Distilling the evolving contributions of anthropogenic aerosols and greenhouse gases to historical low-frequency surface ocean changes

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Key Points:

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13	٠	Over the past century, GHG forced response is characterized by a single dominant
14		mode while AER response consists of two distinct modes.
15	•	AER's first mode features a monotonic increase in global AER and global cool-
16		ing pattern opposite to the GHG warming pattern
17	•	AER's second mode features the recent shift in AER emissions, which drives re-
18		gional warming amplifying the GHG response

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19 Abstract

Anthropogenic aerosols (AER) and greenhouse gases (GHG) – the leading drivers of the 20 forced historical change – produce different large-scale climate response patterns, with 21 varying trend pattern correlations from negative to positive over the past century. To 22 understand what caused the time-evolving comparison between GHG and AER responses, 23 we apply a joint low-frequency component analysis on global sea-surface temperature and 24 sea-surface salinity response over 1921-2020 from CESM1 single-forcing large ensemble 25 simulations. While GHG response is well-described by its first leading mode, AER re-26 sponse consists of two distinct modes. The first one features global AER increase and 27 global cooling, opposite to GHG-induced warming. The second mode features multidecadal 28 variations in AER distributions, where the recent shift from North America/western Eu-29 rope to southeast Asia emissions drives regional changes enhancing the GHG effect. We 30 argue that AER can have both competing and synergistic effects with GHG, as their emis-31 sions change temporally and spatially. 32

³³ Plain Language Summary

Anthropogenically forced climate change over the past century has been mainly caused 34 by two types of emissions: greenhouse gases (GHG) and aerosols (AER). In general, sul-35 fate aerosols from industrial sources can reflect shortwave radiation to yield a cooling 36 effect opposite to the GHG warming effect. However, model simulations isolating GHG 37 and AER forcings show that the large-scale climate effect of AER does not always dampen 38 the GHG effect. Instead, over recent decades, AER have produced surface ocean response 39 patterns more like the GHG response. Using a novel principle component analysis, we 40 find that aerosols have driven two distinct modes of climate change patterns over the his-41 torical period. The first mode is associated with global aerosol increase, resulting in global-42 wide cooling damping the GHG-induced warming. The second mode is associated with 43 the shift in aerosol emissions from north America/western Europe to southeast Asia, which 44 drives regional changes enhancing the GHG effect. Our results highlight the importance 45 of considering the temporal and spatial evolutions of AER emissions in assessing GHG 46 and AER climate effects and attributing historical anthropogenic climate changes to GHG 47 and AER forcings. 48

49 **1** Introduction

Anthropogenically forced climate change over the past century has been primar-50 ily driven by two components: greenhouse gases (GHG) and anthropogenic aerosols (AER). 51 These components modulate the global-mean surface temperature through distinct ra-52 diative effects (Myhre et al., 2014; Forster et al., 2021) - GHG cause surface warming 53 due to absorption and re-emission of longwave radiation, while AER change energy bud-54 get through reflection or absorption of shortwave radiation by scattering (e.g., sulfate) 55 or absorbing species (e.g., black carbon). Additionally, AER have indirect effects on cli-56 mate through cloud-aerosol interactions, where aerosols can serve as cloud condensation 57 nuclei affecting clouds' albedo, lifetime, and properties (Twomey, 1977; Ackerman et al., 58 2004). Over the past century, long-term increases in global-mean GHG and AER have 59 led to a large cancellation between GHG-induced warming and AER-induced cooling ef-60 fects (Deser et al., 2020). 61

While a clear opposing effect from GHG and AER on global-mean surface temperature has been found, comparing the spatial patterns of their climate responses has been less straightforward. Focusing on global GHG and AER forcings in the 20th century, Xie et al. (2013) found that the first leading modes of climate response patterns to GHG and AER bear a great resemblance, suggesting that large-scale climate responses are governed by the same ocean-atmosphere feedbacks intrinsic to the climate system. Wang et al. (2016) further examined the differences in those leading modes, highlighting the unique features of AER forced response associated with interhemispheric temperature asymmetry and
 cross-equatorial circulation change.

