



Simulated aerosol mixing-state response to emission reductions over China

Jinzu Fu¹, Chloe Yuchao Gao¹, and Mingxinyu Lu¹

¹Department of Atmospheric and Oceanic Sciences & Shanghai Key Laboratory of Ocean-Land-Atmosphere Boundary Dynamics and Climate Change, Fudan University, Shanghai, China

Correspondence to: Chloe Yuchao Gao (gyc@fudan.edu.cn)

Abstract. The internal mixing state of aerosol particles governs their hygroscopicity, removal and activation to cloud droplets, yet assessments of emission reductions rarely resolve it. When emissions of sulfur dioxide and black carbon (BC) are reduced simultaneously, the remaining sulfate may either concentrate on the surviving BC-containing particles or redistribute to BC-free particles, with different consequences for coating thickness and cloud condensation nuclei. We examine this response with GISS ModelE2.1–MATRIX-VBS in two simulations with identical meteorology differing only in anthropogenic emissions over China, where SO₂, BC and organic carbon emissions are lower by 86, 83 and 66 % over 2031–2050. The sulfate column over China decreases by 24 %, because the region changes from a net exporter to a net importer of sulfate, and the decrease extends across the North Pacific to the Arctic. Sulfate is removed about three times faster from BC-containing than from BC-free particles, at the source and downwind, and the share of column sulfate on BC-containing particles over China decreases from 35 to 28 %. A closed process budget attributes this to the reduced number of BC cores available to receive sulfate: condensation and coagulation onto them weaken by about 70 %. Boundary-layer particle number over China decreases by 26 % and BC-containing particle number by 65 %, whereas the remaining coated particles grow. Cloud droplet number decreases by 29 %, mainly through loss of BC–organic-carbon particles. Particle number, size and mixing state therefore change more than bulk mass, and mass-based assessments do not resolve them.

1 Introduction

Anthropogenic aerosols are a leading source of uncertainty in climate projections, and the internal mixing state of aerosol particles, that is, how a given mass of each component is distributed across the particle population, is a first-order control on how that aerosol behaves (IPCC, 2021; Bellouin et al., 2020; Bauer et al., 2008; Bond et al., 2013; Riemer et al., 2019). Mixing state regulates hygroscopicity, particle solubility, the efficiency of wet removal and hence lifetime, and the number of particles available to act as cloud condensation nuclei. Most studies of emission-driven aerosol change nevertheless report bulk abundance or population-mean properties, so the response of mixing-state structure itself to emission perturbations remains comparatively unexplored.



Anthropogenic aerosol and precursor emissions over China have fallen substantially since 2013 under successive air-quality and carbon-emission policies (Zhang et al., 2019), and the projections used here continue that decline. East Asian aerosol is carried eastward across the Pacific by the mid-latitude westerlies and contributes to aerosol loading over the North Pacific and North America (Heald et al., 2006; Yu et al., 2012), and regional emission perturbations more generally produce non-local responses in aerosol, temperature and circulation (Westervelt et al., 2020; Liu et al., 2023; Shao et al., 2024). A reduction in Chinese emissions should therefore be felt along the whole trans-Pacific outflow, but how the composition and mixing state of that outflow change, as opposed to its mass, has not been established.

Most climate models rely on globally harmonized inventories such as CMIP6-CEDS for future simulations, which do not resolve China's rapid and spatially heterogeneous emission changes. CEDS overestimates Chinese PM_{2.5} and SO₂ around 2015 and does not capture the full 2015–2019 decline, with consequences for projected aerosol burden (Zhang et al., 2021; Fan et al., 2022). The Dynamic Projection model for Emissions in China (DPEC) links GCAM-China to emission scenarios defined for China and therefore provides a more detailed basis for isolating the effect of emission changes over China (Tong et al., 2020; Cheng et al., 2021).

Aerosol mixing state evolves through gas–particle partitioning, condensation and coagulation (Gao et al., 2017, 2024). These processes modify composition, size distribution and particle number, and they determine which particles carry which components. When sulfur dioxide and black carbon emissions are reduced together, the outcome for mixing state is not obvious in advance. Less sulfur dioxide means less condensable sulfuric acid for every particle, while less black carbon means fewer carbonaceous particles to receive it, so the sulfate that remains may end up more concentrated on the surviving black-carbon-containing particles, or more of it may stay on particles that contain no black carbon at all. Which of these happens determines how thickly the remaining black carbon is coated, how many particles are large and soluble enough to nucleate cloud droplets, and what composition those particles have.

We address three questions. First, how large is the aerosol response to a rapid reduction of emissions over China, how far does it extend, and how closely does it follow the emission change? Second, how is the remaining sulfate reallocated among particle populations, and which microphysical processes control that reallocation? Third, how do particle number and size change in each population, and what does that imply for cloud droplet formation?

To address these questions we drive GISS ModelE2.1–MATRIX-VBS with DPEC projections in a sensitivity experiment that replaces emissions only over China. Horizontal winds are nudged to the same prescribed fields in both simulations and sea-surface temperature is prescribed, so the two simulations share their meteorology and their difference isolates the chemical and microphysical response to the emission change. The scope of the analysis is deliberately restricted to the prognostic composition, number and mixing-state fields of MATRIX-VBS and to the aerosol activation and cloud droplet number diagnosed from them. We report no aerosol optical depth, absorption, radiative forcing or other aerosol optical or radiative quantity, and we draw no conclusions about the radiative consequences of the changes described here (Sect. 2.7). Section 2 describes the model, the emission data, the experimental design and the diagnostics. Section 3 presents and



discusses the bulk, mixing-state, number, activation and cloud responses and their implications for the assessment of emission changes. Section 4 concludes and states the limits of the design.

2 Model and experiments

65 2.1 GISS ModelE2.1–MATRIX-VBS

We use the NASA GISS ModelE2.1 Earth system model (Kelley et al., 2020) with the MATRIX-VBS (Multiconfiguration Aerosol Tracker of mixing state with Volatility Basis Set) aerosol scheme (Gao et al., 2017). MATRIX, derived from the multiconfiguration information tracker of Bauer et al. (2008), resolves internally and externally mixed states of sulfate, nitrate, organic carbon, black carbon, mineral dust and sea salt, together with the mass and number concentration of each
 70 population. Compositions that coexist within a single size range are recorded as separate configurations and updated at every time step through gas–particle interaction, intra- and inter-population mixing, and dry and wet deposition.

MATRIX-VBS couples this mixing-state representation to a volatility basis set that treats the gas–particle partitioning and chemical ageing of semivolatile organic compounds explicitly (Gao et al., 2017). Organic material is distributed among volatility bins, and condensation, oxidative ageing and multiphase reaction are parameterized within each bin (Donahue et al.,
 75 2006). Primary organic aerosol is therefore treated as semivolatile and condensable rather than as inert mass, secondary organic aerosol is followed in both externally and internally mixed states, and particle growth is represented through the condensation of low-volatility vapours. The 15-population implementation of Gao et al. (2024) adds semivolatile organic carbon to every major population except the sulfate Aitken mode, allowing organic aerosol to evaporate, be transported and recondense. Relative to a bulk or population-mean scheme, this framework resolves which particles carry which components,
 80 and therefore particle number and composition as well as mass (Bauer et al., 2008; Gao et al., 2017, 2024). The number of particles of each population that activate to cloud droplets is computed from the simulated size and composition of that population following Abdul-Razzak and Ghan (2000), as in Bauer et al. (2008).

ModelE2.1 is run in its atmosphere-only configuration at $2^\circ \times 2.5^\circ$ horizontal resolution with 40 vertical layers extending from the surface to 0.1 hPa. Gas-phase chemistry uses the CBM-IVD mechanism.

85 2.2 Aerosol populations and diagnostics

Model output follows a population_component naming convention: the suffixes _SU, _BC, _OC and _ALL denote the sulfate, black carbon, organic carbon and total mass carried within a given population. ACC_SU is thus sulfate residing in the accumulation-mode sulfate population, and BOC_SU is sulfate residing in the black-carbon–organic-carbon population. Table 1 defines the populations used in this study. Throughout the paper we distinguish between the total sulfate column,
 90 which sums sulfate over all populations, and the sulfate carried by an individual population; the two need not change in proportion, and the distinction is central to our results.



We group the populations by whether they contain black carbon. The BC-containing populations are BC1, BC2, BCS and BOC; of these, BOC carries more than 97 % of the sulfate and about 90 % of the black carbon in every region considered. The BC-free populations are AKK, ACC, OCC, OCS and the dust and sea-salt populations. The fully mixed population MXX is dominated by dust and sea salt and is assigned to neither group; it carries about 1 % of the sulfate column. Because MATRIX-VBS carries organic aerosol in nine volatility bins in addition to a non-volatile organic-carbon variable, we state the convention explicitly. The organic carbon of a population is the sum of its non-volatile `_OC` variable, where one exists, and all nine volatility bins (OCM2, OCM1, OCM0 and OCP1–OCP6) of that population. All of these variables are archived in the same units and no additional scaling is applied to any of them.

The number concentration of each population is carried by the corresponding MATRIX number tracer, `N_<POP>_1`, archived as a three-dimensional field that gives particles per kilogram of air once the scale factor of 10^{-10} carried in the file metadata is applied. With this conversion the simulated boundary-layer number concentration over China in Control is about $1.4 \times 10^4 \text{ cm}^{-3}$, of which the Aitken mode contributes two thirds. The number-weighted mean dry and ambient diameters of each population are archived as three-dimensional fields, and the number of particles of each population activated to cloud droplets is archived as a three-dimensional field whose absolute normalization we do not rely on; activation is therefore reported only as relative changes and as shares among populations.

