD-AMIP signal as the “radiative forcing”,
201 “radiatively driven/induced/forced” or “atmospheric radiative” response.

202 **Table 1.** CAM5 AMIP experiments and their forcings. Each set of experiments contains 10 ensemble
203 members and covers the period 1920-2030. See text for details.

	RAD-AMIP	SST-AMIP	FULL-AMIP
Radiative Forcing	Time-varying industrial and BMB aerosol emissions. All other radiative forcings fixed at 1920 levels.	All radiative forcings fixed at 1920 levels.	Time-varying industrial and BMB aerosol emissions. All other radiative forcings fixed at 1920 levels.
SST/SIC Forcing	SST and SIC fixed at 1920 levels.	Time-varying SST and SIC induced by industrial and BMB aerosol emissions.	Time-varying SST and SIC induced by industrial and BMB aerosol emissions.

204 *b. AOD and SST patterns*

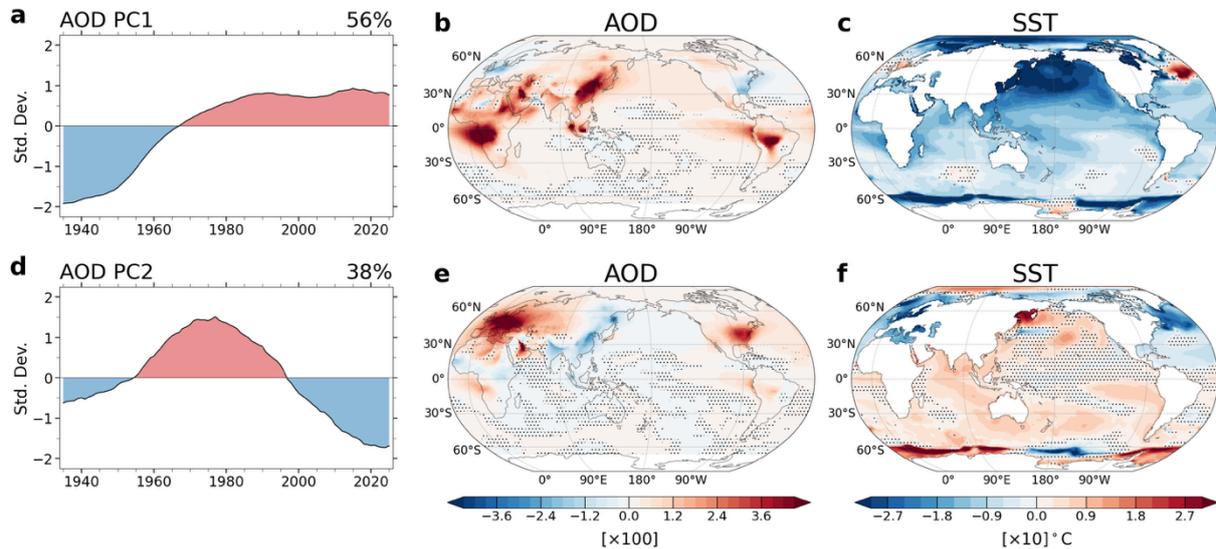
205 To define the dominant patterns of aerosol radiative forcing in FULL-AMIP, we apply
206 Empirical Orthogonal Function (EOF) analysis to JJA AOD (550nm wavelength) based on
207 low-pass filtered (using 10-year running means) ensemble means from 1930-2030 (similar
208 results are obtained using AOD from RAD-AMIP which is not shown; we exclude 1920-1929
209 to minimize the influence of residual memory from ocean initial conditions). The two leading
210 AOD modes account for 94% of AOD variance during 1930-2030 and are thus the focus of our
211 analysis. To define the aerosol-induced SST patterns, we regress 10-year low-pass filtered
212 ensemble mean SSTs in FULL-AMIP onto the normalized AOD Principal Component (PC)
213 timeseries. Applying EOF analysis directly to the aerosol-induced SST field yields nearly
214 identical patterns to those obtained via regression onto the AOD PCs, although there is a slight
215 (3-4 year) phase lag between the SST PCs and their AOD PC counterparts (not shown). The
216 atmospheric responses to the two leading AOD modes are obtained by regressing 10-year low-
217 pass filtered ensemble means of precipitation, sea level pressure (SLP) and land surface air
218 temperature (LSAT) in FULL-AMIP, RAD-AMIP and SST-AMIP onto the normalized AOD
219 PCs. Substituting aerosol-induced SST PCs in place of AOD PCs yields similar atmospheric
220 regression patterns (not shown); thus, for simplicity, we use the AOD PCs throughout this
221 study. Statistical significance of all regression values is assessed using a two-tailed Student's
222 t-test at the 95% confidence level.

223 **3. Results**

224 *a. Leading modes of AOD variability and associated SST patterns*

225 The two leading modes of JJA AOD variability show distinctive temporal and spatial
226 characteristics (Fig. 1). PC1, which explains 56% of the AOD variance, depicts a pronounced
227 (3 standard deviation) increase from the 1930s-1980s with little change thereafter, while PC2,
228 which explains 38% of the AOD variance, shows more sinusoidal behavior, with a positive
229 trend from about 1935-1975 followed by a steep decline to the early 2020s (Figs. 1a,d). The
230 corresponding AOD spatial pattern associated with PC1 exhibits positive regression values
231 over low latitudes of the Eastern Hemisphere, with regional hotspots in East and South Asia
232 associated with industrial emissions and in tropical South Africa, South America, Indonesia
233 and Malaysia associated with BMB emissions; weak negative values are found in the eastern
234 US and western Europe (Fig. 1b). AOD Mode 2 depicts a zonally-asymmetric NH AOD
235 pattern, with large positive values in the eastern US and western Europe juxtaposed against
236 weaker negative values in South and East Asia from industrial aerosol emissions and in eastern

237 Siberia from BMB emissions (Fig. 1e). Qualitatively, Mode 1 captures the rapid
 238 industrialization of Asia and Africa in the past century, along with the peak of major Asian
 239 aerosol emissions in the 2010s through present (Takemura et al., 2012). Mode 2 represents
 240 industrialization followed by clean air regulation in the Western Hemisphere.



241
 242 **Figure 1.** (a,d) Principal Component (PC) time series of Aerosol Optical Depth (AOD) in JJA based on
 243 FULL-AMIP. Percent variance explained by Modes 1 and 2 (PC1 and PC2) are given in the upper right.
 244 (b,c) Regression of JJA AOD ($\times 100$) and SST ($^{\circ}\text{C} \times 10$) in FULL-AMIP onto AOD PC1. (e,f) As in (b,c)
 245 but for AOD PC2. Regions without stippling are significant at the 95% confidence level based on a two-
 246 tailed Student's t-test.

247 The SST anomalies associated with AOD Mode 1, obtained by regressing ensemble-mean
 248 JJA SST anomalies from FULL-AMIP onto AOD PC1, exhibit widespread cooling, as
 249 expected from the global increase in AOD (Fig. 1c). The cooling is greatest over the North
 250 Pacific and along the sea ice margins of both hemispheres. A strong interhemispheric SST
 251 anomaly gradient is evident over the Pacific sector and to a lesser extent the Indian Ocean,
 252 likely associated with aerosol emissions over South and East Asia (Hwang et al., 2024).
 253 Pronounced SST warming is found in the central subpolar North Atlantic. This may be
 254 associated with a strengthening of AMOC and accompanying increase in northward ocean heat
 255 transport in response to AOD reductions over eastern North America and Europe (Cai et
 256 al., 2006; Delworth & Dixon, 2006; Booth et al., 2012; Menary et al., 2020; Dagan et al., 2020),
 257 but local wind-driven processes may also play a role.