The spatial distributions of GHG and AER forcings add additional complexity to 71 the comparison. Unlike well-mixed GHG, AER emissions have much richer structures 72 in their spatial distributions and temporal evolution (Deser et al., 2020). Emissions from 73 North America (NA) and western Europe (EU) have dominated the global total AER 74 loading since the early decades of the 20th century until the 1970s, after which they have 75 declined substantially following emission regulations. On the other hand, emissions from 76 77 southeast Asia (SA) have been increasing gradually since the 1950s, and more recently, have surpassed the emissions from NA and EU since the 1990s. This transition of ma-78 jor AER sources has been found to cause large-scale climate changes in a different way 79 than global-mean AER change (Kang et al., 2021; Wang & Wen, 2022), and to some ex-80 tent can compensate for the global mean AER effect (Shi et al., 2022). 81

To isolate and quantify the respective contributions of GHG and AER to forced 82 historical climate change, single-forcing large ensemble (SF-LE) simulations within fully-83 coupled global climate models (GCMs) have provided valuable insights. Using CESM1 84 SF-LE, Deser et al. (2020) found that the contributions of GHG and AER to the large-85 scale patterns of total forced trends vary over time, with AER being the dominant driver 86 before the 1970s and GHG dominating thereafter. Wang and Wen (2022) further extended 87 the analysis to CMIP5 multi-model comparisons, highlighting both similarities and dis-88 parities in the spatial patterns of the trends driven by AER and GHG. 89

To recap the literature and to illustrate the evolving contributions of GHG and AER 90 to forced historical trends, we begin by showing the ensemble-mean response in CESM1 91 SF-LE for two 40-year periods, 1940-1980 and 1980-2020. Figure 1 shows the trend pat-92 terns for sea-surface temperature (SST) and sea-surface salinity (SSS), and key atmo-93 spheric variables coupled with them, sea level pressure (SLP) and surface water fluxes 94 (i.e., precipitation minus evaporation, P-E). We compare these trend patterns forced by 95 all forcings ("ALL"; from the CESM1 LE project, Kay et al. 2015), GHG, and AER (from 96 Deser et al. 2020). As found in previous studies, during the early period (1940-1980; Fig. 97 1a), the total forced response is dominated by AER, featuring an inter-hemispheric con-98 trast with pronounced cooling due to the SW absorption by AER and salinification trends qq in the northern hemisphere (NH) driven by an enhancement of Atlantic Meridional Over-100 turning Circulation (AMOC; Menary et al., 2020) and increased sea-ice formation in the 101 Arctic ocean. During recent decades (1980-2020; Fig. 1b), the total forced response has 102 been dominated by GHG, featuring broad global warming, a "wet-get-wetter" precip-103 itation pattern, and a "salty-get-saltier" SSS pattern, consistent with the literature (Held 104 & Soden, 2006; Xie et al., 2010; Durack et al., 2012; Capotondi et al., 2012). 105

Perhaps more interestingly, the climate response patterns driven by GHG and AER 106 largely oppose each other during the early period, but show some resemblance during 107 the later period at regional scales (e.g., NH warming and Arctic freshening). We further 108 compute the pattern correlations for running 40-yr trends in global SST and SSS between 109 the ALL and single forcing (GHG or AER) ensemble-means, following Deser et al. (2020). 110 As expected, the pattern correlations between ALL and GHG for both SST and SSS have 111 increased steadily since the 1950s (red lines in Figs. 1c, d), suggesting the increasingly 112 dominant role of GHG in modulating global climate. However, the pattern correlations 113 between ALL and AER decreased only over the first half of the 20th century and have 114 gradually rebounded over recent decades (blue lines in Figs. 1c, d). The same non-monotonic 115 behavior is also found for the pattern correlation between GHG and AER (black lines 116 117 in Figs. 1 c, d), indicating that the surface ocean response patterns to AER have become closer, not opposite, to those forced by GHG over recent decades. 118

This comparison between the evolving AER and GHG forced responses led us to ask: what caused the AER response to change over time, and particularly, to amplify