Table 1. MATRIX-VBS aerosol populations and diagnostic variables used in this study.

Population	Composition	Major components tracked	Description
AKK	Sulfate Aitken mode	SO_4^{2-}	Newly formed or small sulfate particles that may grow into ACC.
ACC	Sulfate accumulation mode	SO_4^{2-} ; semivolatile OC	Accumulation-mode sulfate background; ACC_SU denotes sulfate in this population.
OCC	Organic-carbon population	Non-volatile OC, SO_4^{2-} ; semivolatile OC	Organic-rich population representing externally or weakly mixed organic aerosol.
BC1	Fresh BC (inorganic coating volume fraction < 5 %)	BC, SO_4^{2-} ; semivolatile OC	Fresh or weakly coated BC, used to diagnose an externally mixed tendency.
BC2	Condensation-aged BC (coating > 5 %)	BC, SO_4^{2-} ; semivolatile OC	More aged and soluble BC formed by inorganic condensation and coating.
BCS	Coagulation-aged BC	BC, SO_4^{2-} ; semivolatile OC	Sulfate-coated BC formed mainly by coagulation of BC with sulfate populations.
BOC	Black-carbon–organic-carbon mixed population	BC, non-volatile OC, SO_4^{2-} ; semivolatile OC	Carbonaceous mixed population emphasized here; BOC_SU is sulfate in BOC particles.
OCS	Organic-carbon–sulfate mixed population	Non-volatile OC, SO_4^{2-} ; semivolatile OC	Organic–sulfate population used to compare sulfate responses in ACC and BOC.
SSA	Sea-salt accumulation mode	Sea salt, SO_4^{2-} ; semivolatile OC	Fine sea-salt particles that may carry sulfate and organic coatings.
SSC	Sea-salt coarse mode	Sea salt, SO_4^{2-} ; semivolatile OC	Coarse sea-salt population retained for complete coarse-mode representation.
DD1	Dust accumulation mode (coating < 5 %)	Mineral dust, SO_4^{2-} ; semivolatile OC	Relatively fresh fine-mode dust.
DD2	Dust coarse mode	Mineral dust, SO_4^{2-} ; semivolatile OC	Relatively fresh coarse-mode dust.



Population	Composition	Major components tracked	Description
DS1	(coating < 5 %) Dust accumulation mode (coating > 5 %)	Mineral dust, SO_4^{2-} ; semivolatile OC	Aged or soluble fine dust formed by inorganic coating.
DS2	Dust coarse mode (coating > 5 %)	Mineral dust, SO_4^{2-} ; semivolatile OC	Aged or soluble coarse dust formed by inorganic coating.
MXX	Fully mixed population	BC, non-volatile OC, mineral dust, sea salt, SO_4^{2-} ; semivolatile OC	Complex internally mixed particles not assigned to a more specific population.

Note: NH_4^+ , NO_3^- and H_2O are omitted from the major-component column because MATRIX allocates these inorganic constituents to sulfate-containing populations through thermodynamic equilibrium. Definitions follow the original MATRIX and MATRIX-VBS configurations (Bauer et al., 2008; Gao et al., 2017, 2024).

2.3 Emission inventories

2.3.1 CEDS baseline

The Community Emissions Data System (CEDS) is an open global inventory providing annual, sector- and country-resolved emissions of the major aerosol precursors and greenhouse gases (Hoesly et al., 2018; McDuffie et al., 2020), and is widely used as the common historical and scenario forcing in CMIP6 (Lund et al., 2023). We use the 2019 global CEDS inventory converted to ModelE emission-input format. CEDS resolves agriculture, energy, industry, transport, residential–commercial–other, solvents, waste, international shipping and aviation.

2.3.2 DPEC projections

DPEC combines socioeconomic development with air-pollutant and carbon-emission scenarios for China. It links the historical technological evolution of more than 700 MEIC source types to GCAM-China (Tong et al., 2020) and represents recent emission changes and scenario assumptions for China in greater detail than harmonized global inventories (Cheng et al., 2021). We obtained gridded DPEC projections for China at five-year intervals from 2025 to 2060. Version 1.2 updates the base year from 2015 to 2020, aligns base-year emissions with MEIC v1.4 and resolves near- and medium-term scenario assumptions more explicitly than v1.1. Of the six available pathways, we selected early peak-net zero-clean air. Although DPEC retains a broader source-category structure, the ModelE CEDS emission input used for these experiments exposes five anthropogenic sectors for replacement. We therefore aggregate and map the relevant DPEC categories onto those five model-input sectors: agriculture (AGR), energy/power (ENE), industry (IND), transport (TRA) and residential–commercial–other (RCO).

2.4 Meteorology

Both simulations were atmosphere-only integrations with prescribed monthly sea-surface temperature and sea-ice fields from the PCMDI-AMIP input4MIPs SSTAndSeaIce dataset, version 1.1.0. This dataset is based on merged UK Met Office



HadISST and NCEP OI v2 products and was remapped to the $2^\circ \times 2.5^\circ$ ModelE grid. For 2025–2060, the horizontal winds in both simulations were nudged toward the same future wind fields derived from the bias-corrected CMIP6 dataset of Xu et al. (2021) under the SSP126 scenario. These wind fields were processed and remapped to the 144×89 GISS ModelE grid with 17 pressure levels and four time records per day. The nudging coefficients for the zonal and meridional wind components were both set to 0.001. The pre-2025 nudging winds used to prepare the initial model state were taken from the standard GISS ModelE NCEP reanalysis input files.

The consequence for this study is that the two simulations share their synoptic meteorology. The grid-point correlation between the interannual anomalies of the two runs over 2031–2050 has a median of 0.998 for surface air temperature and exceeds 0.999 for surface wind. The EarlyPeak–Control difference is therefore not a free climate response. It isolates the response of atmospheric chemistry and aerosol microphysics to the emission change under nearly identical transport, together with the limited thermodynamic adjustment that nudged winds and prescribed sea-surface temperature allow. We do not interpret differences in circulation, temperature or precipitation.

2.5 Experimental design

EarlyPeak inputs were constructed on the fixed 2019 global CEDS baseline by a remove-and-fill procedure. A China mask was generated from administrative boundaries and the ModelE emission grid cells falling inside it were identified. Only those cells were removed from the 2019 files; every cell outside China retained its original 2019 value in each year from 2025 to 2060. For each target ModelE cell the DPEC field was mapped from the eight nearest source cells by inverse-distance weighting and written back to the matching month, sector and species after unit conversion. Annual values between five-year DPEC nodes were linearly interpolated. The procedure preserved the original time dimension, vertical injection levels and variable attributes.

Replacement was restricted to the five sectors exposed in the ModelE CEDS input and their mapped DPEC counterparts (AGR, ENE, IND, TRA and RCO), with CEDS ENE paired to DPEC POWER. The replacement is therefore defined by the sector structure of the model CEDS files, not by the full DPEC source-category list. DPEC categories not represented by those five model sectors were not introduced as additional variables. Conversely, CEDS solvent, waste, international shipping and aviation variables were outside the five-sector replacement and remained at 2019 levels in both runs. Species replacement was limited to CO, SO₂, BC, OC, NH₃ and NO_x, for which compatible DPEC and model variables exist. Organic emissions were mapped onto the MATRIX-VBS carbon variables by sector and volatility bin.

Control repeats the global 2019 CEDS emissions in every year and contains no future emission trend. EarlyPeak begins from the same baseline and substitutes year-specific DPEC early peak-net zero-clean air emissions for the compatible species in the five matched sectors within the China mask. All other inputs and the numerical configuration are identical between the two runs. The configuration is summarized in Table 2.



We verified the design from the emission diagnostics archived by the model itself and not only from the input files. For every year of the analysis period and for each of the ten emission sectors applied by the model, the EarlyPeak–Control difference in the emitted SO₂, BC, OC, NH₃ and NO_x is identically zero in every grid cell outside the China mask. The mask comprises 194 model cells, or 1.9 % of the Earth’s surface.

Both EarlyPeak and Control were initialized in 2025 and integrated through 2030 as a spin-up period. The 2025–2060 integrations used the same prescribed meteorology, including the same sea-surface temperature, sea-ice, and SSP126-based nudging wind fields. All reported results were calculated for 2031–2050, after the spin-up period.

Table 2. Meteorological and emission configuration of the two experiments.

Experiment	Meteorology	Emissions over China	Emissions outside China
EarlyPeak	Winds nudged to prescribed fields; prescribed sea-surface temperature and sea ice	Year-specific DPEC early peak-net zero-clean air replacement of five matched sectors on the 2019 CEDS baseline	2019 CEDS, fixed in every year
Control	Identical to EarlyPeak	2019 CEDS, fixed in every year	2019 CEDS, fixed in every year

This design isolates the response to the prescribed Chinese emission pathway while holding every other boundary condition fixed. It is deliberately idealized in two respects. Because emissions outside China do not evolve, the relative influence of the emission change over China is likely to be larger than it would be under a fully evolving global scenario. And because Control is fixed at CEDS 2019 while EarlyPeak uses DPEC, the difference between the runs contains both the difference between the two inventories for present-day China and the future emission reduction along the DPEC pathway; we return to this in Sects. 3.1 and 4.