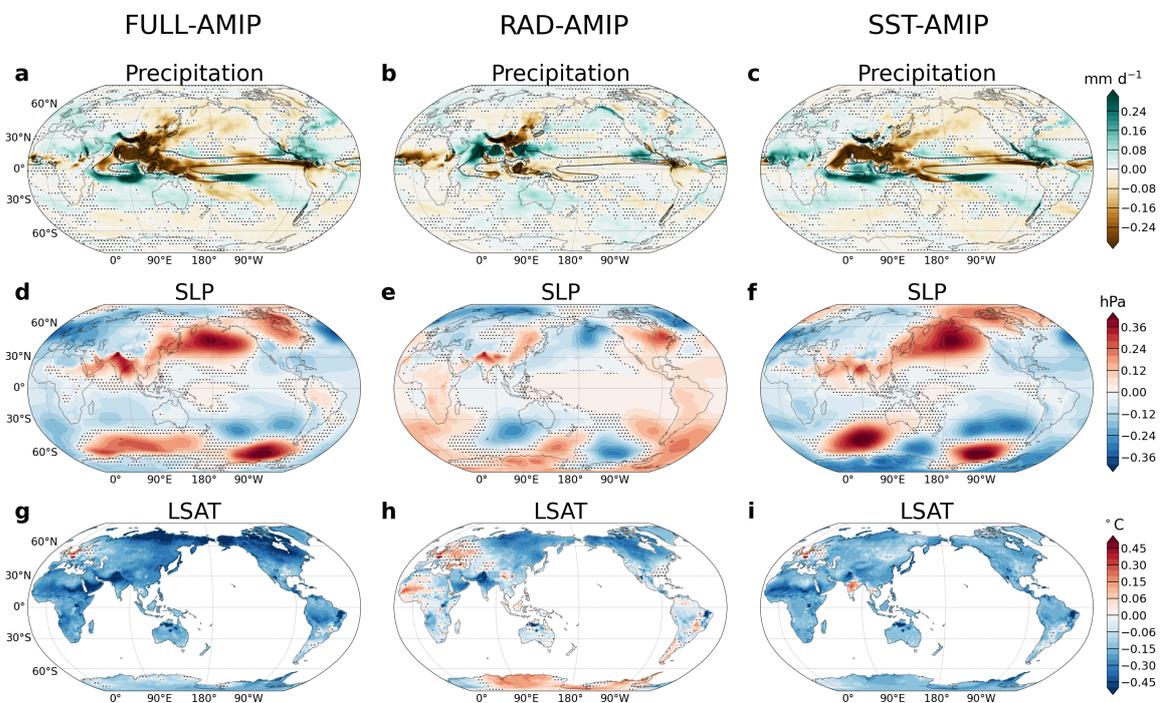
258 The SST anomalies associated with AOD Mode 2 are generally weaker than those
 259 associated with Mode 1 and depict a strong zonal contrast over the NH with widespread cooling
 260 over the North Atlantic and Mediterranean and warming over most of the North Pacific (Fig.
 261 1f). The North Atlantic cooling likely arises in response to increased aerosol emissions over

262 the eastern US and Europe (e.g., Undorf et al., 2018a), while the warming in the Pacific and
 263 Indian Ocean sectors is presumably a result of reduced AOD over eastern Asia and India (Fig.
 264 1f).

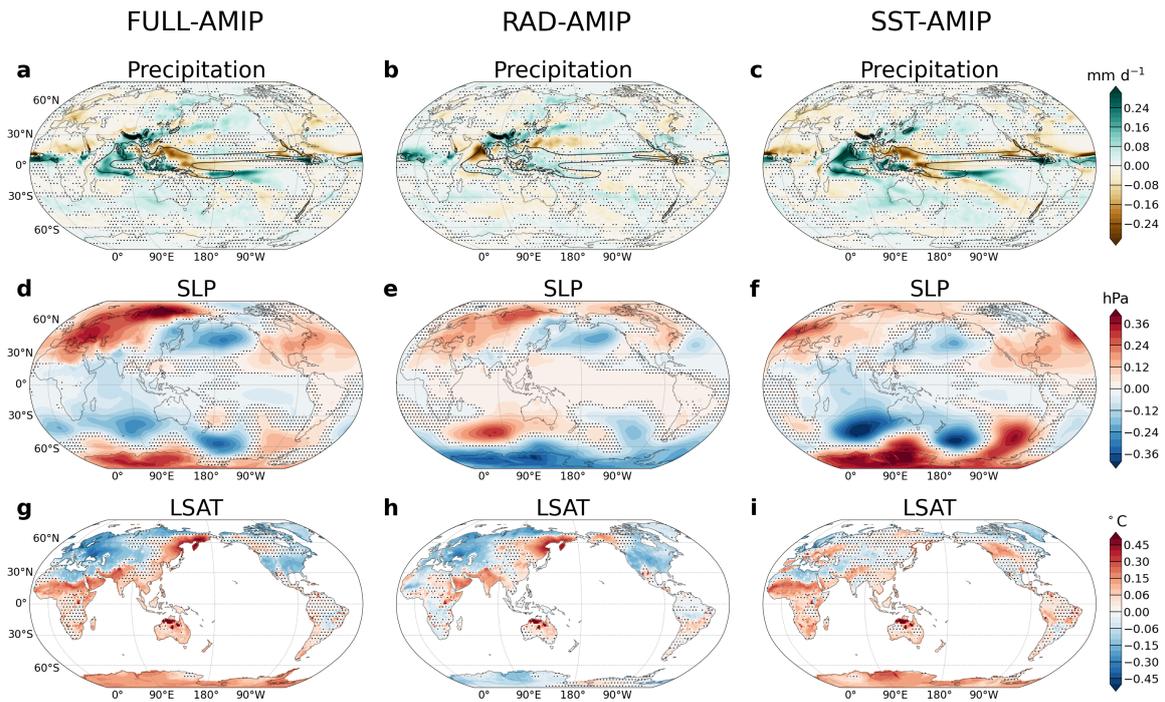
265 Investigation of the mechanisms underlying the SST patterns associated with each AOD
 266 Mode is beyond the scope of this study, but related analyses may be found in Kang et al. (2021),
 267 Shi et al. (2023), Hwang et al. (2024) and Diao et al. (2025), although none considered the
 268 combined effects of industrial and BMB aerosols. Here, our interest is in how these aerosol-
 269 induced SST patterns impact the atmosphere.

270 *b. Global atmospheric response patterns*

271 We begin by examining the global atmospheric response to combined atmospheric radiative
 272 and SST-driven changes associated with each AOD mode. To do this, we regress the FULL-
 273 AMIP JJA ensemble-means onto the PC timeseries associated with each AOD Mode.
 274 Regression maps of precipitation, SLP and LSAT for AOD Mode 1 are shown in Fig. 2 and for
 275 AOD Mode 2 in Fig. 3. Regression maps for associated surface downward short-wave radiative
 276 flux (SWSD) under total-sky, clear-sky and cloudy-sky conditions in both modes are given in
 277 Fig. 4.

