



Aerosol emission estimates through SPEXone/PACE data assimilation in ECHAM6.3-HAM2.3: implications for aerosol radiative forcing

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Abstract. Aerosols exert an effective radiative forcing (ERF_{aer}) on climate through directly scattering and absorbing solar radiation (ARI) and indirectly interacting with clouds (ACI). Aerosol emissions, due to limited observational constraints, remain among the largest sources of uncertainty in modeled ERF_{aer} . Here, we estimate aerosol emissions globally by assimilating observations from SPEXone/PACE for the period 2024-08 to 2025-07, using a local ensemble transform Kalman smoother in combination with the global aerosol–climate model ECHAM6.3-HAM2.3. We assimilate aerosol optical depth at 550 nm (AOD_{550}), Ångström exponent between 440 nm and 670 nm ($AE_{440-670}$), and single scattering albedo at 440 nm (SSA_{440}) and additionally evaluate aerosol absorption optical depth at 440 nm ($AAOD_{440}$). The data assimilation experiment (DAS) reduces root mean square error (RMSE) against SPEXone for all four variables, with global mean bias improving from -0.017 to -0.001 for AOD_{550} and from 0.336 to 0.183 for $AE_{440-670}$. Validation against independent AERONET and MODIS observations likewise shows improvements. The emission estimates based on DAS lead to relatively more emissions of coarse particles, particularly over the ocean, and more weakly absorbing emissions, particularly over biomass-burning regions. Compared to the control experiment (CTL), DAS results in a more negative ERF_{aer} (-2.16 W m^{-2} vs -1.34 W m^{-2}), with changes of -0.53 W m^{-2} from ARI and -0.29 W m^{-2} from ACI. By assimilating SPEXone observations for aerosol emission estimation, this study highlights the critical role of accurate emissions from both natural and anthropogenic sources in determining aerosol radiative forcing.

1 Introduction

Aerosol–climate models are key to properly understand the impact of aerosols on climate; however, their uncertainties play a significant role in climate assessment and in understanding aerosol processes. The latest IPCC AR6 report showed that anthropogenic aerosols exert a significant cooling effect on the climate (Forster et al., 2023) and might partly offset warming



from greenhouse gases (IPCC, 2023, Chapter 6). For future policies related to clean air and geoengineering, it is important to understand aerosol processes. The IMO2020 policy (IMO, 2020), for example, highlights the trade-off between cooling from aerosols and potential health concerns (Yuan et al., 2024). However, large model uncertainties in aerosol effective radiative forcing (ERF_{aer}), consisting of a component due to aerosol–cloud interactions (ERF_{aci}) and a component due to aerosol–radiation interactions (ERF_{ari}), hamper future predictions of climate change (Watson-Parris and Smith, 2022).

Modeling studies using perturbed parameter ensembles (PPEs) have shown that uncertainty in model parameters related to aerosols (emissions, properties, processes) contribute substantially to the overall uncertainty in ERF_{aer} , and may even contribute more to the uncertainty in ERF_{aci} than cloud parameters (Jia et al., 2026a). Here, emissions of aerosols and precursor gases are one of the largest drivers of ERF_{aer} uncertainty (Jia et al., 2026a; Bhatti et al., 2026; Regayre et al., 2026, 2018; Lee et al., 2011), and the forcing impact depends strongly on emission type. Different emission types are represented in climate models at varying spatial and temporal resolutions and are subject to significant uncertainties.

Aerosol emissions from anthropogenic sources are mostly based on bottom-up inventories. One example is Community Emissions atmospheric Data System (CEDS), which provides historical emissions up to present-day emissions by harmonizing existing inventories while remaining transparent and updatable (Hoesly et al., 2018). These datasets are available only at monthly resolution, lacking day-to-day variability. Inventories are first gathered for each country and separated across multiple sectors. Furthermore, proxy data are used to convert country-level emissions to global gridded emissions. However, assigning emissions to the correct sectors, sparse data (particularly in low- and middle-income regions), and preliminary data for the most recent period all contribute to imperfections in these emission estimates (McDuffie et al., 2020; Hoesly and Smith, 2018; Hoesly et al., 2018).

Emissions from biomass burning are related to aerosol emissions from open fires. These datasets are typically derived from top-down approaches by converting satellite observations of fire radiative power and/or burned area to aerosol emissions using emission factors. Many inventories are based on MODIS observations. For example, the fifth version of the Global Fire Emissions Database (GFEDv5) (Van Der Werf et al., 2025) is based on observed burned area; the Fire Inventory from NCAR (FINN2.5) is based on a combination of MODIS active-fire counts and VIIRS observations (Wiedinmyer et al., 2023); and the Global Fire Assimilation System (GFASv1.2) is based on fire radiative power from MODIS (Kaiser et al., 2012; Di Giuseppe et al., 2016). Each inventory differs in accuracy, detection thresholds, and timeliness. When using GFASv1.2, a global scaling factor of 3.4 is recommended to compensate for the underestimation of AOD in the ECMWF model (Kaiser et al., 2012); applying this factor in ECHAM6.1-HAM2.2 also brings the BC lifetime into better agreement with AeroCom models (Veira et al., 2015). However, Tsikerdekis et al. (2023) showed through data assimilation that the optimal scaling factor may be lower and may differ between OC and BC. For the most recent IFS version, a scaling factor of 1.5 has been applied (A. Menochet, personal communication, 2025).

Natural emissions are parameterized as a function of near-surface wind speed for mineral dust (Tegen et al., 2002) and sea spray (Long et al., 2011), with sea spray additionally depending on sea-surface temperature (Sofiev et al., 2011). For dust, surface properties are also taken into account (Zhang et al., 2012; Tegen et al., 2002). Parameterizations in models are challenging because they must compensate for the coarse resolution of aerosol–climate models (in this study approximately



200 km). In particular, the nonlinear relationship between dust emission and wind speed poses a fundamental challenge for accurate dust emission estimates at coarse spatial resolution (Leung et al., 2023; Zhang et al., 2016).

To estimate the uncertainty of commonly used emission inventories, previous studies compared anthropogenic emission inventories for the year 2005 and reported a max/min ratio of 1.28 for BC and 1.32 for SO₂ (Granier et al., 2011). However, these ratios do not directly reflect the true uncertainties, as the inventories may share similar assumptions and underlying data.

For dust emissions, the AeroCom I ensemble showed a relative diversity (standard deviation divided by mean) of 58.3% with a max/min factor of 8.39 (Huneeus et al., 2011). Sea-spray emissions can differ by an order of magnitude among source functions, spanning 3000–70 000 Tg yr⁻¹ even when outliers are excluded (Grythe et al., 2014). For biomass-burning emissions, a comparison of six datasets for the year 2008 shows relative diversities of 52.1% (max/min factor 3.36) for BC and 53.2% (max/min factor 3.77) for OC (Pan et al., 2020).

PPEs have made significant strides in quantifying and constraining these uncertainties; however, the resulting parameter adjustments do not account for temporal variability and are applied at coarse regional (Regayre et al., 2026), or more commonly global, scales.

Data assimilation offers a complementary, spatially and temporally resolved way to constrain aerosol fields against observations, and is widely adopted in aerosol studies. Operational systems such as CAMS assimilate multiple satellite AOD products with a 4D-Var scheme to constrain aerosol concentrations (Garrigues et al., 2023, 2022). Kahnert et al. (2026) additionally assimilate observations from the European E-PROFILE network of ground-based lidars and ceilometers, improving the simulated vertical distribution of attenuated-backscatter and near-surface particulate matter (PM). Using the Local Ensemble Transform Kalman Filter (LETKF), Di Tomaso et al. (2022) assimilate coarse-mode dust optical depth from MODIS retrievals to produce a desert-dust reanalysis (2007–2016) covering Northern Africa, the Middle East and Europe. These studies mainly estimate concentrations, whereas here we estimate emissions.

Zhang et al. (2015) assimilated OMI AAOD to constrain BC emissions over Southeastern Asia, finding that anthropogenic emissions had to increase by up to 500% relative to bottom-up inventories to reproduce the observed absorption. This illustrates both how strongly absorption could constrain BC and why accurate absorption observations matter. Multi-angle polarimeters (MAPs) such as POLDER/PARASOL and the next-generation SPeXone/PACE retrieve aerosol absorption with high accuracy (Hasekamp and Landgraf, 2007; Mishchenko and Travis, 1997). POLDER/PARASOL products have been assimilated to estimate BC, organic aerosol and dust emissions (Chen et al., 2019, 2018) and to quantify aerosol absorption and its direct radiative forcing (Chen et al., 2022). Based on the POLDER retrievals, Tsikerdekis et al. (2021) estimated aerosol mass mixing ratios and Tsikerdekis et al. (2023) estimated aerosol emissions by simultaneously constraining aerosol load, size and absorption, and they have shown that omitting size and absorption information can lead to misinterpretation of the emission estimates.

Assimilating satellite observations for aerosol emissions is nonetheless challenging. The data volume is large, and a time lag separates the moment emissions occur from the moment their signal is observed, because transport, aging and deposition redistribute the aerosol beforehand. To handle the data volume efficiently, we build on the LETKF (Hunt et al., 2007; Miyoshi and Yamane, 2007). In this study we apply its smoother extension, which uses the filter iteratively. The Local Ensemble



Transform Kalman Smoother (LETKS) accounts for the temporal correlation between emissions and their delayed observed signal, and is therefore suited to estimating time-resolved emission fluxes.

LETKS was applied for CO₂ flux estimation by Peters et al. (2005) and later adapted for aerosol emission fluxes by Schutgens et al. (2012), who assimilated MODIS Terra AOD₅₅₀ over ocean and AERONET AOD₆₇₅ and AE_{440–870} over land. As noted above, Tsikerdekis et al. (2023) used POLDER observations of AOD₅₅₀, AE_{550–865}, and SSA₅₅₀ together with the ECHAM-HAM aerosol climate model and LETKS to constrain aerosol properties. The aforementioned studies demonstrated that estimating aerosol emissions requires accurate and diverse observational data to reduce emission uncertainties effectively.

The SPEXone multi-angle spectro-polarimeter aboard the NASA’s PACE mission, launched in February 2024, provides unprecedented retrievals of aerosol properties (Fu et al., 2025), including aerosol amount, size, and absorption. This is especially valuable over land, where emission sources are diverse and accurate constraints on aerosol type are needed for data assimilation. In contrast to Tsikerdekis et al. (2022b), where synthetic SPEXone data were used in an observing system simulation experiment (OSSE), we used real SPEXone observations to constrain natural and anthropogenic aerosol emissions in the ECHAM6.3-HAM2.3 model. Specifically, we estimated emissions from sea salt (SS), dust (DU), black carbon (BC), organic carbon (OC), and sulfur species (DMS, SO₄, and SO₂). The LETKS framework enables aerosol emission estimation at high temporal resolution (2-day cycles) by assimilating SPEXone observations. Although the estimates are technically performed at the grid-cell level, the prior assumes a spatially correlated emission field, which limits the possibility of highly fluctuating estimates between neighboring grid cells. The result is a spatially coherent and temporally resolved representation of aerosol emissions.

This work aims to estimate aerosol emissions from SPEXone observations using the data assimilation system LETKS, assimilating AOD₅₅₀, AE_{440–670} and SSA₄₄₀. We investigate how these estimates differ from the prior and their impact on aerosol properties, cloud properties, and forcing. Section 2 describes the observations used for data assimilation and validation, the model description and emission inputs, and the data assimilation scheme, including the choice of the settings. Section 3 presents the results, starting with a validation of the assimilation against SPEXone observations and an evaluation against independent retrievals from AERONET and MODIS. We then quantify the emission changes by data assimilation and their impacts on modeled aerosol and cloud properties, and ERF_{aer}. The main findings are summarized and discussed in Sect. 4.