Figure 1. CESM1 ensemble-mean trend response to ALL, GHG and AER forcings, for (a) 1940-1980 and (b) 1980-2020. Black contours overlaid on the SST panels are SLP trends (contour interval is 0.16 hPa/40yr, zero contours are thickened, solid contours denote positive SLP trend, dashed contours denote negative SLP trend). Colored contours overlaid on the SSS panels are P-E trends (contour interval is 0.18 mm/day/40yr, zero contours are omitted; green denotes positive P-E and red denotes negative P-E). (c, d) Pattern correlations for 40-yr running trends in ensemble-mean (d) global SST and (e) global SSS between ALL, GHG and AER.

the GHG response over recent decades? Is it because of the decline of global AER emissions, or is it because of the change in the spatial distribution of those emissions, or both? And what implications does this have for detecting and attributing historical low-frequency surface ocean changes? Motivated by these questions, in this work we apply a principal component analysis to investigate the leading modes of low-frequency historical surface ocean changes driven by GHG and AER, and distill their evolving contributions to historical forced climate change.

¹²⁸ 2 Low-frequency component anlaysis

One of the caveats in the trend pattern analysis in Fig. 1 and other studies (e.g. 129 Deser et al., 2020; Kang et al., 2021) is that these trends are computed over arbitrary 130 time intervals, thus may not capture the whole series of the forced response and time-131 evolving forcing patterns. To achieve a more systematic assessment, other prior stud-132 ies have utilized principal component (PC) analysis to linearly separate the total response 133 into several empirical orthogonal functions (EOFs) (e.g., Xie et al., 2013; Wang et al., 134 2016; Bonfils et al., 2020). However, the EOF results can be affected by high-frequency 135 natural variability (e.g., ENSO) that is not completely removed due to insufficient en-136 semble size. Therefore, to robustly examine the *low-frequency forced* response, we ap-137 ply a low-frequency component analysis (LFCA; Wills et al. 2018) to the ensemble mean 138 GHG and AER forced response from CESM1 SF-LE. In the following sections, we first 139 introduce the LFCA method and the data analyzed (section. 2.1), and we next show the 140 results of the leading low-frequency modes for the GHG and AER forced response (sec-141 tion 2.2). 142

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2.1 The LFCA method and data

Unlike conventional PC analysis which maximizes total variance, LFCA finds a linear combination of EOFs that maximize the ratio of low-pass filtered variance to total variance, thereby isolating the leading modes of low-frequency variability (Wills et al., 2018). One can also apply LFCA to multiple spatial-temporal fields jointly, similar to the joint EOF analysis. Previous studies (Wills et al., 2022; Dörr et al., 2023; Bonan et al., 2023) suggest that the use of joint LFCA to account for low-frequency co-variability in multiple fields improves the isolation of long-term forced response.

Therefore, in this study we perform joint LFCA on global monthly SST and SSS 151 anomalies over 1921 – 2020 to study low-frequency modes of forced historical surface ocean 152 changes. The anomalies are relative to the 1921-2020 climatology. We take the SST and 153 SSS response from the ensemble-mean of ALL, GHG and AER ensembles as part of CESM1 154 SF-LE (Deser et al. 2020), each containing 20 members. Note that original simulations 155 in CESM1 SF-LE used the "all-but-one" forcing scenario – that is, all historical radia-156 tive forcing agents are prescribed except their GHG or AER is fixed at the 1920 condi-157 tions. The net effect of GHG and AER can be then obtained by subtracting these "all-158 but-one" SF simulations from the standard ALL simulations. We also note that in this 159 set of LEs, AER specifically refers to industrial aerosols, not including biomass aerosols. 160

For the joint LFCA, we use a 15-year cut-off low-pass filter to isolate multidecadal low-frequency variability, and we retain the 10 leading EOFs, which in total account for 99.9%, 99% and 97% of the joint low-frequency variance for ALL, GHG and AER, respectively. Additionally, to understand the dynamical processes associated with each mode, we regress monthly SLP, precipitation (P), evaporation (E) and P-E anomalies onto the timeseries of each of the PCs.