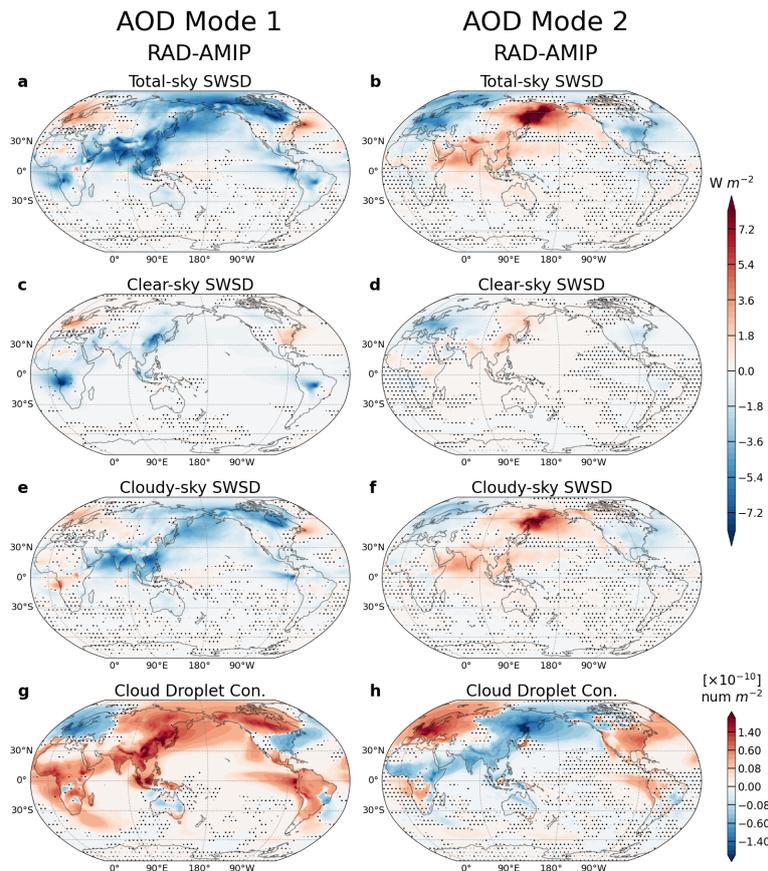


278
 279 **Figure 2.** AOD Mode 1 response of (a,b,c) precipitation (mm d^{-1}), (d,e,f) sea level pressure (SLP; hPa) and
 280 (g,h,i) land surface air temperature (LSAT; $^{\circ}\text{C}$) in FULL-AMIP, RAD-AMIP and SST-AMIP regressed onto
 281 AOD PC1. Regions without stippling are significant at the 95% confidence level based on a two-tailed
 282 Student's t-test. Black contours on precipitation panels show the climatological 6 mm d^{-1} isopleth.



283

284 **Figure 3.** As in Figure 2 but for AOD PC2.



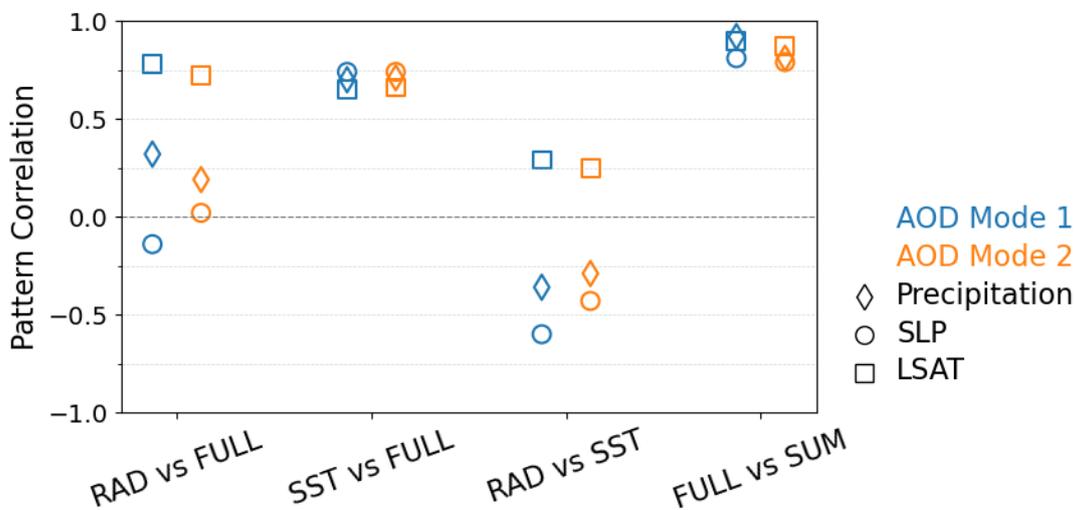
285

286 **Figure 4.** AOD Mode 1 and Mode 2 responses of short-wave surface downward (SWSD) radiative flux (W
 287 m^{-2}) under (a,b) total-sky, (c,d) clear-sky and (e,f) cloudy-sky conditions, and (g,h) vertically integrated
 288 cloud droplet number concentration ($num\ m^{-2} \times 10^{-10}$) in RAD-AMIP regressed onto (left) AOD PC1 and
 289 (right) AOD PC2. Regions without stippling are significant at the 95% confidence level based on a two-
 290 tailed Student's t-test.

291 Both modes exhibit statistically significant responses that are global in scale. AOD Mode
292 1 features prominent drying over South and East Asia, the northern Indian Ocean and the
293 Maritime Continent, regions proximate to the main centers of AOD increase (Fig. 2a),
294 accompanied by reduction in SWSD (Fig. 4a). Compensating areas of wetting occur directly
295 south of the equator in the Indian Ocean and central Pacific. At higher latitudes, widespread
296 drying is found over the Southern Ocean, the North Pacific, northeastern Asia and western
297 North America, while wetting occurs over the subtropical south Indian and southeast Pacific
298 Oceans, tropical Africa, southern Europe, the Caribbean and parts of the North Atlantic. Areas
299 of drying (wetting) generally correspond to anticyclonic (cyclonic) SLP anomalies, with largest
300 SLP magnitudes in the extra-tropics (Fig. 2d). The prominent positive SLP response over the
301 North Pacific may be due to in part to a remote Rossby wave teleconnection excited by
302 precipitation reductions over the northern Indian Ocean and Southeast Asia (Smith et al., 2016;
303 Dittus et al., 2021) in addition to local SST cooling. The circulation response in other regions
304 is discussed in Section 3c. AOD Mode 1 exhibits widespread terrestrial cooling (Fig. 2g), as
305 expected from the global-scale increase in AOD (Figs. 1a,b) with widespread SWSD
306 reductions (Fig.4a). The strongest cooling is found in boreal regions, likely associated with
307 positive ice/snow albedo feedback. A “cooling hole” is seen over far western Europe, reflecting
308 the local decrease in AOD with positive SWSD values (Fig. 4a). SWSD in FULL-AMIP
309 closely resembles that in RAD-AMIP, whereas SST-driven SWSD responses primarily
310 associated with cloud changes are much weaker. Cloud responses to aerosol strongly affect the
311 SWSD response in remote regions (Fig. 4e), whereas the direct radiative effect dominates
312 locally (Fig. 4c).

313 AOD Mode 2 shows generally weaker responses compared to Mode 1, especially for
314 precipitation and LSAT (Fig. 3). Like Mode 1, Mode 2 AOD increases are generally
315 accompanied by local terrestrial drying and cooling (Figs. 1e and 3g). For example, AOD
316 increases over Europe and the eastern US are accompanied by negative SWSD, primarily
317 driven by aerosol-cloud interactions, whereas AOD decreases over eastern Asia and India
318 produce the opposite response (Figs. 4b,f,h). Remote precipitation responses are also evident,
319 most prominently in the tropics, including a zonal dipole pattern of drying over the far western
320 Pacific and wetting over the central Indian Ocean, and an increase in precipitation across the
321 equatorial Atlantic. The large-scale SLP response over the extratropical NH is generally
322 opposite in sign between Modes 1 and 2, in keeping with their contrasting NH AOD signatures

323 (Figs. 2d, 3d and 4a,b). However, the tropical and SH SLP response patterns are distinctive
 324 between the two AOD modes.



325
 326 **Figure 5.** Global pattern correlations of precipitation (diamond), SLP (circle) and LSAT (square) regression
 327 maps associated with AOD Mode 1 (blue) and 2 (orange) between different pairs of AMIP simulations. SUM
 328 refers to the sum of RAD-AMIP (RAD) and SST-AMIP (SST) regressions. FULL refers to FULL-AMIP.