2 Data and methods

2.1 Observations

In this study, we used the SPEXone retrieval products (version based on Fu et al., 2025) of aerosol optical depth at 550 nm (AOD₅₅₀), Ångström exponent between 440 nm and 670 nm (AE_{440–670}), and single scattering albedo at 440 nm (SSA₄₄₀) for data assimilation. The Level 2 (L2) retrieval products are aggregated onto a 1° × 1° grid and filtered according to the quality flag (keeping only quality flag = 0). The data assimilation uncertainties are parameterized as a function of AOD₅₅₀, based on an evaluation of SPEXone L2 uncertainties against the Aerosol Robotic Network (AERONET) observations (Fig. C1). The parameterization yields larger uncertainties in AE_{440–670} and SSA₄₄₀ as AOD₅₅₀ decreases. Figure C2 shows the uncertainties



after aggregating the data to a $1^\circ \times 1^\circ$ grid, as formatted for input to the data assimilation system. For the aggregated data we established lower limits of 0.015 for AOD_{550} , 0.05 for $\text{AE}_{440-670}$, and 0.01 for SSA_{440} . The lower limits ensure that AOD_{550} , $\text{AE}_{440-670}$, and SSA_{440} uncertainties have a minimum value when the L2 uncertainties are aggregated into gridded uncertainties, which are computed as the standard deviation divided by the square root of the number of observations.

We use independent observations from the ground-based AERONET network and Moderate Resolution Imaging Spectroradiometer (MODIS) to evaluate the effectiveness of our data assimilation. The model evaluation in Sect. 3.2 for AOD_{550} and $\text{AE}_{440-670}$ is based on AOD from the AERONET Level 1.5 Version 3 direct-sunlight retrieval product (Holben et al., 2001), interpolated to the required wavelength. The uncertainty in AERONET AOD is 0.01 at mid-visible wavelengths (Giles et al., 2019; Eck et al., 1999). SSA_{440} and AAOD_{440} are taken from the AERONET Level 1.5 almucantar inversion products (Dubovik et al., 2002). Following Dubovik et al. (2000), the retrieval uncertainty in SSA_{440} is ± 0.03 for water-soluble aerosols when $\text{AOD}_{440} > 0.2$, and for biomass-burning and dust aerosols when $\text{AOD}_{440} \geq 0.5$. For Version 3, more comprehensive per-retrieval uncertainty estimates are available (Sinyuk et al., 2020).

Additionally, MODIS Collection 6.1 Level 3 daily data (MODIS Atmosphere Science Team, 2017) are used to evaluate the modeled AOD_{550} and $\text{AE}_{550-870}$ at $1^\circ \times 1^\circ$ resolution. The wide swath of 2330 km provides near-global daily coverage (Barnes et al., 1998), extending the evaluation beyond the spatially sparse AERONET network. For AOD_{550} , we use the combined product, which merges the Deep Blue (DB) retrieval over land and the Dark Target (DT) retrieval over ocean, with an expected error of $\pm(0.05 + 15\% \text{ AOD})$ over land and $+(0.04 + 10\% \text{ AOD}) / -(0.02 + 10\% \text{ AOD})$ over ocean (Levy et al., 2013). We derived $\text{AE}_{550-870}$ over ocean from the Dark Target AOD retrievals at 550 nm and 870 nm, with uncertainties for AOD of $+(0.03 + 20\% \text{ AOD})$ (Sayer et al., 2013).

Appendix A provides the formulas used to derive SPEXone AAOD_{440} , interpolate AOD for AERONET, and calculate AE at wavelengths between 440 nm and 670 nm for AERONET and AE at wavelengths between 550 nm and 870 nm for MODIS.

2.2 Model and emissions

We use the global aerosol–climate model ECHAM6.3-HAM2.3, hereafter referred to as ECHAM-HAM, to link aerosol emissions with atmospheric optical properties. The model is run at T63L31 resolution, which corresponds to a horizontal resolution of $1.875^\circ \times 1.875^\circ$ (roughly $208 \text{ km} \times 208 \text{ km}$) and covers the entire troposphere up to 10 hPa. The model atmosphere is nudged to ERA5 meteorological data. Nudging frequencies are 24 h for temperature and surface pressure, 48 h for divergence and 6 h for vorticity. The ECHAM-HAM model consists of the general circulation model ECHAM, developed by the Max Planck Institute for Meteorology (MPI-M) in Hamburg, Germany (Stevens et al., 2013), and the aerosol module HAM, which is responsible for aerosol representation, aerosol emissions, aerosol processes, and the activation of aerosols into cloud droplets (Tegen et al., 2019).

Aerosols and their processes are represented with the M7 “pseudomodal” scheme (Vignati et al., 2004), comprising seven modes classified by size and solubility, ordered from smallest (nucleation) to largest (coarse): nucleation soluble (NS), Aitken soluble (KS), Aitken insoluble (KI), accumulation soluble (AS), accumulation insoluble (AI), coarse soluble (CS) and coarse insoluble (CI). Each mode is characterized by the total particle number and the mass of a subset of the following species:



DU, BC, OC, SS, and SO_4 . For further details, see Vignati et al. (2004). Particles within a mode are assumed to be internally mixed, while the different modes remain externally mixed from one another. The aerosol processes in the M7 module include nucleation of sulfuric acid–water droplets, coagulation, condensation of sulfuric acid, and aerosol water uptake. A budget study of these processes is presented in Schutgens and Stier (2014), highlighting the different processes that control the evolution of aerosol number and mass. Independent of the M7 module, the HAM2.3 module includes sulfur chemistry to treat DMS, SO_2 , and gas- and aqueous-phase sulfate (Tegen et al., 2019).

Table 1. Emission inventories by sector and species used in this study.

Sector	Species	Emissions
Aircraft	BC	CEDS, 2022 and 2023
Biogenic	OC	aerocom_II, 2000
Domestic	BC, OC, SO_2	CEDS, 2022 and 2023
Energy	BC, OC, SO_2	CEDS, 2022 and 2023
Fire	BC, DMS, OC, SO_2	GFAS, 2024 and 2025
Industry	BC, OC, SO_2	CEDS, 2022 and 2023
Ships	BC, OC, SO_2	CEDS, 2022 and 2023
Ocean	DMS	Lana et al. (2011)*
Terrestrial	DMS	aerocom_II, 2000
Transport	BC, OC, SO_2	CEDS, 2022 and 2023
Waste	BC, OC, SO_2	CEDS, 2022 and 2023

Note: Primary SO_4 emissions are specified as 2.5 % of the SO_2 emissions. *Oceanic DMS fluxes are computed online from prescribed seawater DMS concentrations (Lana et al., 2011) using a gas-exchange parameterization (Nightingale et al., 2000).

In the HAM module aerosol emissions are treated by sector and species, separately. An overview of the emission inventories used in this study is provided in Table 1. Anthropogenic emissions from fossil fuel combustion (FF) are based on the gridded bottom-up CEDS inventory (v2025_04_18) (Hoesly et al., 2024). Since the CEDS data are currently unavailable for 2024 and 2025, emissions from 2022 and 2023 were used instead. Biomass burning (BB) emissions are derived from the CAMS GFAS near-real time (NRT) daily product (Copernicus Atmosphere Monitoring Service, 2022), which covers all open fire sources. Following Kaiser et al. (2012), BB emissions are scaled up globally by a factor of 3.4. This scaling factor is recommended for use with GFAS emissions in ECHAM-HAM. Aerosol emissions from natural sources are parameterized: SS emissions are based on surface wind and sea surface temperature (Long et al., 2011; Sofiev et al., 2011), while DU emissions (Stier et al., 2005; Tegen et al., 2002) depend on surface wind, soil properties, and source region; they include MSG-based Saharan dust sources (Heinold et al., 2016) and updated East Asia soil properties.



2.3 Data assimilation

To estimate aerosol emissions, we apply the Local Ensemble Transform Kalman Smoother (LETKS) to assimilate SPEXone retrieval products AOD₅₅₀, AE₄₄₀₋₆₇₀, and SSA₄₄₀. Most of the setup followed the methodology of Tsikerdekis et al. (2022b); Schutgens et al. (2012).

LETKS is an efficient data assimilation approach. It is parallelizable and therefore well suited for high-dimensional problems such as estimating aerosol emissions grid cell by grid cell for many species simultaneously. In the following, we give a short overview of the data assimilation procedure; for more detailed information on the mathematical framework and efficient implementations we refer to the literature (Hunt et al., 2007; Miyoshi and Yamane, 2007). The workflow starts by creating a prior state: a three-dimensional field of scaling factors (time, longitude, latitude) for the emissions for each ensemble member, where the scaling factors are based on a log-normal distribution (Tsikerdekis et al., 2022b). The aerosol emissions are scaled by the scaling factors that are used by the ECHAM-HAM model to obtain the simulated observations. As summarized in Table S1, the perturbations are applied separately by aerosol species, mode, and emission sector.

LETKS combines the SPEXone observations, the simulated observations, and their uncertainties to estimate the posterior state by applying the Kalman equations:

$$\mathbf{x}^a = \mathbf{x}^b + \mathbf{P}^a \mathbf{H}^T \mathbf{R}^{-1} (\mathbf{y} - \mathbf{H} \mathbf{x}^b), \quad (1)$$

$$\mathbf{P}^a = (\mathbf{I} + \mathbf{P}^b \mathbf{H}^T \mathbf{R}^{-1} \mathbf{H})^{-1} \mathbf{P}^b. \quad (2)$$

Here, in Eqs. (1) and (2), $\mathbf{x}^b$ is the prior state vector (emission perturbation factors) and $\mathbf{x}^a$ is the posterior state vector; $\mathbf{P}^b$ is the prior error covariance matrix and $\mathbf{P}^a$ is the posterior error covariance matrix; $\mathbf{H}$ is the observation operator (in our case ECHAM-HAM and the co-location from the model grid to the SPEXone grid); $\mathbf{y}$ is the observation vector representing the SPEXone retrieval products; and $\mathbf{R}$ is the observation error covariance matrix based on AERONET evaluation (Fig. C2). We assume that observation uncertainties are uncorrelated to make the data assimilation more efficient.

Most settings for the data assimilation follow Tsikerdekis et al. (2023); a brief summary is provided here. Model uncertainty $\mathbf{P}^b$ is represented by an ensemble of 32 members, each assigned its own set of emission perturbation factors. Emission perturbations for any given two-day period are estimated iteratively: first constrained by observations from that two-day window, then successively refined using observations from the following two-day periods, with the mixing ratios updated between each step by the forward model using the preliminary posterior perturbations. For further details, see Tsikerdekis et al. (2022b), Schutgens et al. (2012), and Peters et al. (2005). The multiplicative inflation factor is set to 1, assuming that the forward model's non-linearity ensures sufficient spread.

2.4 Experimental setup

The analysis of all experiments covers the one-year period from 2024-08-01 to 2025-07-31. The control experiment (CTL) consists of an ensemble of perturbed model realizations using default settings without data assimilation. The data assimilation experiment (DAS) optimizes emissions based on the aforementioned data assimilation with SPEXone observations. The model



is first spun up from 2024-01-01 to 2024-03-31, followed by a spin-up from 2024-04-01 to 2024-07-31 for CTL. The DAS
 205 is then initialized from the CTL restart and further spun up from 2024-07-01 to 2024-07-31 to allow the system to reach
 equilibrium.

In addition to the ensemble runs (CTL and DAS), we also perform single model runs to quantify the effects of the emission
 correction by DAS on present-day (PD) aerosol and cloud properties (Sect. 3.4) and ERF_{aer} (Sect. 3.5). For the single-model
 run, the ensemble-mean emissions are used. ERF_{aer} is estimated by differencing two single-model experiments: one with
 210 pre-industrial (PI) emissions and one with PD emissions. For CTL, default unperturbed emissions are used, while for DAS,
 the ensemble-mean emission estimates are applied. By assuming that natural emissions are identical between PI and PD, the
 DAS-PI experiment applies emission corrections only to natural aerosol sources. This results in four experiments in total. Each
 experiment includes a three-month spin-up period, with the constrained emission estimates applied from 2024-07-01 onwards.
 A comparison of AOD_{550} between the ensemble-mean and the single model run driven by the ensemble-mean emissions is
 215 shown in Fig. S1; the results have similar spatial patterns.