Although we take the ensemble mean response first to remove random internal variability before performing the joint LFCA, one can also perform LFCA on each ensemble member to obtain (the best estimate of) its low-frequency forced response and then



Figure 2. Leading low-frequency modes of GHG and AER ensemble-mean responses obtained from joint SST/SSS LFCA. Numbers in the corner of the timeseries plots show the low-frequency variance explained by the corresponding PC. SLP regressions are overlaid on the SST patterns (contour interval is 0.05 hPa, zero contours are thickened, solid contours denote positive SLP and dashed contours denote negative SLP); P-E regressions are overlaid on the SSS patterns (interval is 0.06 mm/day, zero contours are omitted, green denotes positive P-E and red denotes negative P-E). Regressions of aerosol optical depth at the 550nm (no unit, multiplied by 100) onto AER PC1 and PC2 are shown in panels (g) and (k).

average the leading modes across ensemble members (e.g., Wills et al., 2022; Kuo et al.,
2023). We will show in the next section that the results remain the same regardless of
which method is used (cf. Fig. 2 and Fig. S2)

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2.2 Low-frequency modes of GHG and AER forced responses

Figure 2 shows the leading joint LFCA modes of SST and SSS for the GHG and AER forced responses. The GHG forced response is dominated by a single mode that explains 97.4% of the low-frequency variance; we will denote its timeseries as "GHG PC1" hereafter. The AER forced response consists of two leading modes, which both have strong multi-decadal variability and explain 75.2% and 13.9% of the total low-frequency variance, respectively. We will denote their timeseries as "AER PC1" and "AER PC2" hereafter.

The GHG PC1 has increased monotonically throughout the past century, with a 181 pronounced positive trend starting from 1980 (Fig. 2a). The corresponding SST pattern 182 is characterized by broad global warming and enhanced tropical eastern Pacific warm-183 ing (i.e., the El Niño-like SST pattern), accompanied by a reduced tropical zonal SLP 184 gradient (Fig. 2b). The global-scale warming causes precipitation to increase in the trop-185 ics and decrease in the broad subtropics (Fig. S1a), following the "wet-get-wetter" and 186 "warmer get wetter" mechanism (Held & Soden, 2006; Xie et al., 2010). Associated with 187 enhanced global-scale evaporation (Fig. S1b), the net P-E pattern links to a "salty-get-188 saltier" SSS pattern (Fig. 2c Durack et al., 2012; Sun et al., 2021), with an enhanced 189

SSS gradient between tropical and subtropical oceans as well as an amplified climato logical contrast between the Pacific and Atlantic basins.

On the other hand, AER PC1 and PC2 feature distinct surface ocean patterns and 192 time evolutions (Figs. 2d-k). By regressing aerosol optical depth (AOD) onto PC1 and 193 PC2, we find that the first mode is associated with a globally increasing AOD pattern, 194 with the largest source in east Asia (Fig. 2g). The positive AOD anomalies cause global-195 scale SST cooling by reflecting SW radiation, which is most pronounced in the north Pa-196 cific downstream of the east Asian AOD source (Fig. 2e). The AOD-induced surface cool-197 ing further reduces precipitation in east Asia extending to the north Pacific (Fig. S1c), 198 along with the weakly decreased evaporation (Fig. S1d), leading to increased SSS in the 199 north Pacific (Fig. 2f). Additionally, global cooling reduces runoff into the Arctic Ocean 200 from sea ice melting, thereby increasing its SSS. Overall, AER PC1 increases through 201 most of the 20th century, with the strongest positive trend from 1940 to 1980 and a neg-202 ligible trend after 1980 (Fig. 2d). 203

By contrast, AER PC2 is associated with the relative difference in AOD between 204 northeast America/western Europe and southeast Asia (Fig. 2k). This PC has a neg-205 ative trend from the early 20th century to 1970 and a reversed (positive) trend from 1980 206 to present day (Fig. 2h), reflecting the transition of major AER emissions from NA/EU 207 to SA over the course of the 20th century. In the positive phase of PC2, the negative AOD 208 in NA/EU drives NH SST warming confined to mid-to-high latitudes (Fig. 2i) and Arc-209 tic freshening via increased runoff from sea-ice melting (Fig. 2j). The positive AOD in 210 SA drives weak cooling (Fig. 2i) and drying locally in the Indo-Pacific ocean (Fig. S1e), 211 which excites a Rossby wave response weakening the Aleutian Low (Fig. 2i; also see Smith 212 et al. (2016); Dittus et al. (2021)). Due to the zonal-mean energy budget constraint, the 213 ITCZ shifts northward towards the warmer NH as required by cross-equatorial heat trans-214 port (Kang et al., 2008; Hwang et al., 2013), resulting in enhanced precipitation north 215 of the equator and reduced precipitation in the south (Fig. S1e). This zonal-mean pre-216 cipitation dipole pattern further links to a meridional SSS gradient in the tropical Pa-217 cific, with decreased (increased) SSS in the north (south) (Fig. 2j). 218