2.6 Emission perturbation metrics

We quantify the imposed perturbation from the emission diagnostics archived by the model. For species X and year y, let $T_R^q(y)$ be the emission summed over all sectors and integrated over region R in experiment q. The perturbation relative to the Chinese baseline is

$$R_C(y) = \frac{T_C^{\text{EarlyPeak}}(y) - T_C^{\text{Control}}(y)}{T_C^{\text{Control}}(y)} \times 100\%, \quad (1)$$

where C denotes the China mask, and the same perturbation relative to the global emission is

$$R_G(y) = \frac{T_G^{\text{EarlyPeak}}(y) - T_G^{\text{Control}}(y)}{T_G^{\text{Control}}(y)} \times 100\%, \quad (2)$$

where G denotes the globe. R_G is always smaller in magnitude than R_C and is not a measure of the reduction achieved within China. Both describe emission inputs and not simulated burdens. Because the sums include the sectors that were not replaced, R_C is slightly smaller in magnitude than the reduction of the five replaced sectors alone.



2.7 Model evaluation and scope

This study analyses the prognostic aerosol fields of MATRIX-VBS: the mass of each component within each population, the number concentration and size of each population, and the gas-phase precursor and oxidant fields. The ability of the 15-
 190 population MATRIX-VBS configuration used here to represent aerosol composition and number has been evaluated against surface and airborne observations by Gao et al. (2024), who compared simulated mass concentrations of sulfate, organic and black carbon and simulated particle number and size distributions across a range of environments. That evaluation is the basis on which the present diagnostics rest, and we do not repeat it here.

No aerosol optical or radiative diagnostic is used in this study. We report no aerosol optical depth, absorption or extinction
 195 and no radiative forcing or flux, and we draw no conclusion about the radiative consequences of the changes we describe. The analysis is confined to the aerosol mass, number, size and activation fields computed by the microphysical scheme, and the one optical quantity reported, the optical depth of water clouds (Sect. 3.6), is computed by the cloud scheme from droplet number and condensed water.

Because we do not use column optical depth, we also do not use the satellite column-optical-depth comparison that would
 200 conventionally accompany a global aerosol study. Column optical depth cannot in any case distinguish the populations of MATRIX-VBS, so it could not have constrained the mixing-state allocation that is the subject of this paper. Confidence in the results rests on the composition and number evaluation of Gao et al. (2024), on the internal consistency of the component, number and size diagnostics, and on the closed process budgets of Sect. 3.4.

2.8 Regions and statistics

All regional statistics are area-weighted means or integrals on the native model grid. The source region is the China emission
 205 mask of Sect. 2.5. The downwind regions are latitude–longitude rectangles: the Northwest Pacific (20–50° N, 140° E–180°), the Northeast Pacific (20–55° N, 180°–125° W), North America (25–55° N, 125–65° W) and the Arctic (north of 66° N). The Middle East (15–40° N, 35–60° E) and South Asia (5–30° N, 68–95° E) are retained as upwind reference regions. These definitions are used in every figure, table and number reported in this paper.

The share of a regional sulfate column carried on BC-containing particles is the regional-mean sulfate column of the BC-
 210 containing populations divided by the regional-mean sulfate column of all fifteen populations. Changes in shares are given in percentage points. Boundary-layer quantities are air-mass-weighted means over the lowest five model layers, which extend from the surface to about 860 hPa. The activated particle number is averaged over the same layers without mass weighting. The diameter of a population is the number-weighted mean of the archived dry diameter over the same layers. Coating ratios, such as the sulfate-to-BC mass ratio of the BOC population, are ratios of regional-mean column burdens.

All changes are 2031–2050 means of EarlyPeak–Control. Significance is evaluated with a two-sided one-sample *t* test of the
 20 paired annual differences against zero. Because the two simulations share their meteorology (Sect. 2.4), the paired differences contain very little weather noise: the standard deviation of the 20 annual differences of the Chinese sulfate



column is 1.4 percentage points about a mean of -23.7% . Nearly every change reported here is therefore statistically significant, and we regard significance as a necessary check and not as evidence of importance; magnitudes are reported throughout. No multiple-comparison correction is applied. Mapped fields are smoothed with a Gaussian filter of one grid cell for display only, and colour scales are nonlinear so that the far-field response remains visible; every number quoted in the text and every significance test uses unsmoothed data. On maps, stippling marks areas where the change is not significant at $p < 0.05$.

2.9 Closed process budget

MATRIX archives seven column-integrated microphysical tendencies for every mass and number tracer: nucleation (P1), coagulation (P2), condensation (P3), in-cloud processing (P4), intermodal loss (P5), mode transfer (P6) and the total change (P7). We use them as follows.

The total microphysical tendency P7 is the quantity that enters the tracer budget. For a tracer with surface emission E, aqueous-phase production A, and wet and dry deposition W and D (dry deposition includes gravitational settling), the net export from a region is

$$X = E + P_7 + A - W - D. \quad (3)$$

Globally and in the annual mean X should vanish. It does so to within 0.3 % of the total source for each of the sulfate populations analysed here, which establishes that P7 is the net microphysical source and that the archived process terms are in $\text{kg m}^{-2} \text{s}^{-1}$ after the scale factor in the file metadata is applied. Regionally, X is the net horizontal export of the tracer; a negative value is a net import.

The coagulation and intermodal-loss terms P2 and P5 are gross rates and cannot be added to the other terms. For populations whose turnover time is shorter than the model time step they exceed the mass actually transferred by up to an order of magnitude: the archived global intermodal loss of fresh black carbon (BC1) is 50 Tg yr^{-1} , against a total black-carbon emission of 8 Tg yr^{-1} . We therefore diagnose the net transfer of a tracer into or out of a population by coagulation as the residual

$$K = P_7 - P_1 - P_3 - P_4 - P_6. \quad (4)$$

Summed over all populations, K vanishes exactly for black carbon and to within 0.3 % of the gross transfer for sulfate, as it must for a process that moves mass between populations without creating it. The nucleation and condensation terms P1 and P3 summed over populations give the gas-phase source of sulfate, which together with aqueous production and primary emission matches total sulfate deposition.

3 Results and discussion

3.1 Emission perturbation

Figure 1 shows the emission difference between EarlyPeak and Control. Every non-zero cell lies inside the China mask by construction. Averaged over 2031–2050, emissions inside the mask are lower in EarlyPeak by 10.2 Tg yr⁻¹ for SO₂ ($R_C = -86\%$), 1.00 Tg yr⁻¹ for BC (-83%), 4.4 Tg yr⁻¹ for OC (-66%) and 6.0 Tg yr⁻¹ for NH₃ (-50%). Relative to the global emission these are -9.6% , -17% , -9.2% and -9.7% . NO_x is lower by 22 % on average ($R_G = -4.0\%$).

Two properties of the perturbation matter for the interpretation of the response. First, it is close to a step. The SO₂ reduction is already 80 % in 2031 and reaches 91 % in 2050; for BC the range is 77 % to 87 % and for OC 64 % to 68 % (Fig. 1d). Most of the difference between the runs is therefore present from the beginning of the analysis period, which reflects both the emission reduction already built into the DPEC pathway by 2030 and the difference between DPEC and CEDS 2019 for present-day China. The aerosol response is correspondingly almost stationary over 2031–2050 (Sect. 3.7), and we treat the 20 years as samples of one regime. Second, NO_x is the exception: EarlyPeak emits 18 % more NO_x over China than Control in 2031 and 50 % less in 2050, crossing zero in about 2034. Nitrate is consequently the only aerosol component whose response evolves strongly during the analysis period.

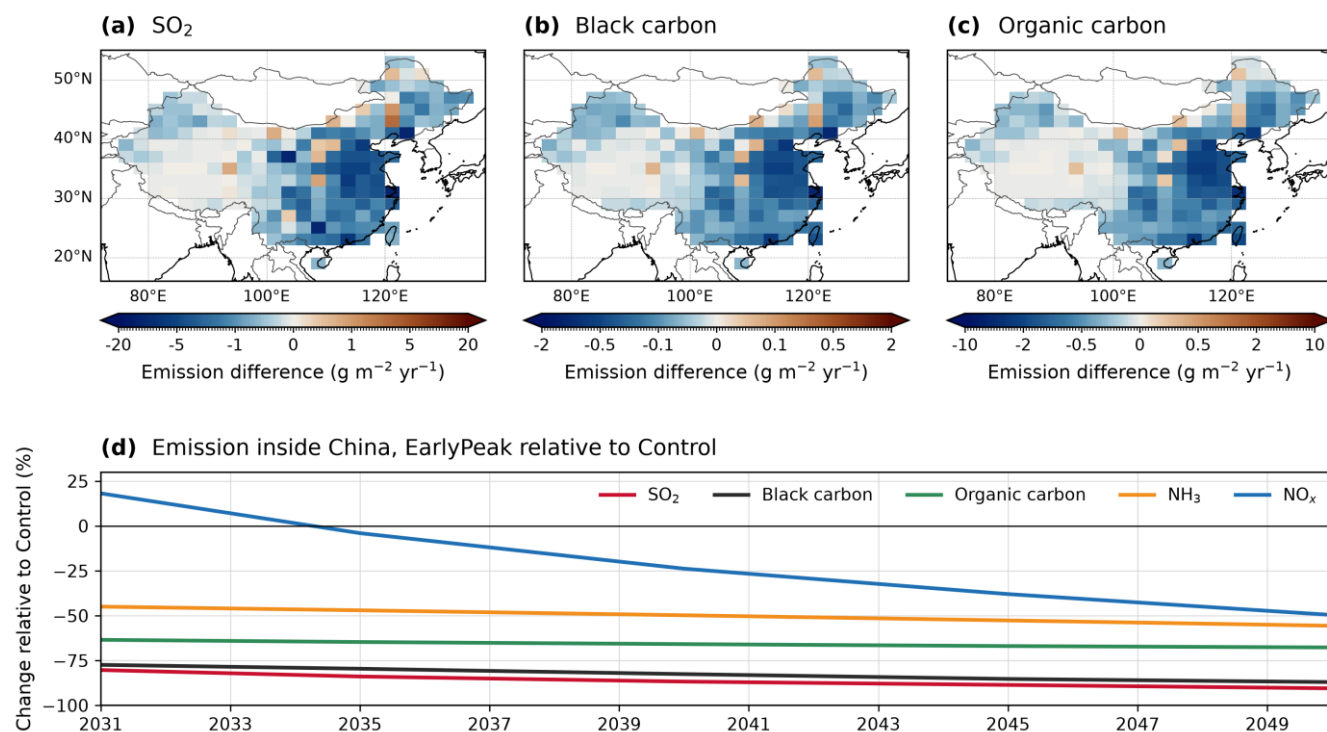


Figure 1. Emission perturbation. (a–c) Mean 2031–2050 difference (EarlyPeak – Control) in the emission of SO₂, black carbon and organic carbon applied by the model, summed over all sectors, shown on the model grid without smoothing; the difference is