3 Results

3.1 Evaluation against SPEXone

Figure 1 shows the annual-mean spatial distributions of AOD_{550} (a), $AE_{440-670}$ (b), SSA_{440} (c), and $AAOD_{440}$ (d) for SPEX-
 one observations, DAS–SPEXone, and CTL–SPEXone differences. The column CTL–SPEXone shows that AOD_{550} is over-
 220 estimated over Canada and Siberia and underestimated over the Amazon basin, equatorial Africa, and India; $AE_{440-670}$ is
 overestimated globally; SSA_{440} is overestimated over the ocean and underestimated over land, particularly in biomass-burning
 regions near the equator, India, and eastern China; and $AAOD_{440}$ is overestimated over biomass-burning regions, Canada, and
 northern Siberia, with a mixed signal over equatorial Africa. DAS corrects most of these biases: As seen under DAS–SPEXone
 the AOD_{550} overestimation over Canada and northern Siberia is reduced, as is the underestimation over the Amazon basin,
 225 equatorial Africa, and India; the positive $AE_{440-670}$ bias is reduced most substantially over the ocean and dust source regions
 such as the Sahara; over land, SSA_{440} is strongly corrected and consequently $AAOD_{440}$.

DAS shows overall better agreement with SPEXone than CTL, consistent with the reduction in mean error (ME) and root-
 mean-square error (RMSE) for all parameters (Fig. 1). For RMSE specifically, DAS yields reductions of 0.041 (–25.0%) for
 AOD_{550} , 0.141 (–23.0%) for $AE_{440-670}$, 0.017 (–25.4%) for SSA_{440} , and 0.011 (–43.5%) for $AAOD_{440}$. Globally, based
 230 on 415 761 co-located data points, the Pearson correlation coefficient (r) is higher for DAS than CTL across all variables:
 AOD_{550} (0.60 vs 0.32), $AE_{440-670}$ (0.57 vs 0.43), SSA_{440} (0.25 vs 0.19), and $AAOD_{440}$ (0.61 vs 0.32).

Figure S2 shows kernel density estimate (KDE) plots for individual SPEXone–model comparisons. The effect of the data
 assimilation differs between ocean and land, reflecting differences in aerosol composition and abundance. DAS improves
 RMSE for all variables over both land and ocean. Significant improvements in ME are found for AOD_{550} over land (–0.009
 235 vs –0.031), $AE_{440-670}$ over ocean (0.145 vs 0.311) and land (0.289 vs 0.408), SSA_{440} over land (–0.011 vs –0.062), and
 $AAOD_{440}$ over land (0.001 vs 0.006). These improvements are consistent with the emission adjustments discussed in Sect. 3.3:

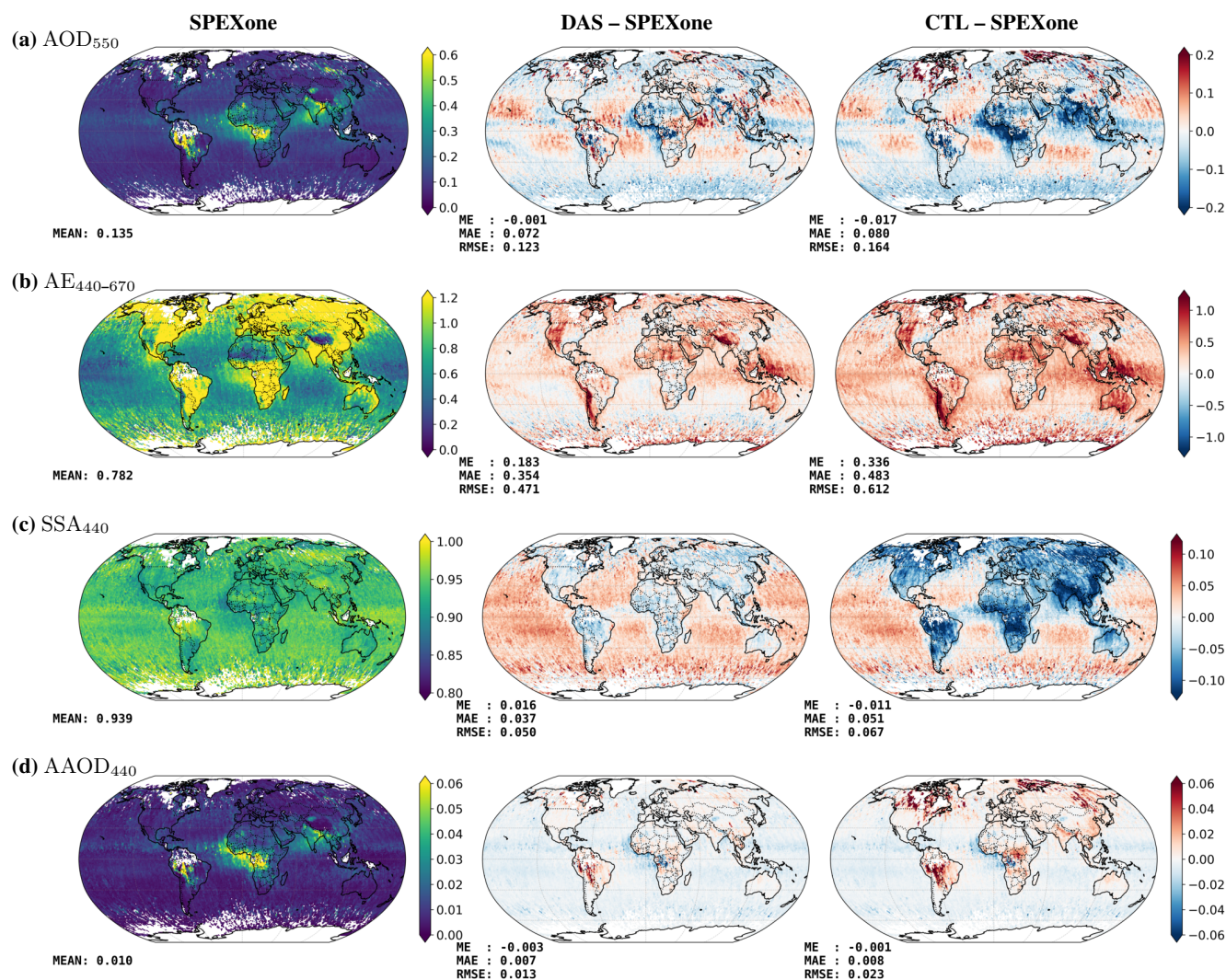


Figure 1. Comparison of time-averaged spatial distributions of (a) AOD₅₅₀, (b) AE₄₄₀₋₆₇₀, (c) SSA₄₄₀, and (d) AAOD₄₄₀ between SPEXone and ECHAM-HAM averaged over the study period (2024-08 to 2025-07). The model data are co-located to the SPEXone data in time and space. The first column shows the SPEXone retrieval product, the second column the bias DAS–SPEXone, and the third column CTL–SPEXone. Each panel includes a text box reporting summary statistics: the first column shows the mean value, while the second and third columns show the mean error (ME), mean absolute error (MAE), and root-mean-square error (RMSE).



increased coarse-mode SS emissions reduce the $AE_{440-670}$ bias over the ocean, while reduced BC emissions over biomass-burning regions increase SSA_{440} , both bringing the model closer to SPEXone observations (see the discussion in Sect. 3.3).

Over the ocean, SSA_{440} is overestimated in DAS (0.025 vs 0.007) and $AAOD_{440}$ is underestimated (-0.004 vs -0.003).
 240 The SSA_{440} overestimation is also visible in Fig. 1 and persists in regions without aerosol outflow. In outflow regions, CTL exhibits a negative SSA_{440} bias that DAS successfully reduces, likely as a consequence of less absorptive aerosol emissions over land being transported to the ocean. However, since this correction is confined to outflow regions, the positive bias in the remaining ocean areas is unchanged, and the overall ocean mean bias increases. This suggests that correcting aerosol emissions alone is insufficient to address structural model biases for SSA_{440} over the ocean. We note that recent algorithm developments
 245 for SPEXone aerosol products (Fu et al., 2026) suggest a possible low bias in SSA over the remote ocean for the data version used in this study.

Figure 2 provides a regional breakdown for ME, using the region definition shown in Fig. D1. Additional statistics are provided in the supplement for RMSE (Fig. S3) and for Pearson correlation coefficient r (Fig. S4). Focusing on AOD_{550} ME (Fig. 2), DAS tends to increase AOD_{550} over the ocean. Only over the Arctic Ocean (AO), the positive CTL bias ($+31.0\%$) is
 250 significantly reduced in DAS, though slightly overcorrected to a negative bias (-15.3%). Over land, the absolute AOD_{550} bias is reduced in all regions except Europe (EUR), where AOD_{550} decreases (DAS -4.4% vs CTL -2.6%). For $AE_{440-670}$, DAS consistently reduces the positive bias across all regions, suggesting a structural positive bias in the model that can be partly corrected by tuning emissions, particularly over the ocean. For SSA_{440} , DAS increases SSA_{440} in every oceanic region. Over the Tropical Pacific Ocean (TPO), South Pacific Ocean (SPO), and South Indian Ocean (SIO), the positive bias worsens even
 255 in regions where CTL is already positively biased. Over land, SSA_{440} also increases in all regions and generally shows better agreement with SPEXone observations, except over North America (NA; ME: -0.012 to $+0.013$) while over Oceania the bias changes sign with unchanged magnitude (AUS; ME: -0.012 to $+0.012$). As noted previously, the data assimilation struggles to constrain SSA_{440} over the ocean through emission adjustments alone; however, the correction over land may propagate to ocean regions through aerosol outflow and might increase the bias over ocean. ME of $AAOD_{440}$ decreases in nearly all regions,
 260 becoming more negative than CTL over the Tropical Atlantic Ocean (TAO), TIO, SAO, SIO, Africa (AF), and AUS. RMSE is lower for almost all variables and regions in DAS compared to CTL (Fig. S3). Exceptions, where DAS RMSE is higher than CTL RMSE, are confined to AOD_{550} in the Tropical Indian Ocean (TIO, $+8.9\%$ relative to CTL), South Atlantic Ocean (SAO, $+5.4\%$), Asia (ASIA, $+4.2\%$), and Tropical Pacific Ocean (TPO, $+2.9\%$). The worsening may be related to violations of the underlying Kalman filter assumptions, such as non-linearity and non-Gaussian error distributions for the emissions. It
 265 may also result from the simultaneous assimilation of AOD_{550} , $AE_{440-670}$, and SSA_{440} , where the specific combination of observed values may not be reproducible by the model which might indicate structural error in the model. Overall, these results demonstrate the ability of the data assimilation to improve the spatial representation of aerosol optical properties across most regions.