In summary, the leading low-frequency modes of GHG and AER forced responses 219 are diverse in both their spatial patterns and temporal evolutions. The GHG response 220 can be largely captured by a single leading mode, which has increased monotonically through-221 out the past century. The AER forced response, however, features two distinct modes. 222 AER PC1 is associated with increasing global AER emissions and resulting global cool-223 ing as well as enhanced regional responses in the NH western Pacific associated with east 224 Asian emissions. AER PC2 represents a multidecadal variation in AER distribution, high-225 lighting the emission shift from high-latitude NA/EU to low-latitude SA over recent decades. 226 This mode features an inter-hemispheric SST gradient and a shift in zonal-mean precip-227 itation and SSS anomalies. The spatial patterns of the two AER modes are consistent 228 with previous studies; however, their relative order and temporal characteristics may de-229 pend on the period analyzed. For example, when accounting for a longer period includ-230 ing the 21st century, Gu et al. (2024) find that the AER shift mode (our 2nd mode) is 231 the leading PC, followed by the AER global mode (our 1st mode). 232

3 Time-evolving contributions of the leading modes of AER and GHG responses

Having quantified the leading modes of AER and GHG responses, in this section, we aim to attribute the changes in the total forced response to these individual modes to gain physical insights.

First, we approximate the time-varying forced responses using their leading LFCA modes, and perform the pattern correlation analysis for running 40-yr trends in global



Figure 3. Pattern correlations for running 40-yr trends in (left) global SST and global SSS (right) response reconstructed using the LFCA leading modes, between (a, b) ALL and GHG or AER responses and (c, d) between GHG and AER responses.

SST and SSS as in Fig. 1 but use the PC-reconstructed responses. We find that using
the first leading mode for GHG response and the leading two modes for AER and ALL
response can faithfully reproduce the results of the simulated total response (cf. Fig. 3
and Fig. 1 c, d). The PC-based pattern correlations capture the increasingly high values between ALL and GHG and the non-monotonic evolution of the correlations between
ALL and AER and between GHG and AER for both SST and SSS trend patterns.

Having verified that the LFCA modes can sufficiently reproduce the total response, now we come back to the questions raised at the beginning: what caused the AER response to change over time and to amplify (rather than offset) the GHG response over recent decades? More specifically, is it because of the change in AER PC1 associated with global AER emissions or the change in PC2 associated with shifting AER emissions, or both?

To answer these questions, we repeat the pattern correlation analysis with individ-252 ual AER PCs. When only accounting for AER PC1, the pattern correlations between 253 ALL and AER decrease monotonically after the 1940s (Figs 3a and b, blue circled lines). 254 unlike the total AER response which bounces back after the 1960s (blue solid lines). Fur-255 thermore, the pattern correlations between AER PC1 and GHG PC1 stay at a constant 256 negative value of -0.88 for SST and -0.69 for SSS (Figs. 3c and d, black circled lines), 257 indicating that the response patterns associated with AER PC1 have continuously op-258 posed those driven by GHG PC1. The high (albeit negative) pattern correlations between 259 GHG PC1 and AER PC1 can already be seen in Fig. 2: both modes feature a global-260 wide SST response driven by global forcing (with sign reversed), and a similar hydro-261 logical cycle response constrained by global warming or cooling, all consistent with ear-262 lier findings by Xie et al. (2013). 263