329 Next, we assess the relative contributions of atmospheric radiative and ocean-mediated
 330 pathways to the atmospheric responses in FULL-AMIP by regressing ensemble-mean fields
 331 from RAD-AMIP and SST-AMIP onto the AOD PCs. The two pathways produce distinctive
 332 and statistically significant global-scale atmospheric responses (Figs. 2 and 3). Comparing to
 333 FULL-AMIP, it is readily apparent that the SST-driven response dominates the large-scale
 334 patterns of precipitation and SLP response, while radiative forcing tends to oppose SST-
 335 induced responses, although the degree of compensation varies with region. On the other hand,
 336 radiatively forced and SST-driven responses make comparable contributions to the LSAT
 337 response in FULL-AMIP. Quantitatively, the leading role of the SST-induced response for the
 338 precipitation and SLP responses is evidenced by the high pattern correlations (exceeding 0.70)
 339 between SST-AMIP and FULL-AMIP, while those between RAD-AMIP and FULL-AMIP are
 340 substantially lower (< 0.33 ; Fig. 5). In contrast, the importance of both radiative forcing and
 341 ocean-mediated effect to the patterns of LSAT response is reflected in the comparable
 342 magnitudes of spatial correlation between RAD-AMIP and FULL-AMIP and between SST-
 343 AMIP and FULL-AMIP (around 0.65 and around 0.75, respectively; Fig. 5). Radiative forcing
 344 partially counteracts the SST-driven response for both precipitation and SLP responses, as seen
 345 by the modest negative pattern correlations between RAD-AMIP and SST-AMIP (Fig. 5). This
 346 negative correlation is particularly pronounced for the SLP response to AOD Mode 1 (-0.60),

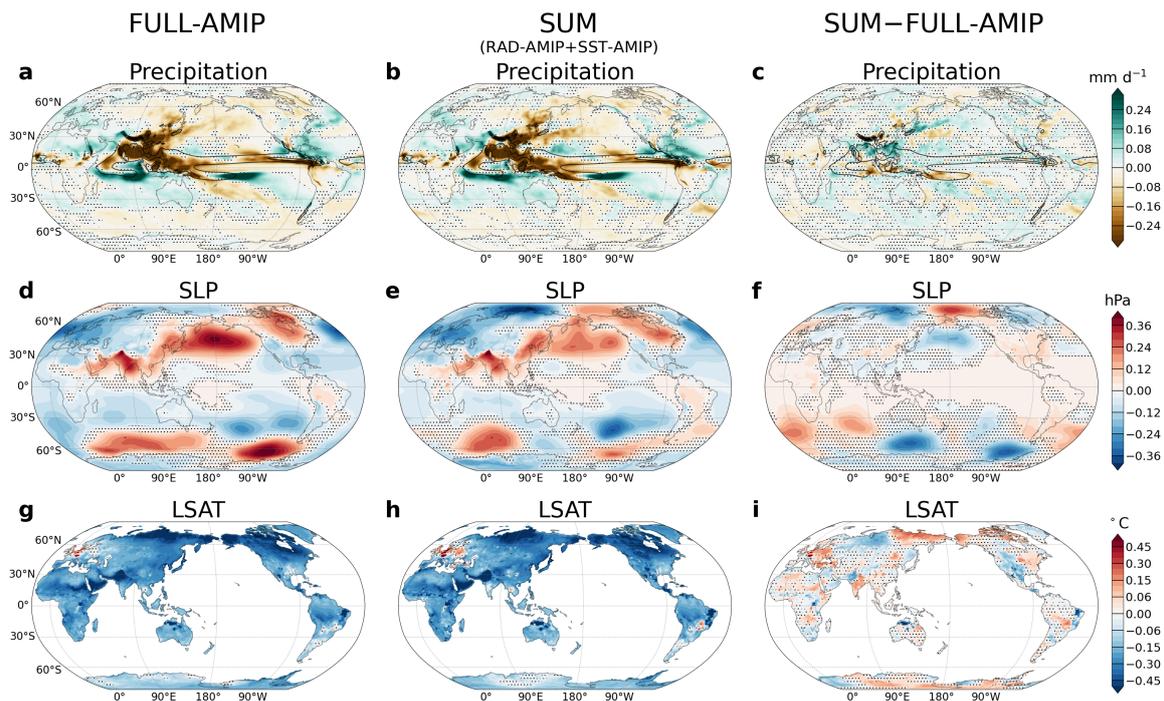
347 as is also visually apparent (Figs. 2e,f). However, for LSAT, the correlations between RAD-
348 AMIP and SST-AMIP are weakly positive (Fig. 5).

349 As mentioned above, the interplay between atmospheric radiative and ocean-mediated
350 pathways depends on the region and quantity of interest. In emission source regions such as
351 East Asia, precipitation responses in both AOD modes are primarily driven by radiative
352 forcing: in Mode 1, AOD increases induce local drying, while AOD decreases in Mode 2
353 induce local wetting (Figs. 1a,d, 2b and 3b). In contrast, the sign of South Asian precipitation
354 response does not always align with local AOD changes because the SST-driven response
355 dominates. For example, in AOD Mode 1, radiative forcing drives a wetting response despite
356 positive AOD values over South Asia, and this radiatively forced wetting is largely offset by
357 SST-driven drying (Figs. 1a and 2b,c). In the NH mid-latitude, the eastern North America and
358 Europe exhibit robust precipitation and LSAT responses in both modes, driven by a
359 combination of radiatively induced and SST-driven thermodynamic and dynamic mechanisms
360 from local and remote forcing sources (Figs. 2b,c and 3b,c). In these regions, in both modes,
361 LSAT changes reflect compensation between radiatively forced and SST-driven responses,
362 whereas western European precipitation results from the reinforcement of the two pathways of
363 response. Over North America, radiatively and SST-driven precipitation and LSAT responses
364 exhibit distinct patterns, reflecting a tug-of-war between locally and remotely induced
365 circulation influences by each pathway. For example, in AOD Mode 1, a radiatively driven
366 negative North Atlantic Oscillation (NAO)-like response (a cyclonic system in the North
367 Pacific) is opposed by SST-driven positive NAO-like response (an anti-cyclonic system in
368 North Pacific). Whether these large-scale circulation responses are triggered by local or remote
369 forcing is hard to determine, warranting further analysis.

370 In low-emission regions, such as the Sahel, for both modes, radiatively forced and SST-
371 driven responses cancel each other, resulting in muted precipitation responses (Figs. 2b,c and
372 3b,c). For AOD Mode 2, radiatively induced and SST-driven SLP responses are of the same
373 sign over most of the NH and tropics but of opposite polarity over the SH high latitudes (Figs.
374 3e,f). The NH LSAT response to AOD Mode 1 shows like-signed contributions from
375 radiatively forced and SST-driven responses (Figs. 2h,i), whereas the LSAT response to AOD
376 Mode 2 depicts nearly orthogonal patterns (Figs. 3h,i).

377 The complex regional interplay between radiatively forced and SST-driven responses
378 would naturally raise a question of to what extent these responses are additive. We address this

379 question by comparing the sum of the RAD-AMIP and SST-AMIP regressions (SUM) with
 380 the FULL-AMIP regressions. Both AOD modes show a large degree of additivity, as evidenced
 381 by the overall similarity in pattern and amplitude between SUM and FULL-AMIP (Fig. 6 for
 382 Mode 1 and Fig. S1 for Mode 2). In general, the differences between SUM and FULL-AMIP
 383 are relatively small in magnitude, albeit statistically significant, except for SLP at high latitudes
 384 where larger discrepancies are found. Pattern correlations between SUM and FULL-AMIP are
 385 high (0.79–0.92) for all three variables in both AOD modes (Fig.5).



386

387 **Figure 6.** As in Fig. 2 but for additivity analysis based on AOD Mode 1 (a,d,g) FULL-AMIP, (b,e,h) the
 388 sum of RAD-AMIP and SST-AMIP (SUM), and (c,f,i) the difference of SUM and FULL-AMIP. Regions
 389 without stippling are significant at the 95% confidence level based on a two-tailed Student's t-test. Black
 390 contours on precipitation panels show the climatological 6 mm d⁻¹ isopleth.

391 c. Regional atmospheric response patterns

392 The diversity of regional features described above makes it difficult to generalize the
 393 relative contribution of atmospheric radiative vs. ocean-mediated pathway on aerosol-driven
 394 atmospheric responses at the regional scale. To further illustrate this regional variation, we
 395 focus here on two areas: the tropical Indo-Pacific sector and Southern Ocean, characterized by
 396 distinct response patterns, to demonstrate how radiative forcing originating from regional AOD
 397 changes interacts with the SST-driven response to influence the atmosphere locally and
 398 remotely.