265 **identically zero outside the China emission mask. Each panel has its own colour scale. (d) Emission integrated over the China mask in EarlyPeak relative to Control for each year.**

3.2 Bulk response

The column burdens of sulfate, black carbon and organic carbon decrease over China and along a plume that crosses the North Pacific and reaches North America and the Arctic (Fig. 2, Table 3). Over China the sulfate column falls from 5.92 to 4.51 mg m^{-2} (−24 %), black carbon from 0.76 to 0.34 mg m^{-2} (−55 %) and organic carbon from 3.83 to 3.10 mg m^{-2} (−19 %).
 270 Over the Northwest and Northeast Pacific sulfate is lower by 13 % and 10 % and black carbon by 33 % and 21 %. Over North America and the Arctic sulfate is lower by 6.2 % and 4.5 %. The sulfate decrease is significant over 98 % of the Northern Hemisphere. Globally, sulfate is lower by 3.6 %, black carbon by 8.4 % and organic carbon by 1.2 %.

There is no comparable response upwind. Over the Middle East the sulfate and black-carbon columns change by −0.6 % and −0.5 %, and over South Asia by −1.1 % and −0.9 %.

275 The response at the source is strongly sub-linear: an 86 % reduction in SO_2 emission lowers the sulfate column over China by 24 %, and an 83 % reduction in black-carbon emission lowers the black-carbon column by 55 %. The budget of Sect. 2.9 shows why. In Control, the sulfate produced and emitted over China (5.9 Tg yr^{-1}) exceeds deposition there, and China exports about 0.5 Tg yr^{-1} of sulfate. In EarlyPeak the source falls to 2.5 Tg yr^{-1} while deposition is 3.0 Tg yr^{-1} , so China imports 0.57 Tg yr^{-1} . Black carbon behaves in the same way, changing from a net export of 0.24 Tg yr^{-1} to a net import of
 280 0.15 Tg yr^{-1} . After the emission reduction, about one fifth of the sulfate deposited over China is transported into the region from outside the mask. The production of sulfate within China falls by 56 %, less than the SO_2 emission, because sulfur dioxide is also imported: SO_2 deposition plus conversion to sulfate over China exceeds the Chinese SO_2 emission in EarlyPeak, whereas in Control about half of the emitted SO_2 is deposited, one third is converted to sulfate within the region and the remainder is exported. Of the two production routes, gas-phase production of sulfate over China falls by 49 % and
 285 aqueous production by 69 %.

The vertical structure separates the source from the far field (Fig. 3). Over China the response is largest at the surface, where sulfate is lower by 48 % and black carbon by 75 %, and it weakens rapidly with height to about −10 % for sulfate in the middle troposphere. Over the Northeast Pacific the response is nearly uniform with height at −9 % to −15 % for sulfate. Over North America and the Arctic it is largest aloft: sulfate is lower by 4 % at the surface over North America and by 10 % at
 290 500 hPa, and by 1 % at the surface in the Arctic and by 8 % near 400 hPa. The far-field signal of the emission reduction is therefore a free-tropospheric signal. The same is true of the weak response over the Middle East, which is confined to levels above 400 hPa (−4 % to −5 %) and is absent below 700 hPa.

The global lifetime of sulfate, defined as burden divided by total deposition, lengthens from 6.93 to 7.08 days (+2.1 %), and that of black carbon from 4.76 to 4.97 days (+4.5 %). This is consistent with the aerosol removed by the emission reduction
 295 being shorter-lived than the global average: the deposition lifetimes of the sulfate and black-carbon populations over China are 3 to 4 days.



Table 3. Regional response to the emission reduction over China, EarlyPeak – Control, 2031–2050 mean. Regions are defined in Sect. 2.8.

	China	NW Pacific	NE Pacific	North Amer.	Arctic	Middle East	Global
Sulfate column (%)	−23.7	−13.2	−10.0	−6.2	−4.5	−0.6	−3.6
Black-carbon column (%)	−55.5	−32.7	−21.4	−3.8	−5.8	−0.5	−8.4
Organic-carbon column (%)	−19.0	−9.1	−4.5	−0.3	−0.6	−0.4	−1.2
Sulfate on BC-containing particles (%)	−40.5	−29.6	−23.1	−12.7	−12.1	−0.6	−9.0
Sulfate on BC-free particles (%)	−14.7	−8.2	−7.0	−4.6	−2.5	−0.7	−2.2
Share on BC-containing, Control (%)	35.4	23.2	19.2	20.0	20.5	37.5	20.8
Share on BC-containing, EarlyPeak (%)	27.6	18.8	16.4	18.6	18.9	37.5	19.7
Boundary-layer particle number (%)	−25.7	−1.9	−0.1	+0.5	+5.4	+0.8	−1.0
Boundary-layer BC-containing number (%)	−64.6	−33.7	−19.3	−1.0	−2.0	−0.1	−9.9
Cloud-top droplet number (%)	−28.7	−6.5	−4.7	−0.9	+0.6	+0.1	−2.1

Note: all entries differ from zero at $p < 0.05$ in the paired test of Sect. 2.8 except the change in the Middle Eastern share (−0.01 percentage points), Northeast Pacific particle number and Middle Eastern cloud-top droplet number.

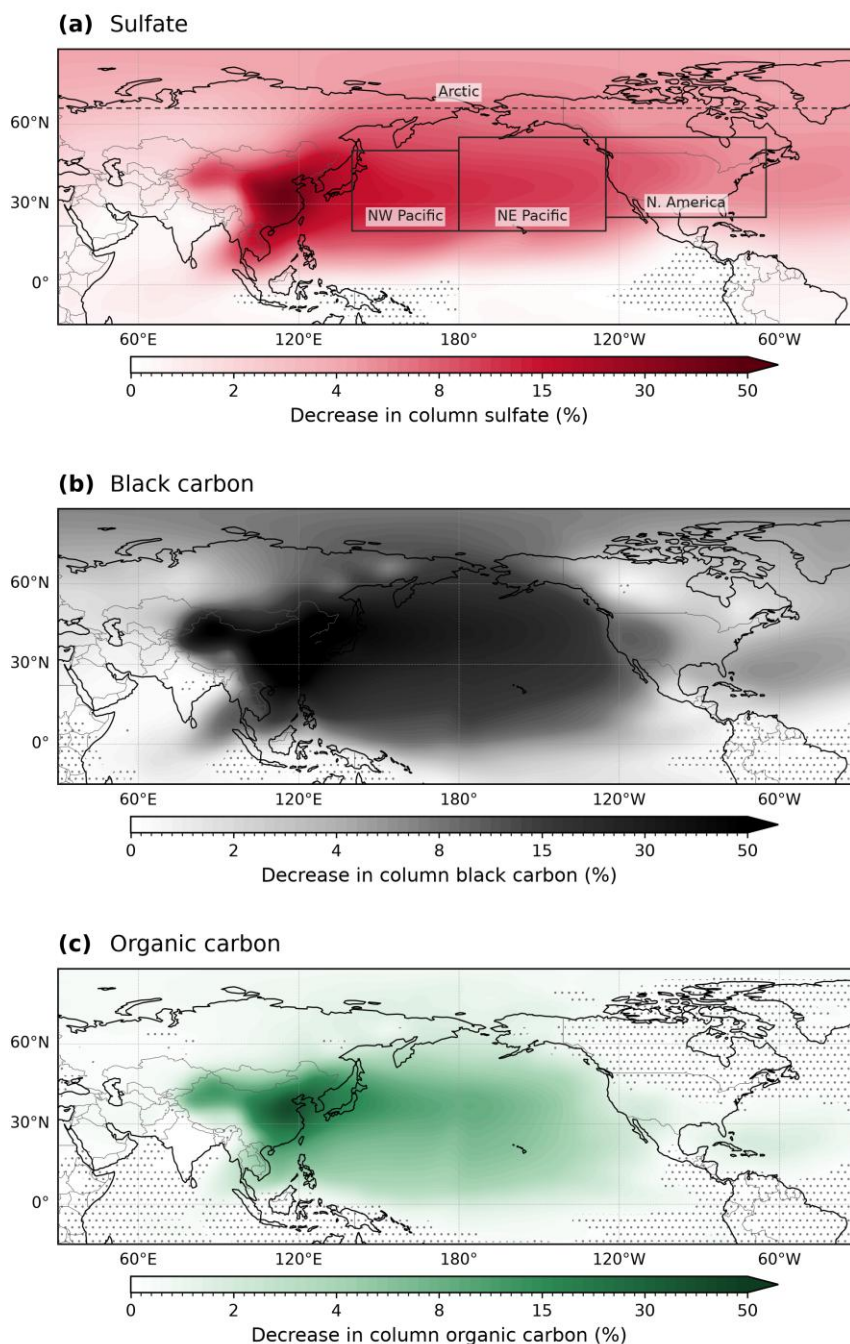


Figure 2. Relative change in column burden, EarlyPeak – Control, 2031–2050 mean. (a) Sulfate. (b) Black carbon. (c) Organic carbon, summed as defined in Sect. 2.2. Boxes in (a) are the downwind analysis regions and the dashed line is 66° N. Stippling marks areas where the change is not significant at $p < 0.05$. Fields are smoothed for display and the colour scale is nonlinear (Sect. 2.8).