Turning to AER PC2, although this mode has a much weaker pattern correlation 264 with GHG PC1 overall, the correlation switches sign from negative to positive around 265 mid-century (Figs. 3c and d, black dashed lines), suggesting that it is AER PC2, not PC1, 266 that makes the total AER forced response patterns more similar to the GHG response 267 patterns (Figs. 3c and d, black solid lines). The abrupt change in the running trend pat-268 tern correlations arises from the phase transition in the AER PC2 timeseries around the 269 1980s associated with the shift in major AER emissions from NA/EU to SA. As AER 270 emissions increase over SA and decrease over NA/EU after the 1980s, this mode produce 271 north Pacific warming, Arctic freshening and SH subtropical drying, similar to the GHG-272 induced local changes (Fig. 2), thus making the AER forced response more like the GHG 273 response. 274

To further illustrate the different contributions of AER PC1 and PC2 to the to-275 tal forced response, we show the trend patterns for AER and GHG PCs over 1940-1980 276 and 1980-2020 (Fig. 4). First, the PC-based trend patterns (Fig. 4, left two columns) 277 are remarkably consistent with the actual simulated trend patterns (Fig. 1), confirm-278 ing that using the leading modes can reproduce the total forced response. Next, turn-279 ing to the trend patterns for AER PC1 and PC2 individually, we find that AER forced 280 trends over 1940-1980 arise primarily from PC1 (Fig. 4a) and over 1980-2020 arise nearly 281 entirely from PC2 (Fig. 4b). Moreover, the increase in global AOD from the early 20th 282 century to the 1980s is predominately associated with PC1 and the moderate decline in 283 global AOD afterward is caused mainly by PC2 (Fig. S3). 284

Overall, these findings suggest that both PC1 and PC2 make significant contributions to the AER response, but their roles vary in time. Over the first half of the 20th century until the 1980s, PC1 dominates the total AER forcing and the forced response, with patterns largely *opposite* to those of GHG. If the spatial pattern of AER emissions had remained unchanged from that of PC1 but continued to increase in magnitude over the past 40 years, the GHG response would have been largely compensated for by AER, resulting in a less detectable anthropogenically forced signal over the past century. How-



Figure 4. Similar to Fig. 1, except for the linear trend patterns associated with GHG and AER PCs for (a) 1940-1980, and (b) 1980-2020.

ever, the geographical distribution of AER emissions did change, shifting from NA/EU to SA, which led to the prominent phase transition in PC2 over recent decades. Although PC2 is associated with small global-mean AOD anomalies, the dynamical response associated with this AER shift mode can be large at regional scales. Some of the regional responses appear to *enhance* the GHG-induced changes, leading to a synergistic effect of AER with GHG over recent decades rather than a competing effect as in earlier periods.

²⁹⁹ 4 Implications for detection and attribution

The time-evolving similarities and disparities between GHG and AER forced responses have important implications for detecting and attributing (D&A) historical climate change. Previous D&A studies, e.g. Bonfils et al. (2020), used an EOF approach with historical simulations and identified two externally forced fingerprints. They argued that the first one, featuring global warming and intensified wet-dry patterns, is driven by GHG, and that the second one, featuring an inter-hemispheric temperature contrast and meridional shift in ITCZ location, is driven by AER.

Using the SST/SSS joint LFCA, we also find two leading modes in the CESM1 ALL 307 ensemble-mean response (Fig. S4), similar to the results of Bonfils et al. (2020). Indeed, 308 at first glance, ALL PC1 and PC2 seem to bear a strong resemblance to GHG PC1 and 309 AER PC1, respectively (Fig. 2). However, there are substantial differences between the 310 patterns of ALL PC1 and GHG PC1. For example, GHG PC1 is characterized by a strong 311 El Niño-like SST warming pattern (Fig. 2b), a zonal-mean wet-dry hydrological pattern 312 (Fig. S1a) and a corresponding zonal SSS pattern (Fig. 2c). By contrast, ALL PC1 has 313 a more uniform tropical SST pattern (Fig. S4b), a zonally-asymmetric precipitation pat-314 tern with drying in the west Pacific and wetting in the central Pacific (Fig. S5a), and 315 a resulting SSS dipole pattern with increased salinity in the western Pacific (Fig. S4e). 316 These mismatches, however, appear to be consistent with AER PC1, with a high pat-317

tern correlation of -0.7 for global SST between ALL PC1 (Fig. S4b) and AER PC1 (Fig. 2e). Similarly, while the response patterns of ALL PC2 are overall anti-correlated with AER PC1, there are noticeable spatial features that cannot be explained by AER PCs but rather resemble GHG PC1. Collectively, this suggests that the two modes of historical anthropogenic fingerprints obtained from CESM1 ALL simulations are forced by the combined effects of GHG and AER, rather than by each forcing agent individually as previously proposed (Bonfils et al., 2020).