399 1) Tropical Indian-Western Pacific sector

400 The pattern of precipitation response to AOD Mode 1 features a distinctive meridional
401 dipole over the tropical Indian-Western Pacific sector in FULL-AMIP (Fig. 7a), indicative of
402 an Asian Summer Monsoon response to shifting historical aerosol emissions that has been a
403 topic of great interest in the literature (Bollasina et al., 2011; Persad et al., 2017; Undorf et al.,
404 2018b; Westervelt et al., 2020). In our simulations, this precipitation dipole response
405 corresponds to a cross-equatorial overturning circulation, with northerly wind anomalies at
406 low-levels and southerly wind anomalies at upper-levels, which feeds anomalous upward
407 motion along 5°-10°S accompanied by anomalous downward motion along 5°-15°N (Figs.
408 7a,b). This cross-equatorial overturning circulation response results from a subtle interplay
409 between radiatively forced and SST-driven response, which impart different meridional scales
410 and polarities of response. The radiatively forced response exhibits a narrow meridional dipole
411 between the Asian subcontinent from the Tibetan Plateau eastward (27°-30°N) and the northern
412 Indian Ocean-West Pacific (14°-23°N; Figs. 7c,d), while the SST-driven response displays a
413 broader latitudinal dipole structure between the northern (5°-20°N) and southern (5°-12°S)
414 portions of the Indian Ocean (Figs. 7e,f). The radiatively forced and SST-driven responses are
415 offsetting over the northern Indian Ocean. As a result, the response in FULL-AMIP shows a
416 hybrid structure, with characteristics of both the SST-driven response (i.e., the broad cross-
417 equatorial dipole, albeit substantially weaker in its northern lobe) and the radiatively forced
418 response (i.e., the narrow meridional dipole over the Tibetan Plateau extending eastward).

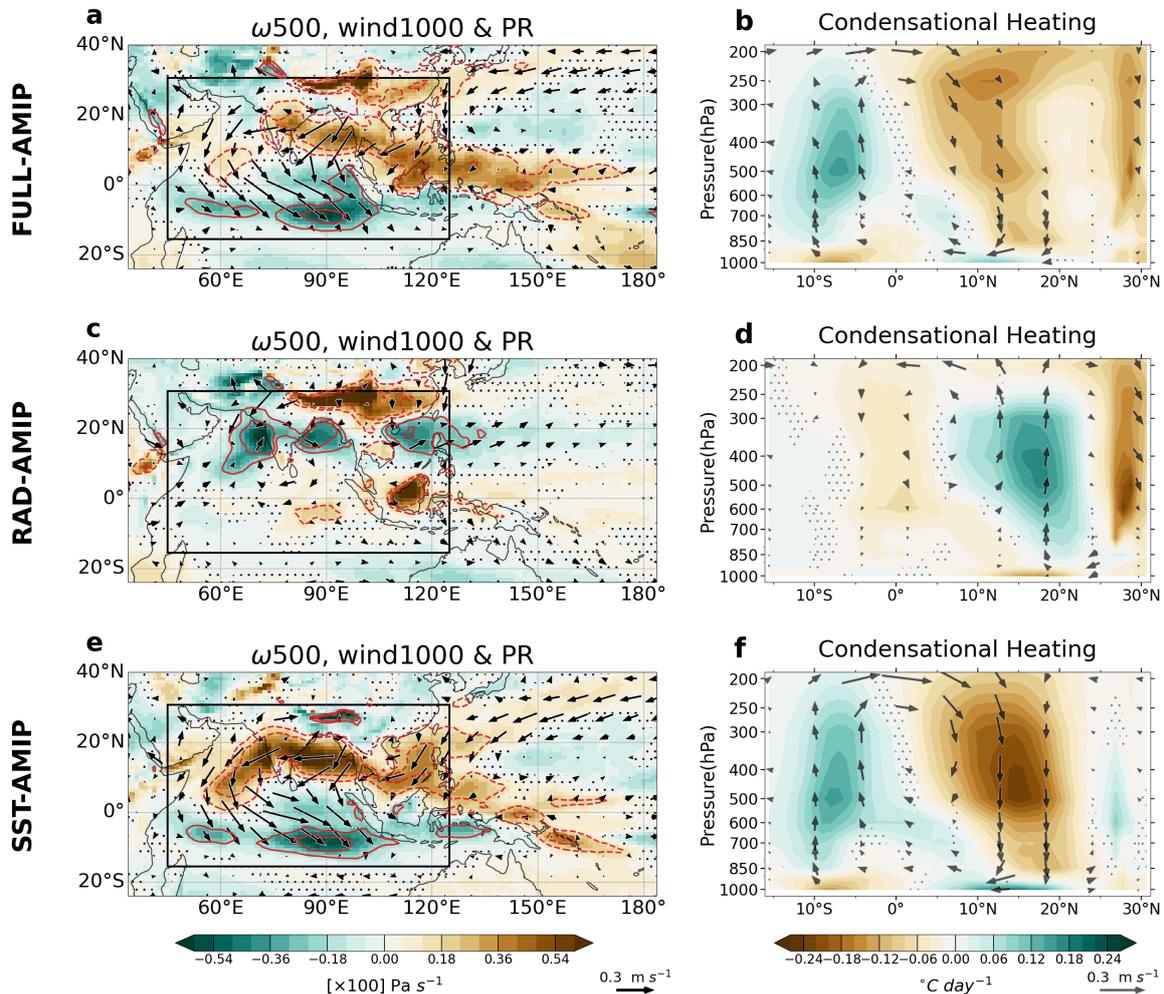
419 The SST-driven precipitation dipole response (Figs. 2a, 7a) likely results from the enhanced
420 meridional temperature gradient over the Indian Ocean (i.e., stronger cooling north of the
421 equator than south; Li et al., 2020), although the pronounced large-scale cooling of the North
422 Pacific may also play a role in displacing the rain belt southward (recall Fig. 1c). Conversely,
423 the radiatively forced precipitation dipole response is likely driven by locally induced drying
424 and subsidence from aerosol emissions over East Asia, with compensating upward motion over
425 the Arabian Sea, Bay of Bengal and the South China Sea (Bollasina et al., 2011; Persad et al.,
426 2017; Westervelt et al., 2020); enhanced cooling over land may also contribute to ascent over
427 the nearby seas.

428 Both aerosol direct and indirect radiative effects contribute to enhanced cooling over
429 Southeast Asia in RAD-AMIP, as seen from the SWSD under clear and cloudy skies,
430 respectively (Figs. 4a,c,e). Aerosol direct effects (e.g., clear-sky SWSD) dominate locally over
431 the aerosol emission source regions (Fig. 4c), while aerosol indirect effects (e.g., cloudy-sky

432 SWSD and associated increase in cloud droplet number concentration) prevail in adjacent
433 regions including the northern Indian Ocean (Figs. 4e,g). Note that the reduction in cloudy-sky
434 SWSD over the northern Indian Ocean and western North Pacific in RAD-AMIP would act to
435 cool the underlying SSTs if the ocean were allowed to respond, consistent with the negative
436 SST anomalies found in SST-AMIP. In SST-AMIP, adiabatic heating associated with the
437 descending branch of the overturning circulation response over the northern Indian Ocean
438 (Figs. 7e,f) reduces cloud cover and allows more insolation to reach the surface (not shown),
439 warming the adjacent Indian Peninsula and parts of central Mainland Southeast Asia (Fig. 2i).
440 However, this warming effect is overwhelmed by radiatively forced cooling (Fig. 2h), resulting
441 in net cooling over these regions in FULL-AMIP (Fig. 2g).