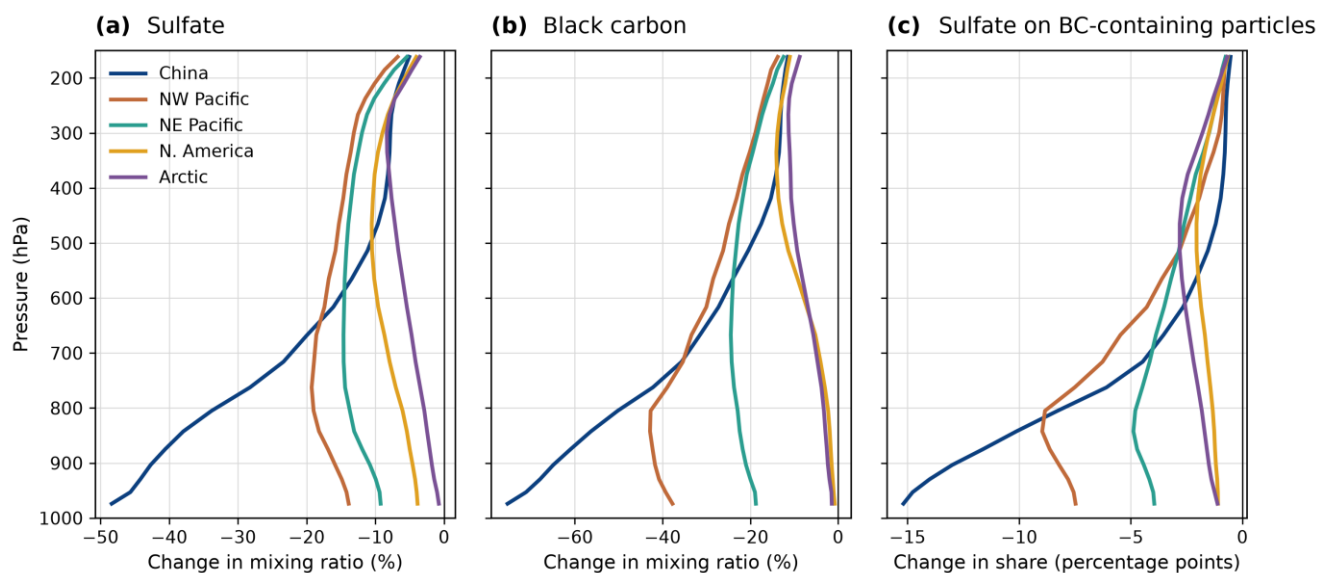


Figure 3. Vertical structure of the response, EarlyPeak – Control, 2031–2050 regional means. (a) Relative change in sulfate mass mixing ratio. (b) Relative change in black-carbon mass mixing ratio. (c) Change in the share of sulfate carried on BC-containing particles, in percentage points.

3.3 Sulfate on BC-containing particles

The emission reduction does not remove sulfate uniformly from the particle population; this is the central result of the study. The three populations that carry nearly all of the sulfate column respond very differently (Fig. 4). Over China, sulfate in the accumulation mode (ACC) decreases by 9 %, sulfate in organic–sulfate particles (OCS) by 20 %, and sulfate in black-carbon–organic-carbon particles (BOC) by 41 %. The contrast is not confined to the source. Over the Northeast Pacific the three changes are –6.5 %, –8.4 % and –23 %, and over the Arctic –2.2 %, –3.3 % and –12 %.

Grouping the populations by whether they contain black carbon makes the pattern explicit (Fig. 5b, Table 3). Sulfate on BC-containing particles decreases by 41 % over China, 30 % and 23 % over the Northwest and Northeast Pacific, 13 % over North America and 12 % over the Arctic. Sulfate on BC-free particles decreases by 15 %, 8 %, 7 %, 5 % and 2.5 % in the same regions. In every region reached by the plume, sulfate is lost from BC-containing particles about three times faster than from BC-free particles, and nearer five times faster in the Arctic.

The share of the sulfate column carried on BC-containing particles consequently falls from 35.4 % to 27.6 % over China, a change of –7.8 percentage points (Fig. 5a). It falls by 4.4 and 2.8 percentage points over the Northwest and Northeast Pacific, by 1.4 over North America and by 1.6 over the Arctic, where it goes from 20.5 % to 18.9 %. The global-mean share falls from 20.8 % to 19.7 %. Over the Middle East and South Asia the share is unchanged (37.5 % and 50.7 % in both simulations). In terms of the individual populations, the sulfate column over China is carried 35 % by BOC, 32 % by ACC and 28 % by OCS in Control, and 27 %, 39 % and 29 % in EarlyPeak: the largest carrier of sulfate changes from the black-carbon–organic-carbon population to the accumulation mode.



330 The reallocation is largest in the surface layer. In the surface layer over China the share of sulfate on BC-containing particles falls from 50 % to 36 %, with sulfate on BC-containing particles lower by 61 % and on BC-free particles by 31 %. It weakens with height in step with the bulk response, and above about 500 hPa over China the share changes by less than 2 percentage points (Fig. 3c). Downwind, the change in share is carried at all levels of the lower and middle troposphere.

335 A bulk diagnosis of the same simulations would report that the sulfate column over China decreases by 24 %. It would not show that the sulfate mixed with black carbon decreases by 41 % while the rest decreases by 15 %. The properties that depend on that distinction, how thickly black carbon is coated and which particles are soluble and large enough to nucleate cloud droplets, are the subject of the following sections.

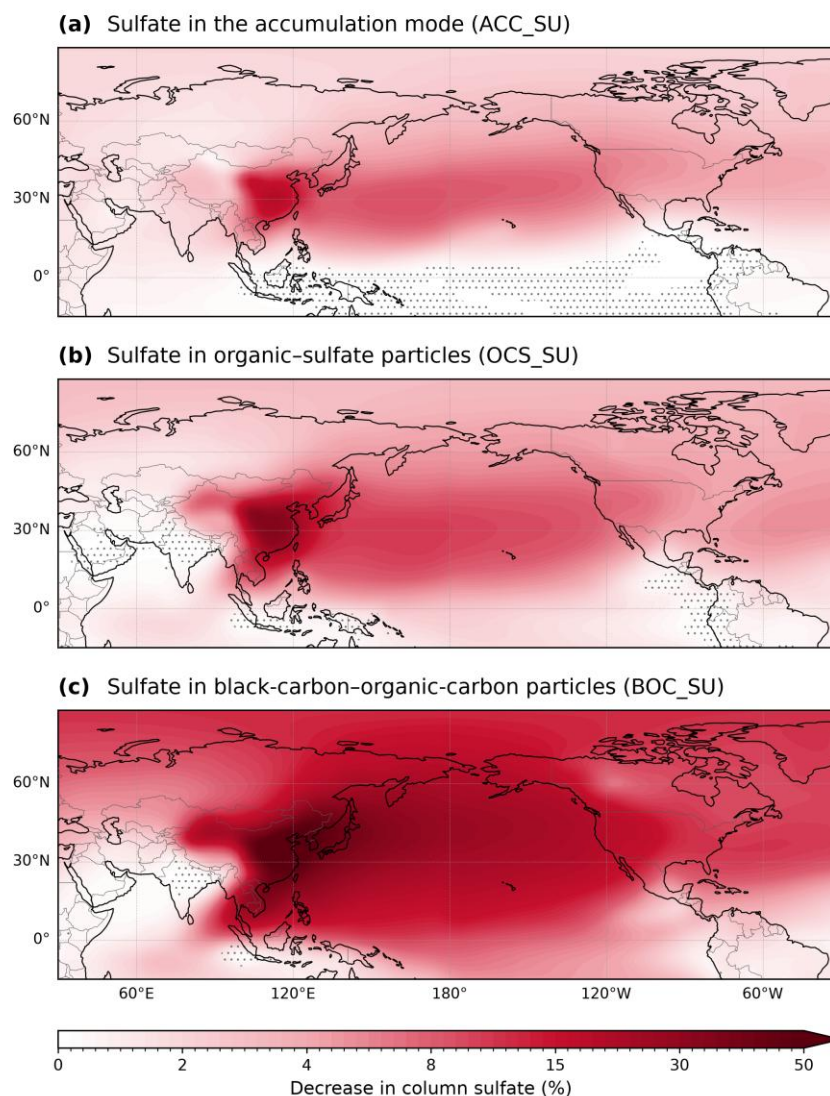
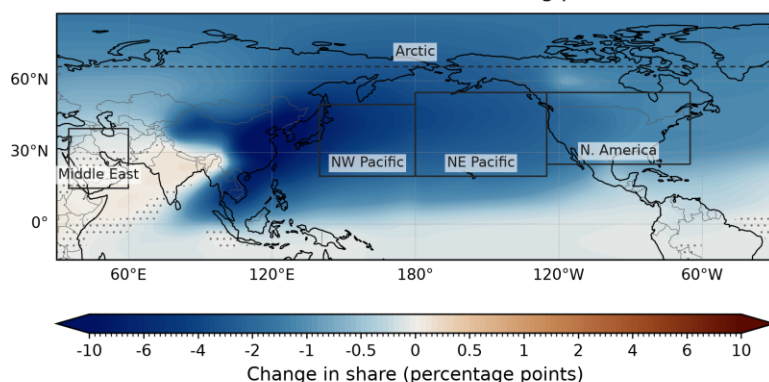


Figure 4. Relative change in the sulfate column carried by the three populations that hold most of the sulfate, EarlyPeak – Control, 2031–2050 mean. (a) Accumulation mode, ACC_SU. (b) Organic-sulfate population, OCS_SU. (c) Black-carbon-organic-carbon population, BOC_SU. Stippling marks areas where the change is not significant at $p < 0.05$.