To assess the consistency of the data assimilation performance and to identify events associated with degraded performance,
 270 Figure 3 shows the ME as a function of time for DAS (blue) and CTL (orange), smoothed with a two-week rolling mean to reduce sampling noise and ensure a consistent number of data points throughout the averaging period. The horizontal colored

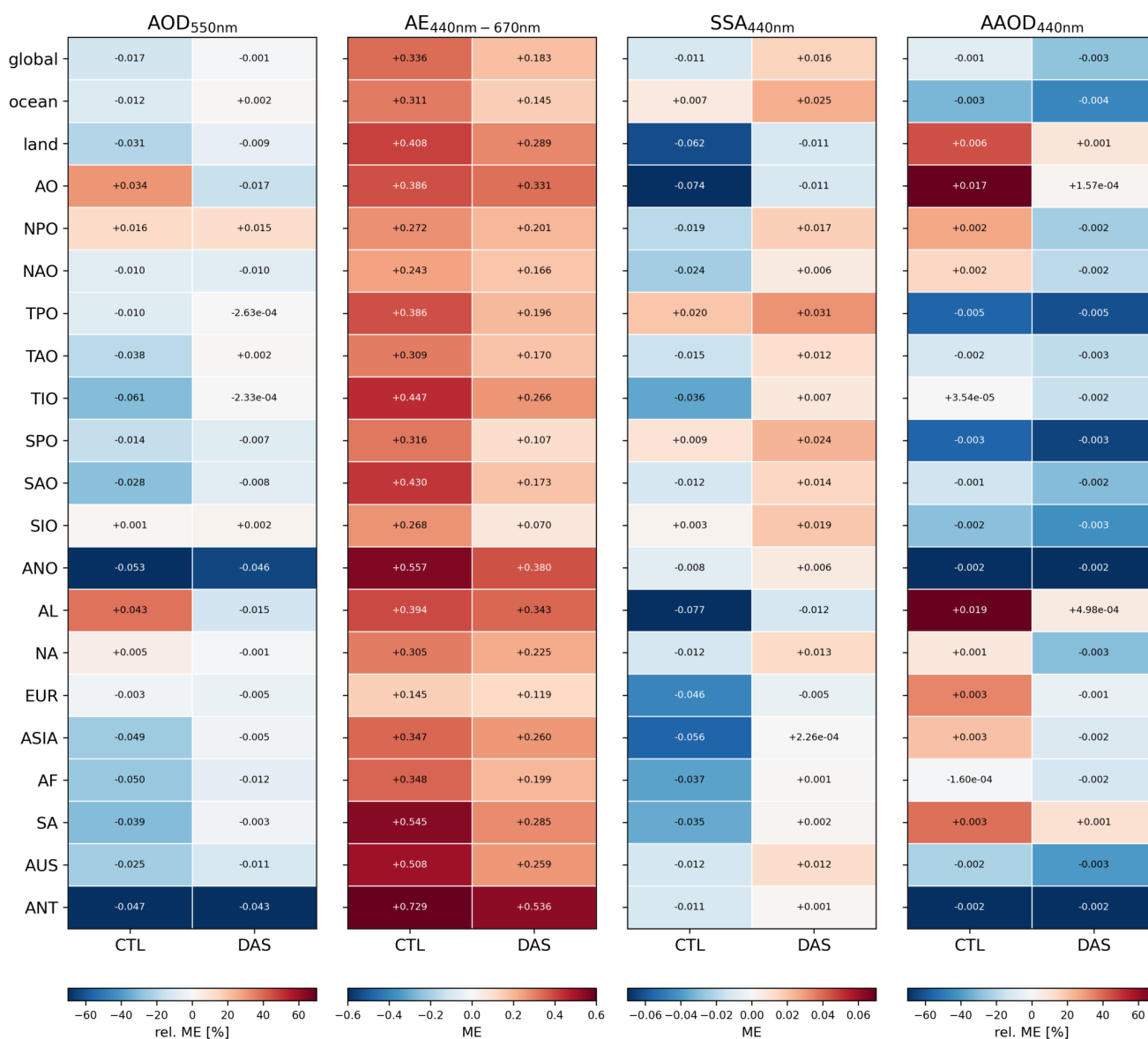


Figure 2. Mean error (ME) for each region of CTL and DAS against SPEXone, where the regions are defined according to Fig. D1 (Jia et al., 2021). For AOD₅₅₀ and AAOD₄₄₀ the colorbar is scaled relative to the SPEXone mean.

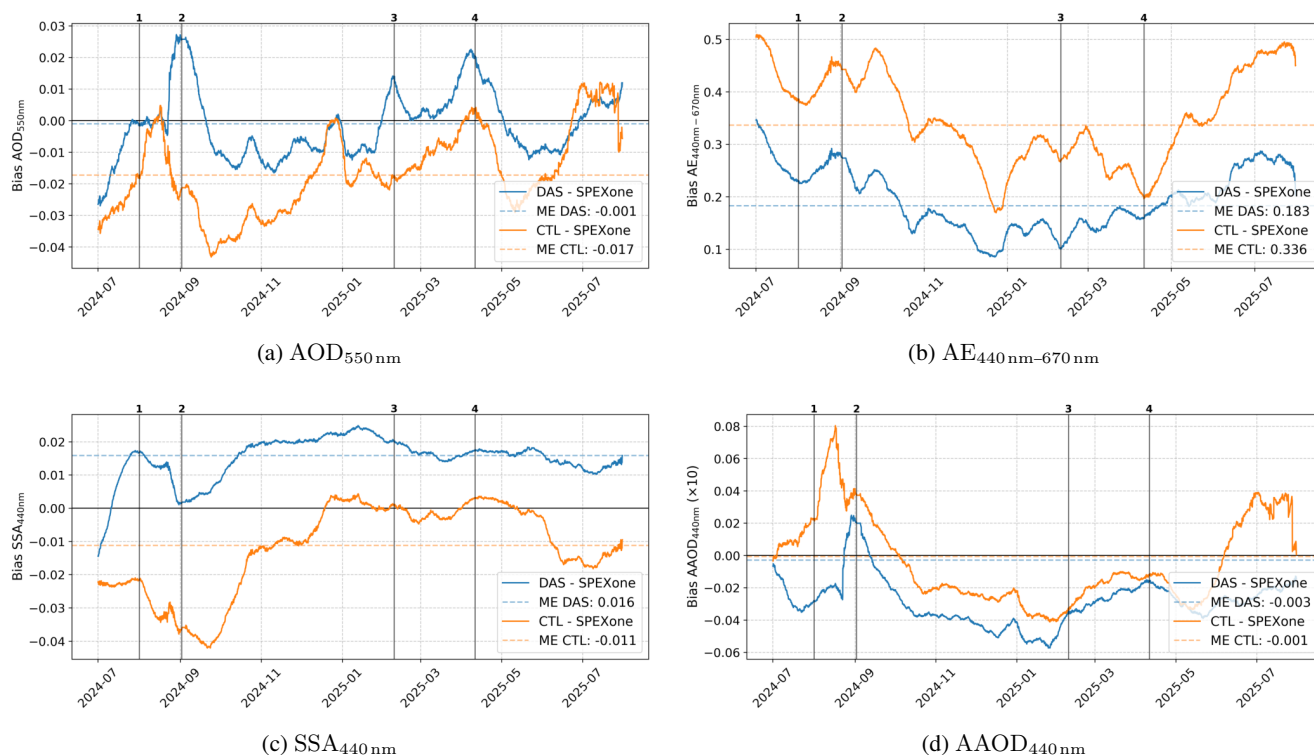


Figure 3. ME as a function of time, smoothed with a two-week rolling mean (one week before and after each data point). Blue and orange curves represent DAS and CTL, respectively. The horizontal colored dashed lines indicate the mean bias over the period 2024-08 to 2025-07, and the horizontal black line marks zero bias. The vertical numbered lines mark the dates referenced in the text. Ocean and land breakdowns are shown in Figures S5 and S6, respectively.

dashed lines indicate the mean bias over the entire period and the horizontal black line marks zero bias. Throughout the period, DAS improves agreement with observations for AOD₅₅₀ (a) and AE_{440–670} (b), with the persistent positive AE_{440–670} bias in CTL consistently reduced by DAS. For SSA₄₄₀ (c) and AAOD₄₄₀ (d), however, DAS exhibits a systematic positive and negative bias, respectively. An ocean–land breakdown is shown in Figures S5 and S6. AOD₅₅₀ improves in DAS over both ocean and land throughout most of the period. For AE_{440–670} and SSA₄₄₀, structural differences emerge between the two regions. Over the ocean, DAS consistently reduces the AE_{440–670} bias. However, SSA₄₄₀ becomes persistently more positively biased in DAS compared to CTL. Over land, both AE_{440–670} and SSA₄₄₀ improve in DAS, though the AE_{440–670} improvement diminishes from early 2025 until around June 2025, with DAS performance becoming comparable to CTL.

Figure S7 shows time series of the model ensemble spread including the global-mean SPEXone uncertainty, and Figure S8 shows the corresponding globally averaged values for comparison. DAS reduces the spread compared to CTL for all variables: AOD₅₅₀ (0.017 vs 0.043), AE_{440–670} (0.100 vs 0.221), and SSA₄₄₀ (0.003 vs 0.027), indicating that the data assimilation effectively constrains the emissions among the ensemble members. However, notable increases in the ensemble spread are



visible around marker (2) for AOD_{550} and SSA_{440} . The markers are discussed in the following. The vertical lines in Fig. 3
 285 mark the following dates: (1) 1 August 2024, (2) 2 September 2024, (3) 9 February 2025, and (4) 11 April 2025, where markers
 (2)–(4) coincide with considerably elevated AOD_{550} bias in DAS.

Marker (1) corresponds to the end of the spin-up period on 2024-08-01. Figure S7 shows a strong decrease in ensemble
 spread at the beginning of the experiment, suggesting that roughly three weeks of spin-up is sufficient for the ensemble to reach
 equilibrium. Note that the time required to reach equilibrium might depend on the time period, the availability of observations,
 290 and the discrepancy between the model and observations.

Marker (2) coincides with a large positive bias in DAS for AOD_{550} and $AAOD_{440}$, related to biomass-burning emissions
 in September and their subsequent outflow, as indicated in the monthly mean bias map for AOD_{550} (Fig. S9a, analogous to
 Fig. 1). Large emissions over equatorial South America and equatorial Africa at the beginning of September (not shown here)
 were transported over the TPO and TAO, resulting in a strong positive bias for DAS relative to CTL. White patches around the
 295 source regions and outflow regions in Fig. S9a indicate missing observations, suggesting that the data assimilation struggles to
 constrain emissions when aerosol loadings are high and observations near the source regions are absent. This is also reflected
 in the ensemble spread, where a notable increase in spread for AOD_{550} and SSA_{440} around marker (2) indicates difficulties in
 constraining biomass-burning emissions during this period.

Around markers (3) and (4), in early February and early April 2025 respectively, DAS shows a positive AOD_{550} bias
 300 originating from excessive dust emissions from Asian desert regions and aerosol emissions from India. The monthly mean bias
 maps for February (Fig. S9b) and April 2025 (Fig. S9c) suggest that large dust emissions from Asian source regions and their
 subsequent transport lead to an overestimation over the North Pacific Ocean (NPO) and NA. A particularly strong positive bias
 over the Arabian Sea is likely attributable to emissions from India. Similar as marker (2), observations near the source regions
 around the Taklamakan and Gobi deserts and their outflow regions are sparse during both periods.

The data assimilation is able to constrain emissions consistently throughout the remainder of the period. The main challenges
 305 arise when observations near strong emission sources are absent, as unconstrained emissions are transported to remote regions
 where they contribute to AOD_{550} biases far from the source. The largest AOD_{550} deviations are associated with biomass-
 burning emissions in equatorial South America and Africa, Asian dust outbreaks, and aerosol outflow over the Arabian Sea.
 Dust emissions are particularly challenging, as they are characterized by short but intense emission episodes and long transport
 310 distances. These difficulties might be related to the previously discussed limitations of the Kalman filter, including non-linearity
 of aerosol processes, non-Gaussian error distributions, and structural errors in the forward model. These periods and regions
 therefore represent a focus for future sensitivity studies assessing data assimilation performance.

3.2 Evaluation against AERONET and MODIS

In this section, we compare model output with independent observations from AERONET and MODIS, with model data
 315 co-located to each observational dataset in both time and space. The AERONET direct-sun measurements of AOD serve as
 the primary validation standard for satellite aerosol retrievals due to its high accuracy. MODIS complements AERONET by
 offering much broader spatial coverage, though its quantitative skill in retrieving aerosol size is limited, especially over land. So

we only focus on ocean when comparing AE. MODIS DT and MODIS DB have been benchmarked against satellite products, AERONET, and Maritime Aerosol Network (MAN) (Schutgens et al., 2020; Sayer et al., 2014).