Thus, we argue that previous D&A approaches that separate the leading modes of historical fingerprints to GHG and AER may be biased, as they don't account for the two distinct modes of AER response and their evolving synergistic and competing climate effects with GHG. Our CESM1 results suggest that robustly detecting and attributing historical forced climate change requires careful separation of GHG and AER responses, which are not mutually independent.

331 5 Summary

In this study, we have analyzed the leading modes of low-frequency climate responses 332 to GHG and AER forcing and distilled their respective contributions to historical forced 333 climate change in CESM1. While the GHG response can be well represented by a sin-334 gle dominant mode, the AER response features two distinct modes. The first mode is 335 associated with an increase in global AER emissions over the past century, driving global-336 wide cooling and regional "wet-get-wetter" precipitation and "salty-get-saltier" salinity 337 response patterns. This AER mode is spatially anti-correlated with the leading GHG 338 mode, largely offsetting impacts from GHG, with some notable regional exceptions such 339 as the western Pacific close to the source of east Asian AER emissions. The second AER 340 mode is associated with a spatial redistribution of AER, featuring the shift of major emis-341 sions from north America/western Europe to southeast Asia over recent decades. This 342 zonally asymmetric AER forcing pattern, however, yields meridional shifts in the zonal 343 mean response of SST, hydrological and SSS, owing to large-scale energy budget con-344 straints. Although this mode has a weaker correlation with the GHG response, the tran-345 sition of this mode from a negative to positive phase over recent decades results in re-346 gional anomalies that can enhance the GHG-induced changes. 347

While our analysis has focused on the past century, the results and conclusions may 348 change for future forcing scenarios. As global AER emissions are projected to decrease 349 with clean-air efforts, AER PC1 will likely change sign in the future. Given the anti-correlation 350 between AER PC1 and GHG PC1, the future AER effect may exacerbate GHG-induced 351 climate change and enhance the detectability. Hence, it remains to be further investi-352 gated how our proposed framework will evolve in the future, accounting for various un-353 certainties arising from model structures, emission scenarios, and representations of AER 354 (direct and indirect) forcings. 355

356 Open Research

The CESM1 Large Ensemble data (Kay et al., 2015) and single-forcing large ensemble data (Deser et al., 2020) are available from https://www.cesm.ucar.edu/community -projects/lens and https://www.cesm.ucar.edu/working-groups/climate/simulations/ cesm1-single-forcing-le. Code to perform LFCA (Wills et al., 2018) is available on Github https://github.com/rcjwills/lfca.

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Supporting Information for "Distilling the evolving contributions of anthropogenic aerosols and greenhouse gases to historical low-frequency surface ocean changes"

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1. Figures S1 - S5



Figure S1. Precipitation (left) and evaporation (right) regressions onto GHG PC1 (top), AER PC1 (middle) and AER PC2 (bottom). Units: mm/day/std.



Figure S2. Results of LFCA applied to each of the individual ensemble members. (left) PC time series. Grey lines are individual members; black lines and color filling are the average of all members; red lines are the LFCA results applied to the ensemblemean response as in Fig. 2. (middle) SST and (right) SSS patterns are averaged over all individual members' PC patterns.



Figure S3. Global-mean AOD anomalies (relative to 1921-2020 climatology) from AER simulations (black) and PC reconstructions (blue and orange). Red dashed line denotes the sum of PC1 and PC2 associated AOD anomalies.



Figure S4. Same with Fig. 2 except for the two leading modes obtained from CESM1 ALL ensemble-mean responses.



Figure S5. Regressions of (left) precipitation and (right) evaporation onto ALL PC1 and PC2.