(a) Share of column sulfate on BC-containing particles



(b) Sulfate by carrier

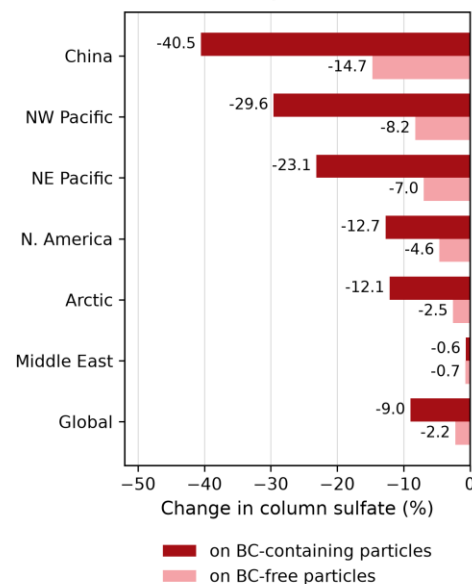


Figure 5. Sulfate on BC-containing particles, EarlyPeak – Control, 2031–2050 mean. (a) Change in the share of the sulfate column carried on BC-containing particles (BC1, BC2, BCS and BOC), in percentage points; boxes mark the analysis regions and stippling marks areas where the change is not significant at $p < 0.05$. (b) Relative change in the regional sulfate column carried on BC-containing and on BC-free particles.

3.4 Process budget

The change in shares could in principle arise without any change in microphysics, simply because less internally mixed air imported from elsewhere makes up a larger part of the column over China after the emission reduction. The closed budget of Sect. 2.9 shows that a process change is also involved, and quantifies it (Table 4).

We first ask where newly formed and emitted sulfate ends up. Summing primary emission, aqueous production and the net microphysical tendency for each population gives the net source of sulfate to that population. In Control, 36 % of the sulfate source over China is delivered to BOC, 29 % to OCS and 30 % to ACC. In EarlyPeak the total source is 59 % smaller, and 25 % of it is delivered to BOC, 40 % to OCS and 30 % to ACC. The emission reduction therefore reroutes about one tenth of the sulfate formed over China from particles that contain black carbon to organic particles that do not, while the accumulation mode receives the same fraction as before. The same shift, smaller in size, is found over the Northwest Pacific (BOC 13 % to 9 %) and in the global budget (20 % to 18 %). Over the Northwest Pacific the total sulfate source falls by only 13 %, so the downwind change in mixing state arises both from the import of less internally mixed aerosol from the source region and from the reduced transfer of locally formed sulfate to the smaller number of BC-containing particles that arrive.

The budget also resolves how sulfate reaches the BOC population. In Control, BOC gains 0.82 Tg yr^{-1} of sulfate over China by condensation of sulfuric acid and 1.30 Tg yr^{-1} by net coagulation with sulfate-bearing particles, so coagulation supplies 61 % of the total. In EarlyPeak the two terms are 0.25 and 0.36 Tg yr^{-1} , lower by 69 % and 72 %. The two routes weaken



almost equally, and both weaken by more than the total sulfate source (-59%). Both routes depend on the number of BOC particles available to receive sulfate, which falls by 52% in the Chinese column (Sect. 3.5); the surface available for condensation and the number of coagulation partners therefore decline together with the sulfate supply itself.

The accumulation mode shows the complementary change. In Control it receives 0.87 Tg yr^{-1} by condensation, 2.05 Tg yr^{-1} by aqueous production and 0.45 Tg yr^{-1} by primary emission, and loses 1.65 Tg yr^{-1} by net coagulation to the mixed populations. In EarlyPeak every source is smaller, but the coagulation loss falls by 63% to 0.61 Tg yr^{-1} , so a larger part of the sulfate that enters the accumulation mode stays there.

For black carbon, the budget shows that ageing slows but remains fast. Fresh black carbon (BC1) over China is converted to coated or mixed populations by mode transfer and coagulation with a turnover time of 1.1 hours in Control and 2.2 hours in EarlyPeak. Ageing is therefore twice as slow when less coating material and fewer coagulation partners are available, but it remains much faster than deposition, and the fresh fraction of the black-carbon column stays at about 2% . Slower ageing therefore cannot account for the black-carbon column falling less than the emission; in the budget, that difference is the change from net export to net import described in Sect. 3.2.

Table 4. Closed process budget over China, 2031–2050 mean, in Tg yr^{-1} of sulfate (first four columns) or black carbon (last column). Entries are Control $\rightarrow$ EarlyPeak.

Term	ACC_SU	OCS_SU	BOC_SU	All sulfate	BOC_BC
Primary emission	$0.45 \rightarrow 0.06$	—	—	$0.45 \rightarrow 0.06$	—
Condensation (P3)	$0.87 \rightarrow 0.58$	$0.76 \rightarrow 0.39$	$0.82 \rightarrow 0.25$	$3.30 \rightarrow 1.67$	—
Aqueous production	$2.05 \rightarrow 0.64$	—	—	$2.05 \rightarrow 0.64$	—
Net coagulation (K)	$-1.65 \rightarrow -0.61$	$0.96 \rightarrow 0.58$	$1.30 \rightarrow 0.36$	0	$0.95 \rightarrow 0.18$
Wet deposition	$1.70 \rightarrow 0.77$	$1.31 \rightarrow 0.88$	$1.50 \rightarrow 0.82$	$4.72 \rightarrow 2.62$	$0.57 \rightarrow 0.24$
Dry deposition	$0.12 \rightarrow 0.07$	$0.23 \rightarrow 0.16$	$0.31 \rightarrow 0.14$	$0.70 \rightarrow 0.41$	$0.18 \rightarrow 0.05$
Net export (X)	$-0.01 \rightarrow -0.11$	$0.18 \rightarrow -0.08$	$0.30 \rightarrow -0.35$	$0.50 \rightarrow -0.57$	$0.20 \rightarrow -0.12$
Share of sulfate source (%)	$30 \rightarrow 30$	$29 \rightarrow 40$	$36 \rightarrow 25$	100	—
Column burden change (%)	-8.7	-20.3	-40.6	-23.7	-55.0

Note: terms are defined in Sect. 2.9. Dry deposition includes gravitational settling. “All sulfate” sums the fifteen populations and additionally includes $0.11 \rightarrow 0.09 \text{ Tg yr}^{-1}$ of nucleation; net coagulation sums to zero over populations by construction of Eq. (4). A negative net export is a net import. The accumulation-mode column also includes a mode-transfer gain from the Aitken mode of $0.08 \rightarrow 0.05 \text{ Tg yr}^{-1}$.

3.5 Particle number and size

In the boundary layer over China the total particle number concentration falls from 1.40×10^4 to $1.04 \times 10^4 \text{ cm}^{-3}$ (-26% ; Fig. 6a, c). The Aitken mode, which holds two thirds of the particles, falls by 26% , and the nucleation rate over China by 24% . The populations that receive primary emissions fall much more: organic carbon (OCC) by 31% , and the four BC-containing populations together by 65% , from 520 to 180 cm^{-3} . The accumulation mode is the least affected, at -5% .



385 Away from the source the number response is weak and changes sign (Fig. 6a, Table 3). The number of BC-containing particles is lower all along the plume, by 34 % and 19 % in the boundary layer of the Northwest and Northeast Pacific. Total particle number, however, is lower by only 2 % over the Northwest Pacific and is unchanged over the Northeast Pacific, and it is slightly higher over much of the northern middle and high latitudes, by 5 % in the Arctic boundary layer. The increase is carried by the Aitken and accumulation modes. It occurs without an increase in the nucleation rate and is consistent with a weaker sink of small particles onto a reduced population of large ones, but we have not attributed it and do not interpret it further.

The size response differs between populations, and in a way that follows from the budget of Sect. 3.4 (Fig. 6d). Over China the number-weighted dry diameter of BOC particles increases by 6.3 %, from 205 to 217 nm, of pure organic particles (OCC) by 4.8 %, and of organic–sulfate particles (OCS) by 2.0 %. The composition of the BOC population explains its growth. Its number falls by 52 % in the column, but the sulfate it carries falls by only 41 % and its organic carbon by 37 %, so each remaining BOC particle carries 22 % more mass. Per unit black carbon, the BOC population carries 32 % more sulfate and 40 % more organic carbon than in Control. Fewer cores share the condensable material, and each is more thickly coated.

Two populations become smaller. The accumulation mode shrinks by 3.1 % over China and by 1 % to 3 % over the Pacific and North America, consistent with less sulfuric acid condensing on a nearly unchanged number of particles. Fresh black carbon (BC1) shrinks by 29 %, from 179 to 128 nm, because the condensed organic and inorganic material that it acquires before being transferred to the aged populations is much reduced.

Downwind of the source the BOC population does not grow. Its dry diameter decreases by about 1 % to 2 % over the Northeast Pacific, North America and the Arctic (Fig. 6b), and its sulfate-to-black-carbon ratio decreases by 9 % over North America and 7 % over the Arctic. Far from the source the number of BC-containing particles is little changed, while less sulfate is available to coat them. “Fewer and larger” therefore describes the coated carbonaceous particles of the source region. It is not a property of the whole population, and it reverses in the far field.