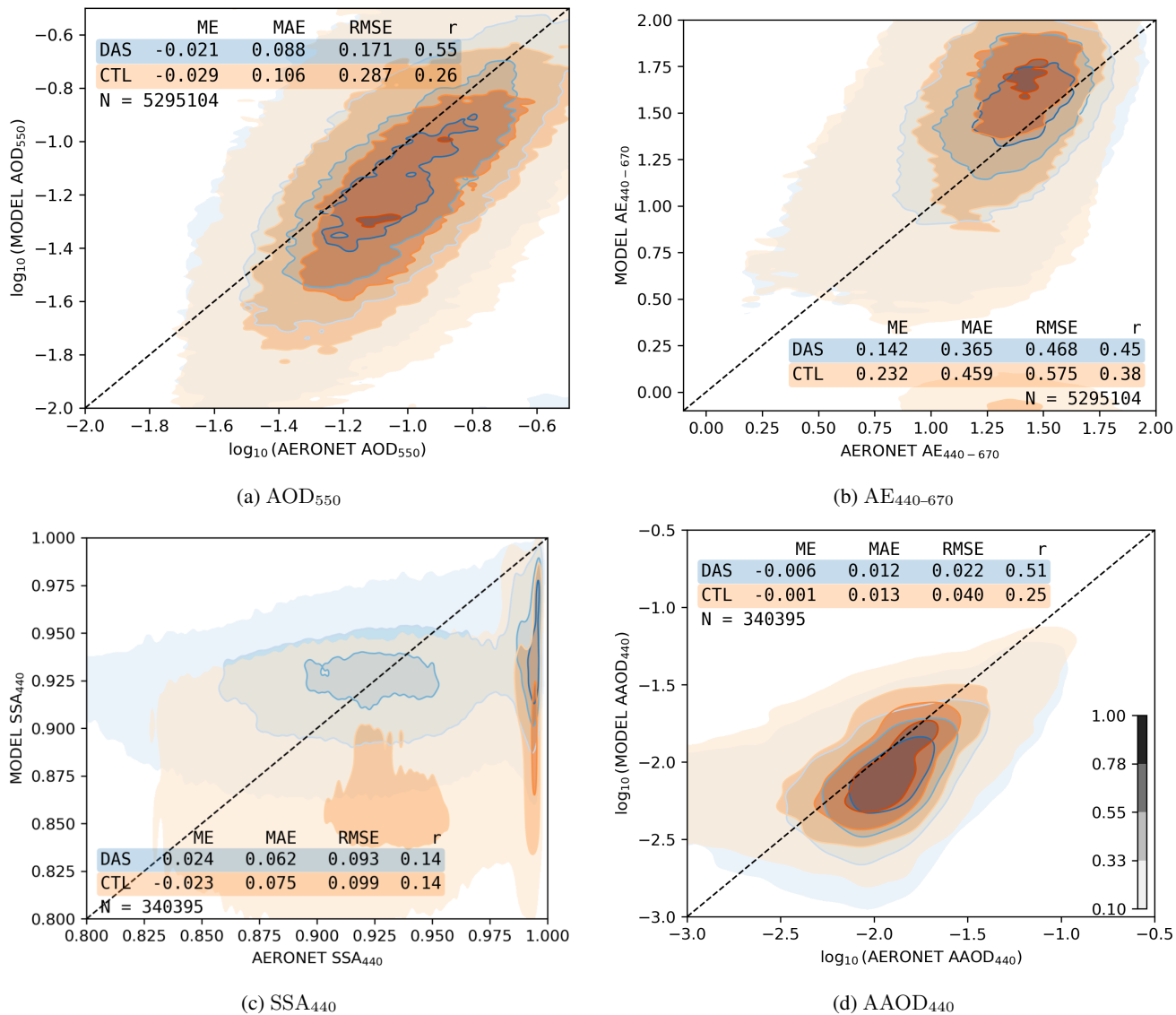


Figure 4. KDE plots of co-located DAS (blue) and CTL (orange) against AERONET observations, analogous to Fig. S2.

320 Figure 4 shows the comparison of CTL and DAS against AERONET measurements. It is clear that RMSE is lower for DAS than for CTL for all parameters: AOD₅₅₀ by 40.4%, AE₄₄₀₋₆₇₀ by 18.6%, SSA₄₄₀ by 6.0%, and AAOD₄₄₀ by 43.8% relative to the RMSE of CTL. ME improves for AOD₅₅₀ and AE₄₄₀₋₆₇₀. However, despite the overall improvement in RMSE relative to CTL, ME does not improve for SSA₄₄₀ and AAOD₄₄₀ in DAS, consistent with the SPEXone comparison (Fig. S2). Unlike



the SPEXone comparison, the SSA_{440} distribution shows two distinct peaks: one around 0.925, and another close to 1.0. DAS improves agreement at both peaks by correcting the underestimation in CTL, though the correction is more pronounced at the first peak. The unequal magnitude of the corrections flattens the SSA_{440} distribution, which may explain why r remains essentially unchanged (0.137).

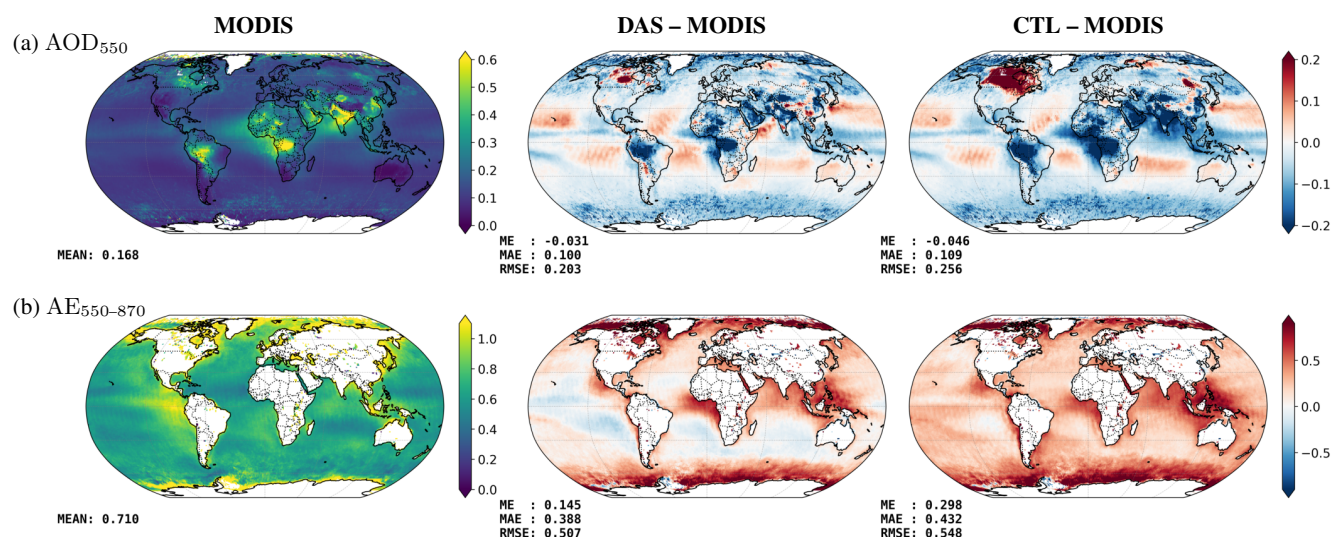


Figure 5. Time-averaged MODIS AOD₅₅₀ (a) and AE₅₅₀₋₈₇₀ (b) (left), and the differences DAS–MODIS (center) and CTL–MODIS (right).

The comparison with MODIS AOD₅₅₀ (Fig. 5a) reveals a spatial pattern broadly consistent with the SPEXone comparison (Fig. 1a): the model underestimates AOD₅₅₀ over anthropogenic source regions in Asia and biomass-burning regions in Africa and South America, and overestimates it over Siberia and boreal regions in Canada. DAS improves upon CTL, with ME changing from -0.046 to -0.031 , and RMSE from 0.256 to 0.203. Despite the similar spatial patterns, error statistics are generally larger for the MODIS–model comparison than for the SPEXone–model comparison. Figure 5b shows the comparison of MODIS AE₅₅₀₋₈₇₀ with model AE₅₅₀₋₈₆₅. An analogous comparison using SPEXone data is shown in Fig. S10. The model–MODIS comparison reveals a pattern broadly consistent with the model–SPEXone comparison: CTL remains positively biased (0.298), and DAS substantially reduces this bias (0.145). RMSE also improves from 0.548 to 0.507.

Figure S11 shows the KDE plots for AOD₅₅₀ and AE₅₅₀₋₈₆₅, using MODIS AE₅₅₀₋₈₇₀ in place of AE₅₅₀₋₈₆₅. As seen before for SPEXone and AERONET, DAS improves agreement with MODIS across all metrics for both variables, with r increasing from 0.28 to 0.44 for AOD₅₅₀ and from 0.37 to 0.42 for AE₅₅₀₋₈₆₅. A land–ocean breakdown for AOD₅₅₀ is also included (Fig. S11c,d). DAS improves both ME and RMSE relative to CTL, with a more pronounced improvement over land (RMSE: 0.412 to 0.265; -35.7%) than over the ocean (RMSE: 0.188 to 0.179; -5.1%).

To conclude, the SPEXone-based data assimilation yields a model that agrees not only better with SPEXone itself but also with independent observations, demonstrating the effectiveness of assimilating SPEXone data. DAS shows improved

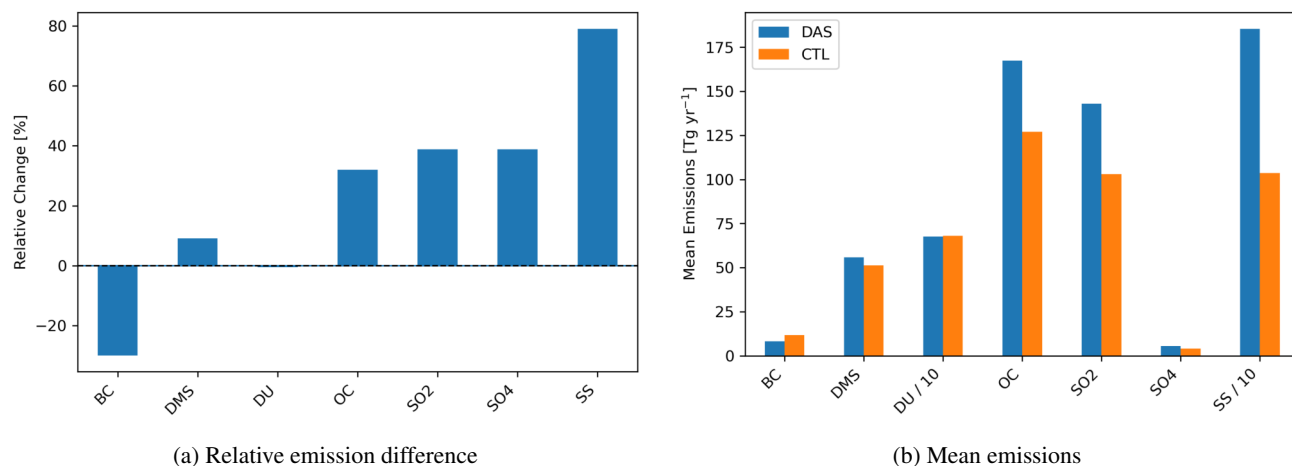


Figure 6. (a) Relative emission difference (DAS – CTL)/CTL and (b) mean emissions for CTL (orange) and DAS (blue). SS and DU emissions are scaled down by a factor of 10. The emission values are listed in Table 2.

agreement with AERONET observations, demonstrating its ability to constrain emissions such that the model better reproduces the actual aerosol state. On the other hand, DAS also brings the model closer to the spatially extensive MODIS observations, showing that even only assimilating relatively sparse SPEXone data can still enhance model performance on larger spatial scales, supporting the conclusion from a synthetic data assimilation experiment (Tsikerdekis et al., 2022b).

3.3 Aerosol emission estimation

In this section, we present the aerosol emission estimates from DAS and their changes relative to CTL. Figure 6 compares global aerosol emissions from CTL (orange) and DAS (blue) for the period 2024-08 to 2025-07, showing the relative change with respect to CTL (a) and the total global emissions averaged over time (b).

DAS produces substantial emission changes relative to CTL: DMS (+9.2%), SO₂ and SO₄ (+38.8%), OC (+32.0%), and BC (–29.9%). SS emissions increase significantly (+79.0%), driven primarily by the coarse mode (SS_{CS}: +89.3%), while fine-mode SS decreases (SS_{AS}: –29.5%). Total DU remains essentially unchanged (–0.5%), but its mode distribution shifts: the accumulation-insoluble mode increases substantially (DU_{AI}: +30.5%) while the coarse-insoluble mode decreases slightly (DU_{CI}: –1.2%). Overall, the emission adjustments result in coarser, less absorptive aerosols with greater total mass, consistent with the data assimilation reducing AE_{440–670} over the ocean and increasing SSA₄₄₀ and AOD₅₅₀ over land to improve agreement with SPEXone observations (Sect. 3.1).