442 The relative role of RAD-AMIP (i.e. atmosphere radiative effects) and SST-AMIP (i.e.
443 ocean-mediated effects) provides valuable insight on the relative importance of in-situ vs. large
444 scale responses to aerosol changes in driving summertime rainfall trends in this region, which
445 has been a topic of substantial debate (Bollasina et al., 2011; Persad et al. 2017; Dong et al.,
446 2019; Li et al., 2018; Wang et al., 2019; Westervelt et al. 2020). AOD Mode 1 captures
447 prevailing northly wind anomalies and precipitation reductions over Asian Summer Monsoon
448 land regions, indicating the weakening of Asian Summer Monsoon (Li et al., 2018; Wang et
449 al., 2019). The SST-driven cross-equatorial dipole circulation accounts for the precipitation
450 response over the central to southern Indian Peninsula and adjacent oceans. Meanwhile East
451 Asian Monsoon weakening arises primarily from radiatively dominated in-situ responses,
452 including subsidence, land cooling and positive SLP anomalies, that suppress ascending
453 motion particularly over the Tibetan Plateau extending eastward. These results suggest that
454 historical South Asian Summer Monsoon weakening reflect effects of East Asian aerosols
455 (Shawki et al, 2018) in Mode 1 here, given the importance of ocean-mediated response. But,
456 when not decomposed by mode, the full response of South Asian Summer Monsoon to aerosols
457 likely includes the contribution from other remote sources such as Europe and North America
458 (Bollasina et al., 2014; Undorf et al., 2018b). In contrast to South Asian Summer Monsoon
459 dominated by remote aerosols, East Asian Monsoon weakening is primarily attributable to local
460 aerosols through atmospheric radiative pathway. Given that the total response of East Asian
461 Summer Monsoon precipitation to aerosols is dominated by Mode 1 (not shown), which is
462 compensated by weak precipitation increase in response to negative local AOD values in Mode
463 2 (Figs. 1d and 3a), this indicates that historical East Asian Summer Monsoon weakening and

464 associated land drying is likely dominated by local aerosols through the atmospheric radiative
 465 pathway.



466
 467 **Figure 7.** AOD Mode 1 tropical Indo-Pacific response of (a,c,e) 500hPa ω ($Pa s^{-1} \times 100$; color shading),
 468 1000 hPa vector wind ($m s^{-1}$; reference vector in lower right) and precipitation ($mm day^{-1}$; red contours), and
 469 (b,d,f) zonally-averaged ($45^{\circ}-125^{\circ}E$) condensational heating ($^{\circ}C d^{-1}$; color shading) and v, ω vectors (v : m
 470 s^{-1} , reference vector in lower right; ω : $Pa s^{-1} \times 50$) in FULL-AMIP, RAD-AMIP and SST-AMIP regressed
 471 onto AOD PC1. Red contours in (a,c,e) show precipitation regression slopes, starting from $\pm 0.2 mm d^{-1}$ and
 472 contoured at $\pm 0.2 mm day^{-1}$ intervals, with solid lines for positive values and dashed lines for negative
 473 values. Region for the zonally-averaged cross section is outlined in (a,c,e). Areas without stippling are
 474 significant at the 95% confidence level based on a two-tailed Student's t-test. Horizontal wind magnitudes
 475 $< 0.01 m s^{-1}$ are omitted for clarity.

476 *2) Southern Hemisphere Circulation*

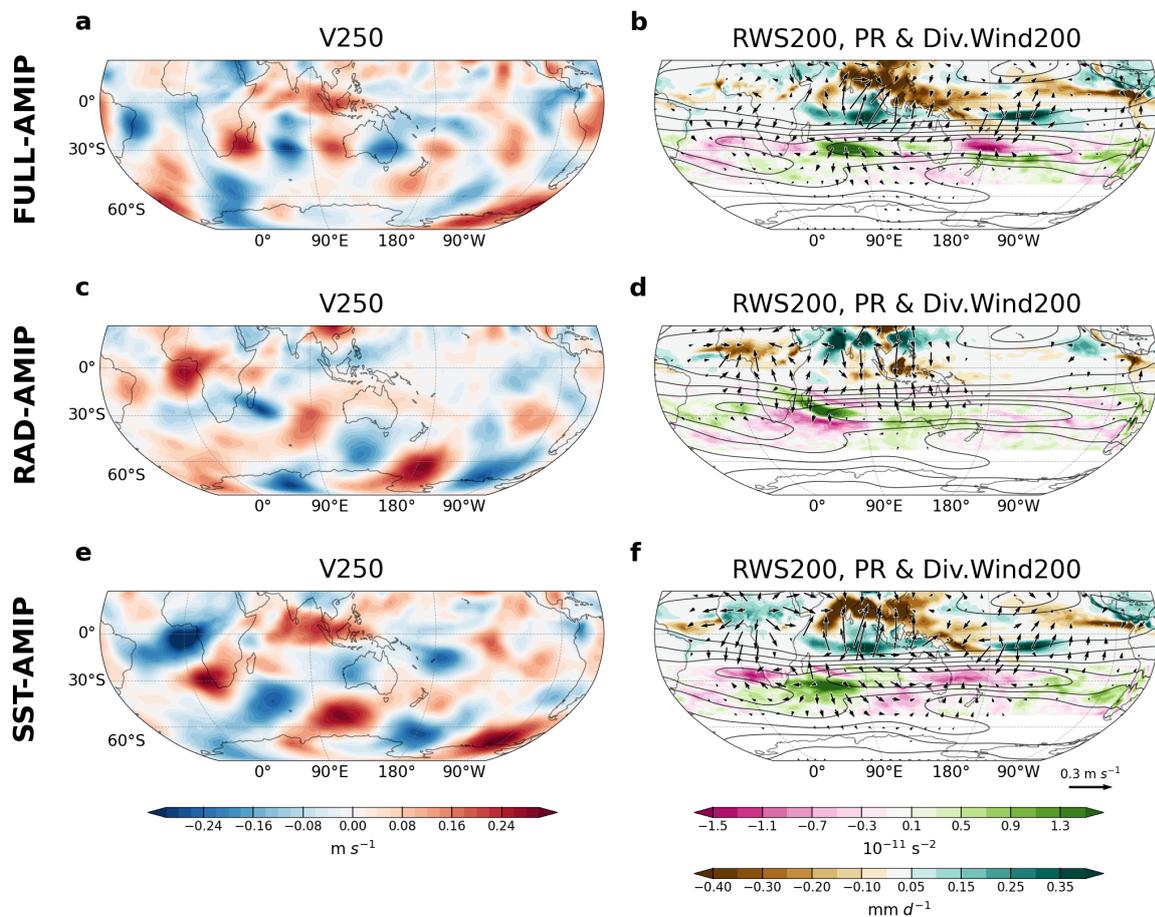
477 A notable aspect of AOD Mode 1 is the SLP response pattern over the SH, which resembles
 478 a Rossby wave train in both RAD-AMIP and SST-AMIP, but with opposite sign (Figs. 2e,f).
 479 In contrast, FULL-AMIP shows a zonally symmetric structure over the SH extra tropics, with
 480 positive (negative) SLP anomalies at high (middle) latitudes (Fig. 2d). SH circulation trends
 481 have been linked to anomalous heating in the tropics which can trigger tropical–extratropical

482 teleconnections (Berbery et al. 1992; Hoskins and Ambrizzi 1993). Guided by this theory, we
483 pose an open question: can low-latitude aerosols, particularly central African biomass burning
484 emissions given their geographic proximity, modulate SH circulation through teleconnection
485 dynamics via an atmospheric radiative pathway, and how does this compare with
486 teleconnections driven by the ocean-mediated response? These SH anomalous upper-level
487 circulations can potentially influence Australian precipitation (Ashok et al., 2003; Cai et al.,
488 2011) and Antarctic climate through the Tropical-Antarctic teleconnection (Ding and Steig,
489 2013; Li et al., 2021).

490 To understand the origin of the Rossby wave train responses, we examine the upper-level
491 circulation and its connection to tropical precipitation. The 250 hPa meridional wind (V250)
492 anomalies clearly delineate poleward propagating Rossby wave trains of opposite sign in
493 response to the radiatively forced and SST-driven responses, which appear to originate in the
494 Gulf of Guinea and propagate southeastward over the southern Indian Ocean and into the
495 Pacific sector of the Southern Ocean (Figs. 8c,e). The SST-driven Rossby wave train takes a
496 slightly more poleward route than the radiatively forced one. FULL-AMIP lacks a poleward-
497 propagating Rossby wave signature due to the large degree of cancellation between RAD- and
498 SST-AMIP (Fig. 8a). Instead, FULL-AMIP shows a zonally-oriented V250 wave train
499 emanating from the tip of South Africa and extending eastward across the Indian and Pacific
500 Oceans along the latitude (30°S) of the subtropical jet maximum, which acts as a wave guide
501 (Fig. 8b).