The black-carbon response consequently has to be reported as a population total and as a per-particle property separately, because the two move in opposite directions. Over China there are two thirds fewer BC-containing particles in the boundary layer and 41 % less sulfate resides on black carbon, while each remaining BC-containing particle is larger and carries about one third more sulfate and 40 % more organic carbon per unit black carbon. Properties that scale with the number or total mass of mixed particles, such as the number of coated particles available as cloud condensation nuclei, decrease strongly. Properties that are set per particle, such as coating thickness, the hygroscopicity of an individual BC-containing particle and its susceptibility to wet removal, move towards more aged particles. The slower conversion of fresh black carbon (Sect. 3.4) is a third, separate effect, and in these simulations it is too small to matter for the black-carbon burden. A statement about “the effect of emission reductions on black-carbon coating” therefore has no single sign: it depends on whether the black carbon and the sulfate that would coat it are reduced together, as at the source, or separately, as in the far field.

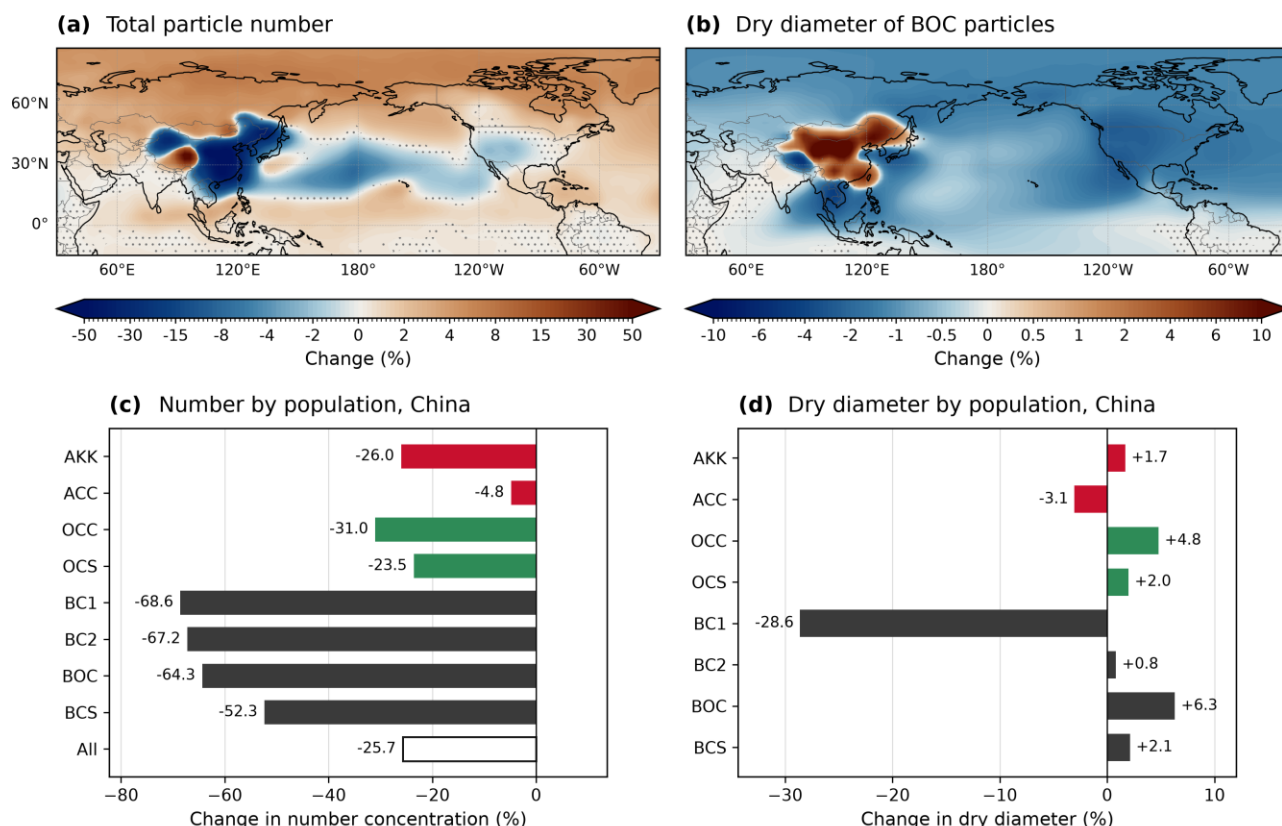


Figure 6. Particle number and size in the boundary layer (lowest five model layers), EarlyPeak – Control, 2031–2050 mean. (a) Relative change in total particle number concentration. (b) Relative change in the number-weighted dry diameter of the BOC population. Stippling in (a) and (b) marks areas where the change is not significant at $p < 0.05$. (c) Relative change in the number concentration of each population over China; “All” is the total over the fifteen populations. (d) Relative change in the number-weighted dry diameter of each population over China.

3.6 Activation and cloud properties

The changes in number, size and mixing state combine in the number of particles that activate to form cloud droplets, which the model computes from the simulated size and composition of each population. Activation and the cloud properties that follow from it are the downstream consequences that can be examined without the aerosol optical calculation. We report them as relative changes and shares only (Sect. 2.2).

In the boundary layer over China the number of activated particles decreases by 23 %, and the droplet number concentration near the top of stratiform clouds decreases by 29 %, from 330 to 235 cm^{-3} (Fig. 7a). Droplet number is lower by 6.5 % and 4.7 % over the Northwest and Northeast Pacific and by less than 1 % over North America, and the cloud-top effective radius is larger by 4.6 % over China. The two diagnostics are consistent with each other: across the Northern Hemisphere the spatial correlation between the relative changes in activated number and in cloud-top droplet number is 0.86.



The cloud scheme carries the droplet-number change into the optical depth of water clouds, which it computes from droplet number and condensed water. Over China the optical depth of water clouds is lower by 5.9 %, from 26.9 to 25.3, while the cloud condensed water column is higher by 1 % and the effective radius of warm stratiform clouds is larger by 3.9 %; the change is therefore a change in droplet size at nearly constant cloud water. Over the Northwest and Northeast Pacific the optical depth is lower by 0.5 %, over North America by 0.2 % and globally by 0.3 %, and it is unchanged over the Middle East. Water-cloud cover changes by less than 0.3 percentage points everywhere. We take the analysis no further than this cloud property. With nudged winds and prescribed sea-surface temperature the simulations contain no circulation, precipitation or temperature response to the change, and the radiative flux diagnostics are not used (Sect. 2.7).

The population-resolved diagnostics show where the loss comes from (Fig. 7b). In Control, 38 % of the activated particles over China are BOC particles, 35 % are OCS and 21 % are accumulation-mode particles, although BOC accounts for less than 3 % of particle number; BOC particles are the largest of the fine populations and are coated with soluble material, so they activate readily. In EarlyPeak the number of activated BOC particles is lower by 52 %, which by itself lowers the total by 19.8 percentage points, or 85 % of the total decrease. Activated OCS particles contribute a further 4.8 points and the accumulation mode 0.7, while the larger pure organic particles of Sect. 3.5 activate more often and offset 2.1 points.

The decrease in droplet-forming particles over China is therefore predominantly a decrease in the number of coated carbonaceous particles, and only to a small extent a consequence of less sulfate in the accumulation mode. The growth of the individual BOC particles described in Sect. 3.5 does not compensate for their loss in number. This attribution requires a model that carries mixing state and number; a scheme that diagnoses cloud condensation nuclei from sulfate mass alone would attribute the same change to the 24 % decrease in sulfate. An earlier study with the MATRIX scheme likewise found that reductions in carbonaceous emissions lower cloud droplet number (Bauer et al., 2010).

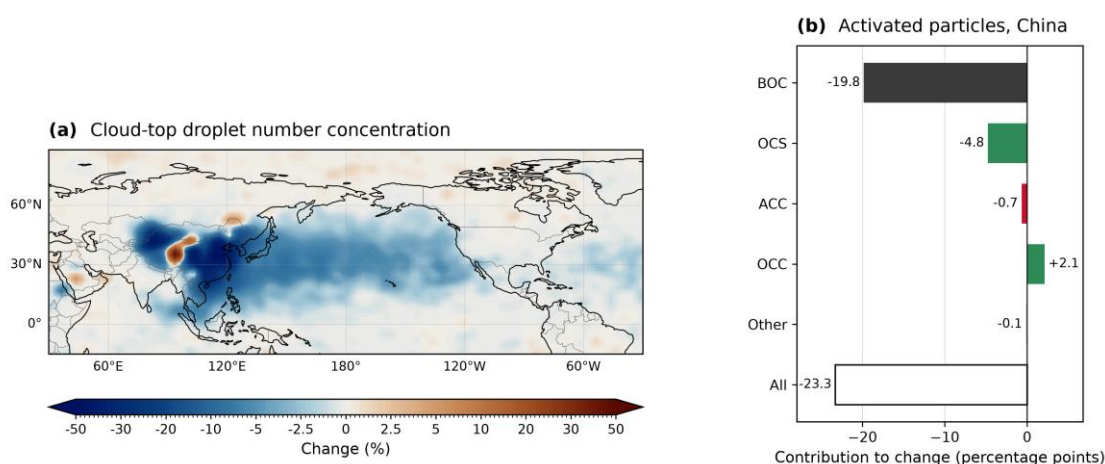


Figure 7. Aerosol activation and cloud droplet number, EarlyPeak – Control, 2031–2050 mean. (a) Relative change in droplet number concentration near the top of stratiform clouds; cells where the change is not significant at $p < 0.05$ are set to zero before the display smoothing. (b) Contribution of each population to the change in the number of activated particles in the boundary layer over China, in percentage points of the Control total.