Specifically, for fire emissions, BC decreases by 38.6% and OC increases by 37.0%, corresponding to GFAS scaling factors of 4.66 for OC (Fig. S12a) and 2.09 for BC (Fig. S12c). For the fossil fuel emissions, the adjustments are more moderate: OC increases by 28.6% and BC decreases by 19.4%, while biogenic OC increases by 11.6%. Additionally, it should be noted that 65% of BB OC emissions are emitted in soluble form (Stier et al., 2005), making them particularly important for clouds.

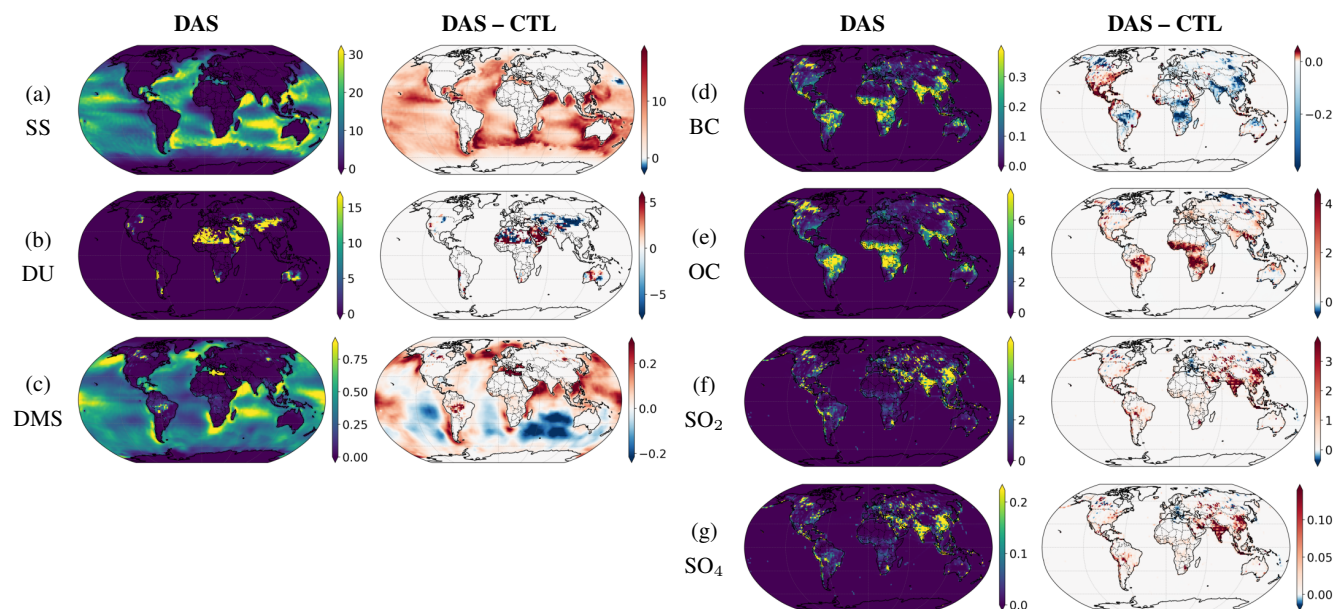


Figure 7. Time-averaged aerosol emissions in kilograms per square kilometer per day for DAS and next to it the mean difference DAS–CTL for each species. The color map for the difference field is piecewise-linear, comprising two slopes that meet at a center fixed at zero.

The emission changes also exhibit pronounced regional variability. The following discussion is based on the spatial distributions shown in Figure 7, which displays DAS emission estimates alongside the DAS–CTL difference for each species. Regional statistics are summarized in Figure 8, where the left panel shows the absolute difference (DAS–CTL) by species and region, and the right panel shows the relative change normalized by the mean CTL emissions.

For natural emissions, total SS emissions increase consistently across ocean regions (Fig. 7a). The mode breakdown (Fig. S13) reveals that this increase is driven by decreasing fine-mode SS (c) while increasing coarse-mode SS (d) globally, with the exception of polar regions, along the shores of Europe, East Asia and Antarctica. DU emissions (Fig. 7b) decrease over Europe (EUR, –14.9%) and Asia (ASIA, –21.6%), particularly around the Caspian Sea and Central Asia, but increase over the Sahara in Africa (AF, +10.7%). Over Asia, particularly over the Gobi Desert, DU shifts toward finer modes (ASIA DU_{AI}: +55.6%, DU_{CI}: –23.6%). This shift may explain the positive AOD₅₅₀ bias over the NPO and NA, as smaller particles are more readily transported over long distances. Marine DMS emissions (Fig. 7c) increase over most northern hemisphere ocean regions and coastal areas but decrease substantially over the southern Indian Ocean, consistent with the regional decrease in SS_{AS} in that area.

Biomass-burning and anthropogenic aerosol emissions likewise exhibit strong regional differences. BC, OC, and sulfate emission (Figs. 7d–g) changes vary substantially by region. Over biomass-burning regions near the equator, BC emissions decrease over South America (SA, –32.9%) and Africa (AF, –30.3%) while OC emissions increase (SA: +42.0%, AF: +60.0%). In contrast, over higher-latitude biomass-burning regions in Arctic Land (AL), such as northern Canada and Siberia,

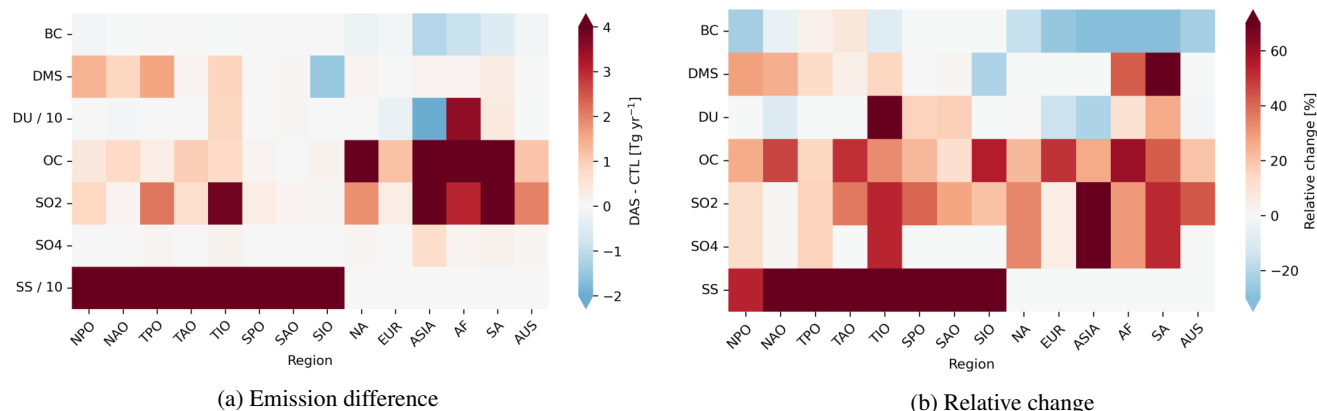


Figure 8. Panel (a) shows the emission difference DAS–CTL by species and region; DU, DU_{CI} , SS, and SS_{CS} are scaled down by a factor of 10. Panel (b) shows the relative emission difference $(DAS - CTL)/CTL$ by species and region, with values excluded where mean CTL emissions fall below 0.2 Tg yr^{-1} . Stippling indicates accumulation mode and hatching indicates coarse mode. The corresponding values are also listed in Table S2 for CTL and Table S3 for DAS.

both BC and OC emissions decrease (AL BC: -69.4% , OC: -14.5%). This suggests that emissions in higher latitudes are overestimated, while equatorial emissions are underestimated but too absorptive in CTL. Over India and China, BC emissions decrease while OC and sulfate emissions increase (ASIA: BC -40.4% , OC $+26.5\%$, SO_2 and SO_4 $+70.5\%$). A similar pattern is found over Europe and North America, though with a smaller sulfate increase: EUR (BC -27.0% , OC $+50.4\%$, SO_2 and SO_4 $+5.2\%$) and NA (BC -16.2% , OC $+23.0\%$, SO_2 and SO_4 $+33.7\%$). These regions are characterized by relatively high anthropogenic OC (Fig. S12b) and BC (Fig. S12d) emissions, coinciding with relatively high sulfate emissions. The data assimilation therefore appears to reduce strongly absorptive BC emissions while simultaneously increasing OC and sulfate emissions.

Finer inspection within the defined regions (Fig. D1) reveals notable sub-regional differences. Over NA, BC emissions decrease over Canada, while OC emissions decrease in the north and increase in the south; over the US, OC, BC, and sulfate emissions all increase. The strong sulfate increase over India and eastern China likely dominates the ASIA signal (SO_2 and SO_4 , $+70.5\%$). Note that the regional variability in the emission changes partly reflects the spatial structure of the underlying emissions themselves, where some of the defined regions are smaller than the correlation length of our perturbed emissions. Below the correlation length, meaningful variations of the perturbation are not expected; emissions are likely to be scaled by a constant factor throughout those small regions.

Furthermore, we compare our emissions estimates with those of Tsikerdekis et al. (2023), who used the same model and data assimilation system but assimilated POLDER observations for the year 2006. Compared to the POLDER-based experiment, our DAS yields lower SO_2 , lower BC, higher OC, comparable SS, and substantially lower DU emissions (DAS: $\sim 677 \text{ Tg yr}^{-1}$ vs POLDER DAS: $\sim 1419 \text{ Tg yr}^{-1}$, Table 2). These differences may partly reflect the different time periods analyzed: the POLDER study covers 2006, whereas our experiment covered 2024–2025, where high-emission events such as wildfires and



dust outbreaks are expected to differ substantially between periods. Additionally, our experiments used a scaling factor of 3.4 for wild fires emissions (instead of no scaling applied by Tsikerdekis et al., 2023). For dust specifically, Tsikerdekis et al. (2023) report CTL and DAS emissions of 1105 Tg yr^{-1} and 1419 Tg yr^{-1} , respectively, compared to 681 Tg yr^{-1} and 677 Tg yr^{-1} in the present study. The discrepancy may partly reflect differences in the dust emission scheme, interannual variability, correlation length and the data used for the data assimilation. Our configuration includes MSG-based Saharan dust sources, which yield up to +20% higher annual Saharan dust emissions (Heinold et al., 2016). This would be expected to increase rather than reduce dust loadings, suggesting that the discrepancy more likely stems from other sources, e.g. interannual differences (2006 vs 2024/2025) (Aryal and Evans, 2021). Furthermore, in our study we imposed the correlation length for the perturbation uniformly, whereas over the Sahara region Tsikerdekis et al. (2023) imposed no correlation length. Additionally, they excluded POLDER $\text{AE}_{550-865}$ over the Sahara region from the data assimilation because the bias was too large.

Table 2. Comparison of aerosol emission estimates for CTL, DAS, and DAS using POLDER data from Tsikerdekis et al. (2023). All emissions are given in teragrams per year.

Category	CTL	DAS	POLDER DAS for 2006
DMS	51.2	55.9	–
SO ₂	103	143	198
SO ₄	4.0	5.5	–
BC	11.7	8.2	13.3
OC	126.9	167.5	153.6
SS	1036	1855	1850
SS _{AS}	89.6	63.2	–
SS _{CS}	946.3	1791.5	–
DU	681	677	1419
DU _{AI}	15.5	20.2	–
DU _{CI}	665.1	657.0	–

3.4 Changes in aerosol and cloud properties

In this section, we discuss the results of the single model run using the ensemble-mean emission estimates from DAS. The changes in aerosol emissions can alter the aerosol amount and mixture, in turn, modify the capacity of aerosols to serve as cloud condensation nuclei (CCN), and consequently affect cloud properties. Figure 9 shows spatial distributions of AOD_{550} (a), water AOD_{550} (b), CCN at 0.3% supersaturation at the surface (c), in-cloud cloud droplet number concentration at cloud-top (CDNC) (d), liquid water path (LWP) (e), ice water path (IWP) (f), and cloud fraction (CF) (g), for DAS, CTL and their differences.



DAS yields a higher global mean AOD₅₅₀ than CTL (0.13 vs 0.11), with substantial regional differences. AOD₅₅₀ increases over biomass-burning regions in South America and equatorial Africa, anthropogenic combustion source regions in India and south-east China, the eastern Sahara, and over most ocean regions, likely associated with enhanced aerosol outflow from land and an overall increase in SS emissions. An exception is found over the southern Tropical Indian Ocean, where AOD₅₅₀ decreases, consistent with the reduction in DMS and fine-mode SS emissions in that region.