502 Both the radiatively forced and SST-driven V250 Rossby wave trains appear to be initiated
503 by tropical precipitation and associated diabatic heating anomalies over Africa and the Indian
504 Ocean (Figs. 8d,f). In SST-AMIP, upper-level divergence associated with increased
505 precipitation over the Sahel produces the negative (northerly) V250 anomaly centered over the
506 Gulf of Guinea (Fig. 8f). As the anomalous northerly divergent winds cross the equatorward
507 flank of the climatological subtropical westerly jet, it generates a negative Rossby wave source
508 (RWS) to the west of South Africa, which triggers a downstream Rossby Wave response (Fig.
509 8f). Increased precipitation in the tropical Indian Ocean along 10°S may also act to reinforce
510 the Rossby Wave response. These results are consistent with previous studies which show the
511 critical role of the subtropical jet in amplifying upper-level circulation anomalies emanating
512 from tropical heating (Berbery et al. 1992; Hoskins and Ambrizzi 1993; Li et al., 2015) and
513 serving as a waveguide into the SH extra tropics (Gillett et al. 2021). In RAD-AMIP, upper-

514 level convergence associated with diminished precipitation over the Sahel produces the
 515 positive (southerly) V250 anomaly centered over the Gulf of Guinea (Fig. 8d). Unlike SST-
 516 AMIP however, RAD-AMIP shows a large AOD increase over the African Democratic
 517 Republic associated with local BMB emissions (recall Fig. 1b). The resulting anomalous heat
 518 source from absorbing BMB aerosols is balanced by adiabatic ascent, accompanied by upper-
 519 level divergence (Fig. 8d). The divergent V250 anomaly crossing the equatorward flank of the
 520 subtropical jet over the southwestern Indian Ocean generates a positive RWS anomaly, which
 521 in turn triggers the downstream Rossby wave train response (Fig. 8d).



522

523 **Figure 8.** AOD Mode 1 Southern Hemisphere response of (a,c,e) 250 hPa meridional wind (V250; m s^{-1})
 524 and (b,d,f) 200 hPa Rossby wave source (RWS200; pink-green color bar; 10^{-11} s^{-2}), precipitation (PR; green-
 525 brown color bar; mm d^{-1}) and 200 hPa divergent wind vector (m s^{-1} ; reference vector in lower right) regressed
 526 onto AOD PC1 in FULL-AMIP, RAD-AMIP and SST-AMIP. For clarity, RWS200 (precipitation)
 527 regressions are only shown in the latitude band 52° - 20° S (19° S- 26° N) in FULL-AMIP and SST-AMIP and
 528 52° - 6° S (6° S- 26° N) in RAD-AMIP; wind vectors with magnitudes $< 0.03 \text{ m s}^{-1}$ are omitted. The
 529 climatological 250hPa zonal wind is contoured in black (contour interval = 10 m s^{-1} from 5-45 m s^{-1}).

530 Due to the largely canceling Rossby wave trains, the SH SLP response in FULL-AMIP
 531 shows a more zonally symmetric structure than either RAD-AMIP or SST-AMIP (recall Figs.
 532 2d-f). We do not have a definitive explanation for this pattern of response, other than to

20

533 conjecture that it may be linked to the zonally-symmetric precipitation response in the deep
534 tropics over the Indian and Pacific sectors, and to note that it stems from a subtle balance
535 between radiatively and SST-driven effects, which are not entirely additive (recall Fig. 6).

536 The opposing Rossby-wave-like signatures in RAD- and SST-AMIP and their tropical
537 origins suggest a plausible teleconnection pathway by which low-latitude aerosols can
538 influence SH circulation through anomalies in tropical convection. In RAD-AMIP, the
539 response originates from divergent flow associated with adiabatic ascent induced by absorbing
540 African aerosols and Sahel drying, together with precipitation anomalies over the tropical
541 Indian Ocean, whereas in SST-AMIP it arises primarily from diabatic heating associated
542 African convection. Despite originating from different processes, both pathways generate
543 poleward-propagating Rossby wave trains of opposite sign. Their near cancellation, along with
544 the relatively weak influence of Sahel precipitation anomalies featuring a meridional dipole
545 pattern, may help explain the muted, zonally symmetric SH signal in FULL-AMIP. We
546 reiterate that this is a working hypothesis, and firm attribution requires further verification such
547 as using barotropic linear model experiments forced with the anomalous heating or divergent
548 flow from our simulations to test whether similar wave train responses can be reproduced.

549 **4. Summary and Discussion**

550 There is increasing recognition that the global climate response to anthropogenic aerosols
551 is sensitive to the pattern of regional emissions, which has shifted over time, necessitating a
552 broader view of their role in historical climate change (Persad and Caldeira, 2018; Deser et al.
553 2020; Kang et al. 2021; Wang and Wen, 2022; Persad, 2023; Shi et al. 2023; Dong et al. 2024;
554 Diao et al. 2025). In particular, two distinct modes of AOD variability from industrial sources
555 have been identified over the past century: the “global increase mode” primarily associated
556 with Asian emissions but modulated by emissions over North America and Europe, and the
557 “shift mode” reflecting a zonal redistribution of NH emissions between the western and eastern
558 hemispheres (Kang et al. 2021; Wang and Wen, 2022; Shi et al. 2023; Dong et al. 2024). These
559 AOD modes can produce synergistic and competing effects with GHGs depending on the
560 period of interest, complicating detection and attribution efforts (Dong et al. 2024). The
561 dynamical mechanisms underpinning the global response to the “global increase” and “shift”
562 modes of industrial aerosol emissions have been investigated in idealized coupled modeling
563 studies, revealing the importance of both atmospheric and oceanic processes (Kang et al. 2021;
564 Wang and Wen, 2022; Shi et al. 2023; Diao et al. 2025). However, non-linear interactions

565 between aerosol emissions in different regions complicates a full understanding and
566 interpretation of the combined effects of the two AOD modes (Diao et al. 2025).

567 In this study, we provide additional insight into the pathways by which time-evolving
568 anthropogenic aerosols from combined industrial and BMB sources over the past century
569 influence boreal summer (JJA) climate. In particular, we utilize a novel atmospheric modeling
570 framework to separate the atmospheric radiative and ocean-mediated pathways of response.
571 We decompose the full spatio-temporal evolution of historical aerosol emissions into two
572 leading modes and elucidate the response pathways for each mode.

573 AOD variations arising from industrial and BMB sources can be decomposed into two
574 leading modes, which together explain 94% of global JJA AOD variance during 1930-2030
575 (56% for Mode 1 and 38% for Mode 2). AOD Mode 1 increases monotonically over the first
576 half of the record with little change thereafter, in contrast to Mode 2 which exhibits sinusoidal
577 behavior, with a positive trend from the mid-1930s to the mid-1970s followed by a steep
578 decline to the early 2020s. Mode 1 features positive AOD anomalies at low latitudes, with
579 regional hotspots in East and South Asia associated with industrial emissions and in tropical
580 South Africa, South America, Indonesia and Malaysia associated with BMB emissions,
581 accompanied by weak negative AOD anomalies in western Europe and the eastern US. While
582 Mode 2 is characterized by large positive AOD values in western Europe and the eastern US,
583 juxtaposed against weaker negative values in South and East Asia and eastern Siberia. The two
584 AOD modes are broadly consistent with the “global increase” and “shift” modes reported in
585 prior studies that considered industrial aerosols alone (e.g., Wen and Wang, 2022; Shi et al.,
586 2023; Dong et al., 2024).