3.7 Time evolution and other species

Because the perturbation is close to a step (Sect. 3.1), the response is nearly stationary (Fig. 8). The change in the Chinese sulfate column goes from -22% in 2031 to -25% in 2050, that in black carbon from -52% to -58% , and the share of sulfate on BC-containing particles in EarlyPeak stays between 26% and 29% throughout. Over the Northeast Pacific the sulfate response is -9% to -11% in every year.

Nitrate is the exception. Its column over China is almost unchanged in 2031 (-2%), when Chinese NO_x emissions in EarlyPeak exceed those of Control, and is lower by 39% in 2050, when they are 50% lower. Over the Northeast Pacific, in contrast, nitrate is lower by about 50% throughout, in step with the ammonia burden there (-54%) and not with Chinese NO_x . Ammonium decreases by 29% over China and the Pacific. MATRIX carries nitrate, ammonium and aerosol water as bulk species distributed over the sulfate-containing populations by thermodynamic equilibrium, so their response cannot be attributed to individual populations, and we do not analyse it further.

Mineral dust and sea-salt burdens change by less than 1% in every analysis region except China, where the sea-salt column is 2% lower. One feature of the dust response nevertheless mirrors the behaviour of black carbon. Over China the fraction of dust mass that remains in the dust populations, instead of being transferred to the fully mixed population, rises from 4.1% to 5.6% , consistent with fewer fine particles and less sulfate being available to mix with dust.

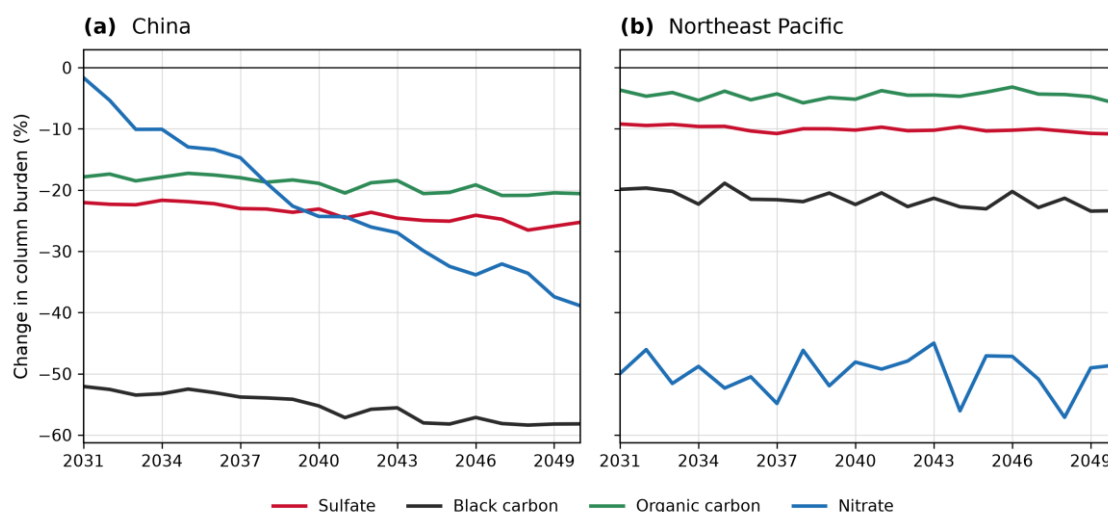


Figure 8. Annual relative change in column burden, EarlyPeak – Control, for each year of the analysis period. (a) China. (b) Northeast Pacific.

3.8 Implications for assessing emission changes

Previous assessments of aerosol emission perturbations have largely reported total or bulk-mean responses (Stjern et al., 2016; Samset et al., 2018). Resolving mixing state and number has three implications for such assessments.



First, mass-based metrics under-report what changes. Over China the sulfate column is lower by 24 %, but the quantities that
480 change most are the number of BC-containing particles (−65 %), the number of activated particles contributed by them
(−52 %) and the sulfate mixed with black carbon (−41 %), none of which is the sulfate or black-carbon mass that an
assessment would normally report, and none of which can be recovered from component masses. The same holds downwind:
the preferential loss of sulfate from BC-containing particles is found with a ratio of about three along the whole outflow, and
the share of sulfate on BC-containing particles is measurably lower in the Arctic.

485 Second, the reductions of sulfur dioxide and of carbonaceous emissions are not separable in their effect on cloud-forming
particles. Most of the decrease in activated particles comes from a population that exists because black carbon, organic
carbon and sulfate are emitted together and mix; a pathway that reduced sulfur dioxide alone or black carbon alone would be
expected to change the number and the composition of cloud condensation nuclei differently. We have not run such single-
species experiments, and the statement is an inference from the budget, not a result.

490 Third, once emissions over China are small, the sulfate and black-carbon columns there are sustained in part by transport
from outside the region, so the aerosol burden over China becomes less sensitive to further local reductions. This dependence
on transport is at its largest in the present design, in which emissions outside China are held fixed.

Establishing what these changes imply for aerosol optical properties and for the radiative effect of the cloud response
requires optical and radiative diagnostics that this study does not use (Sect. 2.7) and cannot be taken by inference from the
495 composition and cloud changes reported here.

4 Conclusions

Using GISS ModelE2.1–MATRIX-VBS and the DPEC early-peak-net-zero-clean-air pathway, we replaced anthropogenic
emissions only over China while holding emissions outside China at the 2019 CEDS baseline, in two simulations with
nudged winds and prescribed sea-surface temperature that share their meteorology. Over 2031–2050 the Chinese emissions
500 of SO₂, black carbon and organic carbon are lower by 86 %, 83 % and 66 %. The analysis is restricted to the composition,
number, size and activation fields of the aerosol scheme and to the droplet number and optical depth of the clouds that form
on it.

The bulk response is sub-linear at the source and is exported downwind. The sulfate and black-carbon columns over China
are lower by 24 % and 55 %, because China changes from a net exporter to a net importer of both. The reduction extends
505 across the North Pacific to North America and the Arctic, where it is largest in the free troposphere. There is no response
above 1 % over the Middle East or South Asia.

The emission reduction removes sulfate preferentially from BC-containing particles. Sulfate on BC-containing particles is
lower by 41 % over China and sulfate on BC-free particles by 15 %, and the ratio of about three between the two persists
along the outflow as far as the Arctic. The share of the sulfate column carried on BC-containing particles falls from 35 % to
510 28 % over China and from 20.5 % to 18.9 % in the Arctic. A closed process budget shows that the fraction of newly formed



sulfate delivered to the BOC population falls from 36 % to 25 % while the fraction delivered to organic–sulfate particles rises from 29 % to 40 %, and that condensation and coagulation onto the BOC population weaken equally, by about 70 %. Particle number in the Chinese boundary layer falls by 26 % and the number of BC-containing particles by 65 %. The coated carbonaceous particles that remain are larger and carry about one third more sulfate per unit black carbon, whereas the accumulation mode and fresh black carbon become smaller; far from the source the BC-containing particles are instead slightly smaller and less coated. The number of activated particles over China falls by 23 %, cloud-top droplet number by 29 % and the optical depth of water clouds by 6 %, and 85 % of the loss of activated particles is a loss of BOC particles. The effect of the emission reduction cannot be described by a change in bulk aerosol mass alone. The largest changes are in how many particles there are, how large they are and which of them carry the sulfate, and these changes reach well beyond the region in which emissions are reduced. Assessments of aerosol emission changes would therefore benefit from resolving mixing state and particle number explicitly alongside bulk abundance. Five limits of the design bound these conclusions. Emissions change only over China, so the contribution of transport from outside the region is at its maximum, and the perturbation is the response to Chinese anthropogenic emissions about 85 % below the CEDS 2019 level rather than a projection of the change between the present day and 2050. Winds are nudged and sea-surface temperature is prescribed, so the simulations contain no circulation, precipitation or temperature response to the emission change, and the small statistical uncertainty that follows from the shared meteorology is not a measure of structural uncertainty. The process terms are column integrals and net coagulation is a residual, so the budget gives the net gain or loss of each population but not which populations exchange mass or at what height. No observational product resolves the ACC, BOC, OCS and BCS populations, so the Control share of sulfate on BC-containing particles, 35 % in the column and 50 % at the surface over China, is unconstrained by observation and is the quantity most in need of measurement; the activation diagnostic is likewise used only in relative terms. The radiative and climate consequences of the changes reported here remain to be established, with the optical and radiative diagnostics this study does not use (Sect. 2.7) and a free-running ensemble. Single-species emission experiments, and comparison with single-particle and black-carbon coating measurements in Chinese outflow before and after the emission reductions of the last decade, would provide the most direct tests of the reallocation described here.

Code and data availability

CEDS, DPEC and the bias-corrected CMIP6 meteorological dataset are available from the sources cited in Sect. 2. Processed model output, regional statistics, plotting data and the complete set of Python scripts used for post-processing and figure production are archived at Zenodo (DOI: 10.5281/zenodo.23131046). The release version and licence will be specified when the Zenodo record is published. The archive includes the scripts that produce every quoted number (analysis_v8.py) and every figure (figures_v8.py) from the annual-mean model output.



Author contributions

CYG and JF conceptualized the study. JF and ML designed the experiments and performed the model simulations. JF and CYG processed and analysed the data and prepared the figures. JF and CYG wrote the original draft, and all authors reviewed and edited the manuscript. All authors approved the submitted version.

Competing interests

The authors declare that they have no conflict of interest.

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