The overall increase in AOD₅₅₀ is associated with larger aerosol mass emissions and a higher fraction of soluble aerosols, which promotes stronger water uptake. Consequently, water AOD₅₅₀ increases from 0.06 in CTL to 0.08 in DAS, with a similar spatial pattern (Fig. 9b).

Compared to CTL, DAS yields a higher global mean CCN at surface (181.83cm^{-3} vs 166.93cm^{-3}), with pronounced regional differences (Fig. 9c). CCN at the surface increases in almost all regions, with particularly strong increases over biomass-burning and anthropogenic source regions such as North America, South America, equatorial Africa, South Africa, India, and East Asia, consistent with the AOD₅₅₀ increases shown in Fig. 9a. A slight CCN increase is also found at higher latitudes, including the Arctic and Antarctic regions, while a small decrease is apparent over remote ocean areas, particularly over South of TPO, consistent with changes in DMS emissions (Fig. 7) and reduced fine-mode SS emissions (SS_{AS}: Fig. S13c).

Consequently, the global mean CDNC is higher in DAS than in CTL (91.58cm^{-3} vs 88.79cm^{-3}). Overall, the spatial pattern of CDNC change follows that of surface CCN but less so with that of AOD₅₅₀ (Fig. 9d). This is consistent with the observational evidence that surface CCN is a better proxy for cloud-base CCN than AOD (Jia et al., 2026b; Gryspeerdt et al., 2017). However, an exception occurs over eastern China and India, where CCN increases throughout the atmospheric column (Fig. S14) but CDNC decreases. This apparent contradiction is related to the mismatch in temporal sampling between CCN and CDNC when comparing annual mean: CCN is sampled at all time steps, whereas CDNC is sampled only when liquid clouds are present. The seasonal plots (similar to Fig. 9) illustrate that, during DJF and MAM when liquid clouds are scarce over eastern China and India, CCN is markedly enhanced in DAS (Fig. S15), but CDNC differs little from CTL (Fig. S16). During JJA and SON when liquid clouds dominate (Fig. S17), CCN shows decrease or little variation.

According to cloud adjustment theory (Albrecht, 1989), changes in CDNC subsequently lead to changes in cloud water and cover. LWP changes broadly follow the pattern of CDNC changes (Fig. 9e), where the global mean is changed by -0.78% . Changes in aerosol emissions may also affect ice-nucleating particles (INP) and hence IWP, analogous to the effect on LWP (Lohmann and Feichter, 2005). IWP (Fig. 9f) decreases over the Amazon and equatorial Africa, consistent with the reduction in BC emissions in those regions. In ECHAM-HAM, heterogeneous nucleation in mixed-phase clouds is initiated either by contact nucleation by mineral dust or by immersion nucleation by black carbon and mineral dust (Lohmann and Neubauer, 2018). Changes in BC and DU emissions therefore lead to changes in INP concentrations and subsequently in IWP.

CF changes by a small amount at the global scale (by 0.002%) and is spatially noisy (Fig. 9g), indicating a weaker response compared to CDNC, LWP and IWP. In ECHAM-HAM, CF is parameterized through a critical relative humidity scheme (Quaas, 2012; Sundqvist et al., 1989) rather than being directly linked to cloud condensates, which limits the direct influence of emission changes on CF (Gryspeerdt et al., 2020). Aerosols can affect relative humidity, but only through cloud droplet activation and subsequent water vapor uptake. However, regional-scale differences might be more important. Visible changes

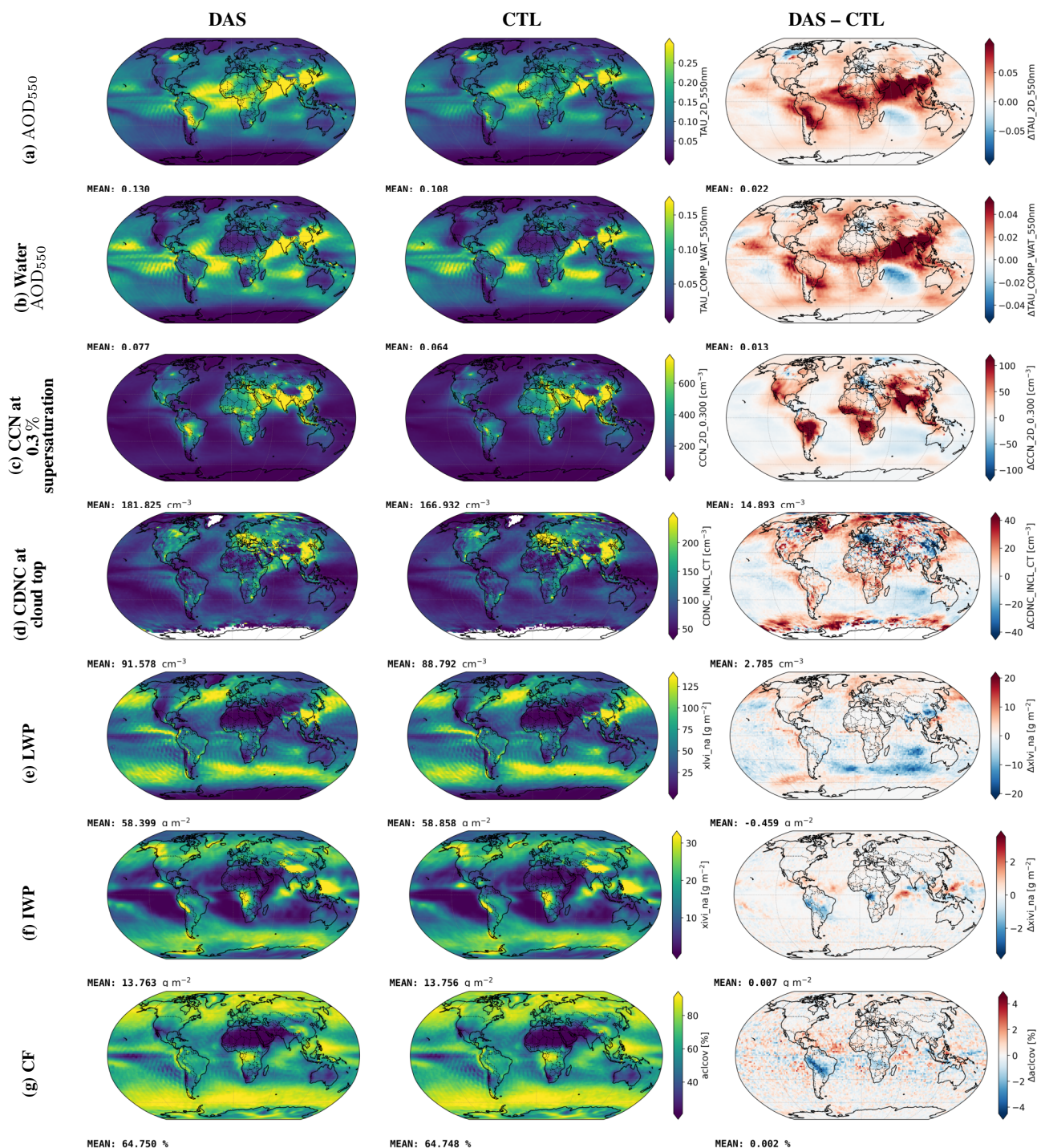


Figure 9. Time-averaged model data related to aerosols and clouds from DAS, CTL, and their difference DAS-CTL.



in CF are found over South America and equatorial Africa, closely following the IWP pattern, likely related to changes in the humidity field and cloud lifetime through ice formation.

3.5 Impacts on aerosol radiative forcing

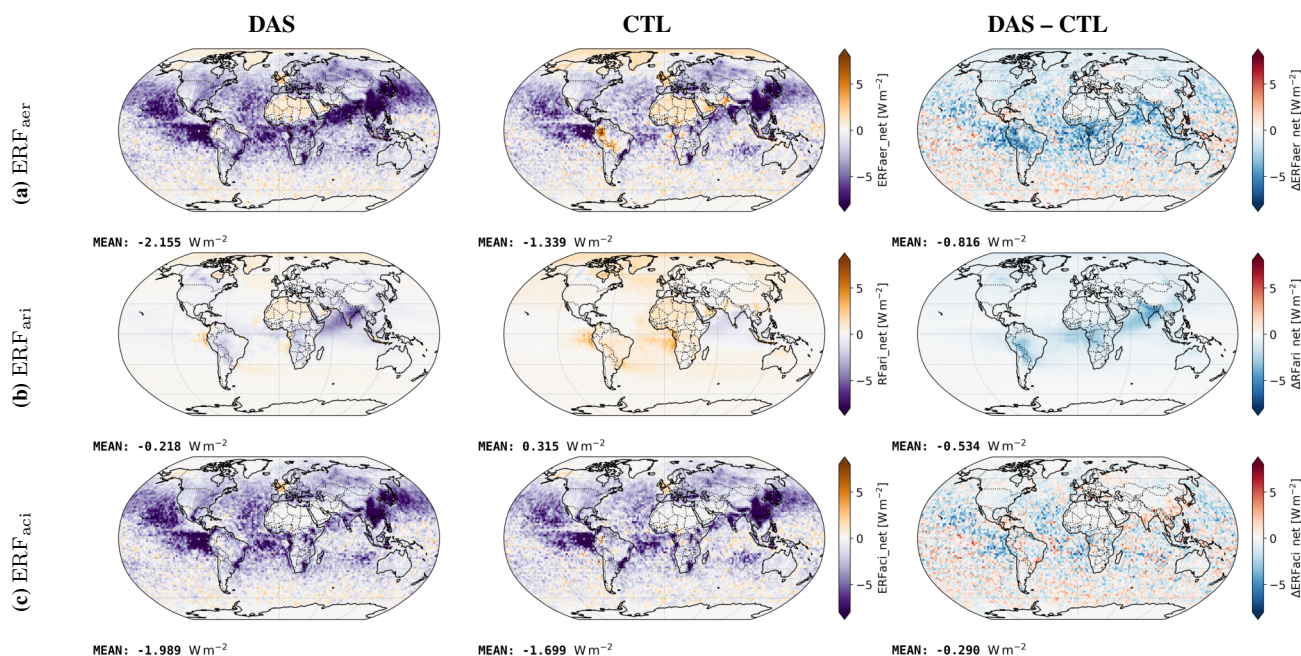


Figure 10. ERF_{aer} (a), ERF_{ari} (b), and ERF_{aci} (c) for DAS, CTL, and their difference DAS–CTL.

In this section, we assess changes in aerosol effective radiative forcing (ERF) resulting from the ensemble-mean emission adjustments of DAS, as applied in the single model run described in Sect. 3.4 (for the CTL single run default emissions were used). For DAS-PI experiment, aerosol corrections are applied only for natural species (DU, SS, and DMS), while both natural and anthropogenic emissions are adjusted in the DAS-PD experiment. The formulas used to compute ERF are provided in Appendix B. Figure 10 shows the total net ERF_{aer} (a), the net ERF due to aerosol–radiation interactions (ERF_{ari} , panel b), and the net ERF due to aerosol–cloud interactions (ERF_{aci} , panel c), each for CTL (left), DAS (center), and their difference DAS–CTL (right).

DAS produces a more negative ERF_{aer} (-2.16 W m^{-2}) compared to CTL (-1.34 W m^{-2}), indicating substantially stronger cooling. This shift is driven primarily by ERF_{ari} , which changes from positive ($+0.32 \text{ W m}^{-2}$) in CTL to negative (-0.22 W m^{-2}) in DAS, and to a lesser extent by ERF_{aci} , which becomes more negative in DAS (-1.99 W m^{-2}) than in CTL (-1.70 W m^{-2}). The ERF_{aer} (and particularly ERF_{aci}) estimated from DAS lies at the lower end of the IPCC assessed range (IPCC, 2023, Chapter 7). However, a direct comparison with the IPCC assessed range should be interpreted with caution. In our setup, fire emissions are treated as anthropogenic, meaning the PD and PI experiments use different fire emission



inventories: GFAS for PD (scaled by 3.4) and CMIP6 biomass-burning for PI. Additionally, the model may exhibit an overly high sensitivity of cloud droplets to CCN perturbations and excessively strong updraft velocities (Virtanen et al., 2025), which could amplify ERF_{aci} .