587 By spanning through 2030, our results provide additional insight on historical and near-
588 present aerosol variability. Our simulations use RCP8.5 aerosol emission trajectory from 2006
589 onward, which projects a peak of Asian aerosol emissions in the 2010s, followed by a decline
590 in the 2020s-2030s, due to decreasing emissions in Japan offset by increases in other Asian
591 regions (including China; see Fig.6b in Takemura et al., 2012). In contrast, CMIP6 shows
592 higher AOD in China than CMIP5 prior to 2006 (see Fig. 2a in Wang et al., 2014) and also
593 fails to capture China’s AOD decline from 2006 to 2014 (Wang et al., 2021). Meanwhile, in
594 the recent Shared Socioeconomic Pathway (SSP) future scenarios, SSP 3-7.0, which is
595 comparable to RCP 8.5 in terms of the greenhouse gas trajectory, projects China’s aerosol
596 emissions increase through the 2050s (see Fig.2 in Lund et al., 2019). Therefore, though

597 imperfect, the middle-of-road aerosol emission trajectory of RCP 8.5 more closely resembles
598 Asian aerosol emission conditions from 2006 to present compared to the latest CMIP6 and
599 SSPs, and the resulting global and regional AOD estimates remain within the observational
600 error range (IPCC AR6; Lund et al., 2019; Takemura et al., 2012; Wang et al., 2024).

601 In AOD Mode1, the shift of PC1 from a positive slope to a negative slope in the mid 2010s
602 likely reflects the projected Asian aerosol emissions' peak in the 2010s and BMB decline in
603 South America, Southeast Asia (Takemura et al., 2012), and NH mid-latitude (see Fig. 4c in
604 Chemke and Coumou, 2024) under RCP8.5. The difference between RCP8.5 and CMIP6 may
605 help explain why Mode 2 shows smaller Asian AOD changes relative to the mid-latitudes, in
606 contrast to the "shift" mode based on industrial aerosols from 1930-2010 identified by Wang
607 and Wen (2020) using CMIP6, which showed comparable low- and mid-latitude contributions.
608 These imply that regional emission variations, particularly in Asia, are crucial in shaping the
609 dominant modes of global aerosol variability, and can shift the balance between atmospheric
610 radiative and ocean-mediated pathways of aerosols, altering both the strength and spatial
611 pattern of associated global climate responses. This further underscores the need to integrate
612 evolving regional trajectories into future emission scenarios.

613 Having established the leading AOD modes, we next addressed two key questions: through
614 which pathways do aerosols influence climate, and how do these pathways differ across regions
615 and aerosol modes? By separating atmospheric radiative and ocean-mediated pathways, we
616 showed that the ocean-mediated pathway tends to dominate the global large-scale circulation
617 and precipitation responses to both AOD Modes, with offsetting contributions from the
618 atmospheric radiative pathway. However, the interplay between the two pathways is complex
619 and regionally dependent, making it difficult to generalize. NH land surface air temperature is
620 an exception, where the two pathways have distinct roles in each mode. In Mode 1 (low-latitude
621 aerosols), radiative forcing and SST-driven influences are comparable, whereas in Mode 2
622 (mid-latitude aerosols), radiative forcing dominates. This suggests that low-latitude aerosols
623 can perturb local land surface air temperature through both pathways, making them efficient in
624 driving regional changes. In contrast, mid-latitude aerosols primarily drive in-situ surface air
625 temperature changes through the atmospheric radiative pathway. Although mid-latitude
626 aerosols exert weaker local impacts, they have high efficacy in driving remote surface air
627 temperature responses, including contributing to global mean surface air temperature (Shindell
628 and Faluvegi, 2009; Persad and Caldeira, 2018).

629 The tropical Indo-Pacific and SH tropospheric circulation responses to AOD Mode 1
630 exemplify how low-latitude aerosols exert both local and remote influences on circulation
631 through two pathways. The tropical Indo-Pacific sector displays a distinctive meridional dipole
632 response in precipitation and tropospheric overturning circulation. The latitudinal structure of
633 this response represents a hybrid between the broad cross-equatorial scale resulting from the
634 ocean-mediated pathway, associated with remote and local aerosols (Bollasina et al., 2014;
635 Undorf et al., 2018b), and the narrower NH dipolar pattern driven by the atmospheric radiative
636 pathway, which likely reflects the influence of aerosols from adjacent sources (Fig. 5 in Persad,
637 2023). In the SH, both the radiatively forced and SST-driven responses produce poleward-
638 propagating Rossby wave trains emanating from the tropics into the extratropics, but with
639 opposite sign. Their largely canceling responses appear to be initiated by tropical diabatic
640 heating anomalies from convection changes over the Sahel and tropical Indian Ocean in
641 response to remote industrial aerosols, along with adiabatic heating anomalies associated with
642 central Africa BMB aerosols in the atmospheric radiative case. These results suggest a potential
643 aerosol-driven tropical–extratropical teleconnection in the SH, though confirmation with linear
644 models or targeted experiments is needed.

645 Under the transient framework, we showed that the interplay of atmospheric radiative and
646 ocean-mediated effects is neither uniform nor stationary. Radiative effects can exert not only
647 short-lived adjustments but also persistent, decadal-scale influences. This blurs the
648 conventional distinction between “fast” and “slow” responses to radiative forcing, as long-term
649 evolving radiative forcing itself may encompass a slow component. With aerosol emissions
650 expected to decline, and clean air legislation already enacted in some countries, it is critical to
651 account for both atmospheric radiative and ocean-mediated components of aerosol effects
652 when predicting regional hydroclimate and risks such as heat extremes, given the mobility and
653 geography-dependent nature of their nuanced interplay.

654 Much remains to be understood about the mechanisms underlying the atmospheric radiative
655 and ocean-mediated pathways of response to the two AOD Modes documented here, including
656 the relative contributions of local vs. remote aerosol emissions and their mode dependence,
657 warranting further analysis of our experiments. Our study has utilized one climate model
658 (CESM1) and one (CMIP5) representation of historical time-evolving industrial and biomass
659 burning aerosols, both of which have known strengths and shortcomings (e.g., Holland et al.
660 2024). Repeating our approach with other models and updated aerosol emissions inventories

661 would be a useful next step for addressing structural and forcing uncertainty. Models with
662 improved representation of aerosol microphysical processes and aerosol-cloud interactions,
663 enhanced vertical and horizontal resolution, and interactive wildfire capabilities, may be
664 especially valuable in this regard. Applying our protocol to regional aerosol forcing
665 experiments such as those conducted as part of the Regional Aerosol Model Intercomparison
666 Project (Wilcox et al. 2023) may yield additional mechanistic insights, although non-linear
667 interactions between forcings in different regions may complicate the picture (Diao et al. 2025).
668 Additionally, this work focuses on boreal summertime (JJA) signals, particularly those
669 associated with NH monsoon circulations and mid-latitude large-scale precipitations. Although
670 the long-term variation of JJA AOD patterns likely does not differ from that of the annual mean
671 and other seasons, addressing the seasonality of both the forcing and the response (for example,
672 boreal winter as in Diao et al., 2025) would be a worthwhile next step.

673 Looking forward, anthropogenic aerosol emissions are expected to undergo substantial
674 changes. With continued air quality regulation in Asia and growing industrial activity in
675 tropical regions, the center of aerosol emissions may shift to South Asia, South America, and
676 Africa, though the timing of this transition remains uncertain (Scholten et al., 2024). This shift
677 is especially concerning given the high hydrological sensitivity of the tropics and its ability to
678 influence climate worldwide via teleconnections (Smith et al., 2016; Dittus, et al., 2021; Liu et
679 al., 2024). Given the critical role of anthropogenic aerosols in shaping historical changes in
680 regional precipitation over densely populated areas (White et al. 2025), there is an essential
681 need to incorporate information on anticipated near-term and long-term changes in both
682 industrial and biomass burning aerosol emissions into climate projections and risk assessments
683 (Persad et al., 2023). Importantly, understanding the physical pathways by which aerosols
684 impact global climate would not only improve our interpretation of historical climate trends,
685 but also enhance the reliability of future projections in a world increasingly shaped by GHGs
686 and shifting aerosol regimes.

687

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694 *Data Availability Statement.*

695 The CESM1 CAM5 AMIP single-forcing experiment data will be available through a
696 NCAR GLOBUS guest collection. The CESM1 Large Ensemble simulations used in this
697 study are available from <http://www.cesm.ucar.edu/experiments/cesm1.1/LE/#single-forcing>
698 and <http://www.cesm.ucar.edu/projects/community-projects/LENS>.

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