470 Since ERF is closely related to the differences in PD–PI (Δ) in aerosol and cloud properties, we also show these differences in Fig. S18. The shift in ERF_{ari} can be understood through ΔAOD_{550} and $\Delta AAOD_{440}$. The ΔAOD_{550} is larger in DAS than in CTL (0.05 vs 0.03), while $\Delta AAOD_{440}$ is smaller (0.004 vs 0.005), indicating a shift toward a more scattering and less absorbing PD atmosphere. The enhanced cooling is strongest over biomass-burning regions in Canada, South America, and equatorial Africa, and over India, where reduced BC emissions (Fig. 7) lower $AAOD_{440}$ and enhanced OC and SO_4 increase
 475 scattering AOD_{550} . Over the ocean, particularly in biomass-burning outflow regions, the replacement of absorbing BC by more reflective OC and sulfate produces a stronger cooling effect, consistent with the simultaneous decrease in $AAOD_{440}$ and increase in AOD_{550} seen in Fig. S18. ERF_{aci} is more negative in DAS compared to CTL (Fig. 10c), though the spatial pattern appears more noisy due to the CF variability. The distribution of ERF_{aci} changes broadly agrees with $\Delta CDNC$ (DAS: 26.39 cm^{-3} , CTL: 23.22 cm^{-3}) and ΔLWP (DAS: 5.30 g m^{-2} , CTL: 4.60 g m^{-2}) shown in Fig. S18, through the interactions
 480 of aerosols and liquid clouds. Although ice formation can influence IWP and ice cloud fraction (included in CF), its impact is limited to the longwave ERF_{aci} , which is negligible compared to the shortwave forcing (Fig. S19).

4 Discussion and conclusion

This study presents global aerosol emission estimates for the period 2024-08 to 2025-07, derived by assimilating SPEXone multi-angle polarimetric observations from the NASA PACE mission into the ECHAM6.3-HAM2.3 global aerosol–climate
 485 model using the LETKS data assimilation framework. The assimilated observations constrain aerosol amount, size, and absorption through AOD_{550} , $AE_{440-670}$, and SSA_{440} , respectively. The resulting emission estimates improve model agreement not only with the assimilated SPEXone observations but also with independent observations from AERONET and MODIS.

The data assimilation reduces BC emissions (-29.9%) while increasing SS ($+79.0\%$), DMS ($+9.2\%$), SO_2 and SO_4 ($+38.8\%$), and OC ($+32.0\%$); total DU remains essentially unchanged (-0.5%). Two dominant adjustment mechanisms are
 490 identified: over the ocean, $AE_{440-670}$ is corrected through increased coarse-mode SS emissions; over land, SSA_{440} is corrected by replacing strongly absorptive BC with OC and sulfate. Both mechanisms are consistent with the findings of Tsikerdekis et al. (2023) using POLDER observations for 2006. For fire emissions specifically, DAS implies global scaling factors of 2.09 for BC and 4.66 for OC relative to the GFAS inventory. The BC scaling is close to the POLDER-based estimate (1.90), while for OC the scaling is higher (POLDER: 2.93). This discrepancy may reflect differences in the prior emission fields used by the data
 495 assimilation, as well as differences in the study period with different contributions from BB. Additionally, GFAS fire emission estimates rely on MODIS fire radiative power retrievals, and potential sensor degradation between 2006 and 2024–2025 may further influence the inferred emission magnitudes and scaling factors.

The emission adjustments lead to higher AOD_{550} globally, driven by an increased abundance of soluble and scattering aerosols and stronger water uptake. Surface CCN and CDNC at cloud-top both increase globally but with pronounced regional



500 differences. A case study of eastern China and India demonstrates that an annual increase in CCN alone is not sufficient to increase annual mean CDNC, and that factors such as the seasonal timing of emission changes also need to be considered.

The updated emission estimates lead to a substantially more negative ERF_{aer} in DAS (-2.16 W m^{-2}) compared to CTL (-1.34 W m^{-2}). This is driven primarily by a shift in ERF_{ari} from $+0.32 \text{ W m}^{-2}$ in CTL to -0.22 W m^{-2} in DAS, and by a more negative ERF_{aci} in DAS (-1.99 W m^{-2}) compared to CTL (-1.70 W m^{-2}). DAS exhibits stronger cooling compared
 505 to CTL which can be explained by the replacement of absorbing BC by more soluble and reflective aerosols. These changes lead to increased atmospheric scattering and increased cloud droplet number. Comparing these estimates with the PPE-based ensemble of Bhatti et al. (2026) suggests that the DAS cooling may represent an extreme case.

Two systematic limitations are identified when comparing DAS against SPEXone. First, SSA_{440} is overestimated over remote ocean regions outside of land-to-ocean outflow areas, reflecting an inherent model bias that cannot be corrected through
 510 emission adjustments alone. This may suggest either an unrepresented source of absorbing aerosols over the ocean in the model, or an underestimated lifetime of land-sourced absorbing aerosols. However, it should be noted that a newer version of SPEXone retrievals (Fu et al., 2026) results in higher SSA over ocean which may reduce the bias. Second, AOD_{550} is overestimated in regions of high relative humidity, such as the Tropical Indian Ocean and East Asia. Following Tsikerdekis et al. (2023), these regions are characterized by high concentrations of soluble aerosols such as SS and sulfates, whose optical
 515 properties are sensitive to the RH profile. Model overestimation of RH in these regions may lead to incorrect SS and sulfate emission estimates. The non-linear nature of aerosol water uptake makes this particularly challenging for the data assimilation system to correct.

We can address parametric biases, for instance by running the data assimilation with a PPE-constrained version of ECHAM-HAM (Bhatti et al., 2026). That would be an important step toward more physically consistent emission estimates. Given the
 520 importance of aerosol emissions for radiative forcing and the broader impact of aerosols on global climate, the ultimate goal is to combine a model with reduced structural uncertainty with spatially and temporally resolved emission estimates, which could substantially reduce uncertainties in aerosol forcing.

Code and data availability. The ECHAM-HAM version that was used in this study can be found in the following repository: https://redmine.hammoz.ethz.ch/projects/hammoz/repository/echam/show/echam6-hammoz/branches/uni_amsterdam_vrije/WC20220422 (Tsikerdekis et al.,
 525 2022a). This repository can be accessed after registration at <https://redmine.hammoz.ethz.ch/projects/hammoz> (HAMMOZ, 2023). The SPEXone observations can be found at <https://public.spider.surfsara.nl/project/spexone/RemoTAP-SPEXone-GRIDDED/> (last access: August 2026; Fu et al., 2025). The MODIS Aerosol Cloud Water Vapor Ozone Daily L3 Global product (MYD08_D3, Collection 6.1) is available from the NASA LAADS Distributed Active Archive Center (DAAC): https://ladsweb.modaps.eosdis.nasa.gov/archive/allData/61/MYD08_D3/ (last access: August 2026; MODIS Atmosphere Science Team, 2017). The AERONET (<https://aeronet.gsfc.nasa.gov/>;
 530 AERONET, 2023) data are freely available.



Appendix A: Observation calculations

AAOD₄₄₀ at wavelength λ_1 for SPEXone is computed as

$$\text{AAOD}_{\lambda_1} = \text{AOD}_{\lambda_1} (1 - \text{SSA}_{\lambda_1}) \quad (\text{A1})$$

The AE_{440–670} between wavelengths $\lambda_1 < \lambda_2$ is calculated as

$$535 \quad \text{AE}_{\lambda_1-\lambda_2} = - \frac{\log\left(\frac{\text{AOD}_{\lambda_1}}{\text{AOD}_{\lambda_2}}\right)}{\log\left(\frac{\lambda_1}{\lambda_2}\right)}, \quad (\text{A2})$$

where this formula is applied to both AERONET and MODIS retrievals.

For AERONET, we use

$$\text{AOD}_{\lambda} = \text{AOD}_{\lambda_2} (\lambda/\lambda_2)^{-\text{AE}_{\lambda_1-\lambda_2}}, \quad (\text{A3})$$

with $\lambda_1 \leq \lambda \leq \lambda_2$ and $\lambda_1 \neq \lambda_2$. For AERONET, to compute AOD₅₅₀, we used AOD at 440 nm and 675 nm.

540 Appendix B: Radiative forcing calculations

The calculation follows established conventions (Ghan, 2013). All fluxes are evaluated at the top of the atmosphere (TOA) and follow the convention that net downward radiation is positive. The net radiative flux F is computed from the downwelling radiative flux $r^\downarrow$ and the upwelling shortwave and longwave fluxes $r_{\text{sw}}^\uparrow$ and $r_{\text{lw}}^\uparrow$, respectively:

$$F = r^\downarrow - r_{\text{sw}}^\uparrow - r_{\text{lw}}^\uparrow. \quad (\text{B1})$$

545 We compute F for the following conditions: all = all-sky, cs = clear-sky, noa = no-aerosol, and csnoa = clear-sky no-aerosol. To evaluate anthropogenic forcing, two experiments are conducted: PI = pre-industrial and PD = present-day. Here, PI refers to anthropogenic aerosol emissions representative of 1850. Using the definitions above, the three plotted quantities are computed as:

$$\text{ERF}_{\text{ari}} = \left[(F_{\text{PD}}^{\text{all}} - F_{\text{PD}}^{\text{noa}}) - (F_{\text{PI}}^{\text{all}} - F_{\text{PI}}^{\text{noa}}) \right], \quad (\text{B2})$$

$$550 \quad \text{ERF}_{\text{aci}} = \left[(F_{\text{PD}}^{\text{noa}} - F_{\text{PD}}^{\text{cs,noa}}) - (F_{\text{PI}}^{\text{noa}} - F_{\text{PI}}^{\text{cs,noa}}) \right], \quad (\text{B3})$$

$$\text{ERF}_{\text{aer}} = F_{\text{PD}}^{\text{all}} - F_{\text{PI}}^{\text{all}}. \quad (\text{B4})$$

Note that for the forcing calculation, $r^\downarrow$ is not required.



Appendix C: Observation uncertainties

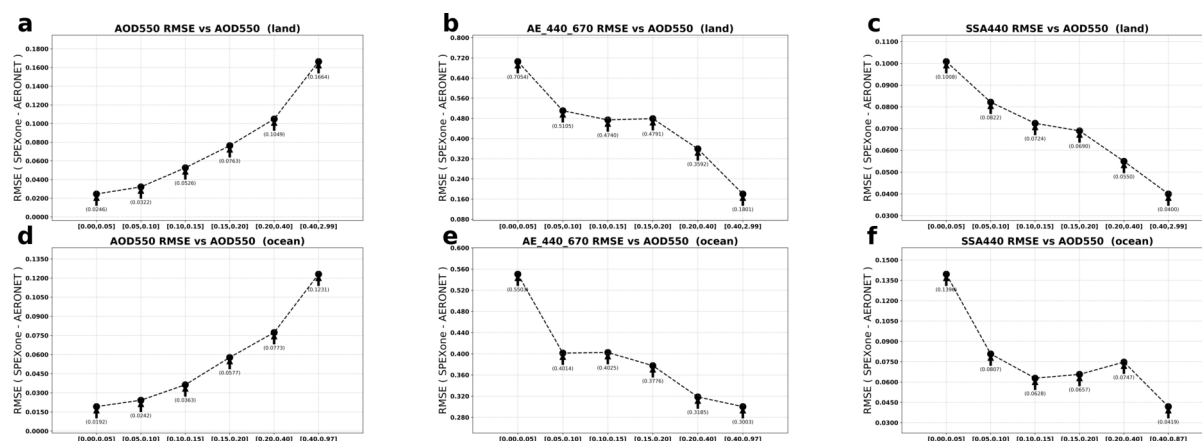


Figure C1. SPeXone validation against AERONET at pixel level (5 km) for AOD₅₅₀, AE_{440–670}, and SSA₄₄₀. The uncertainties are binned and separated for land and ocean.

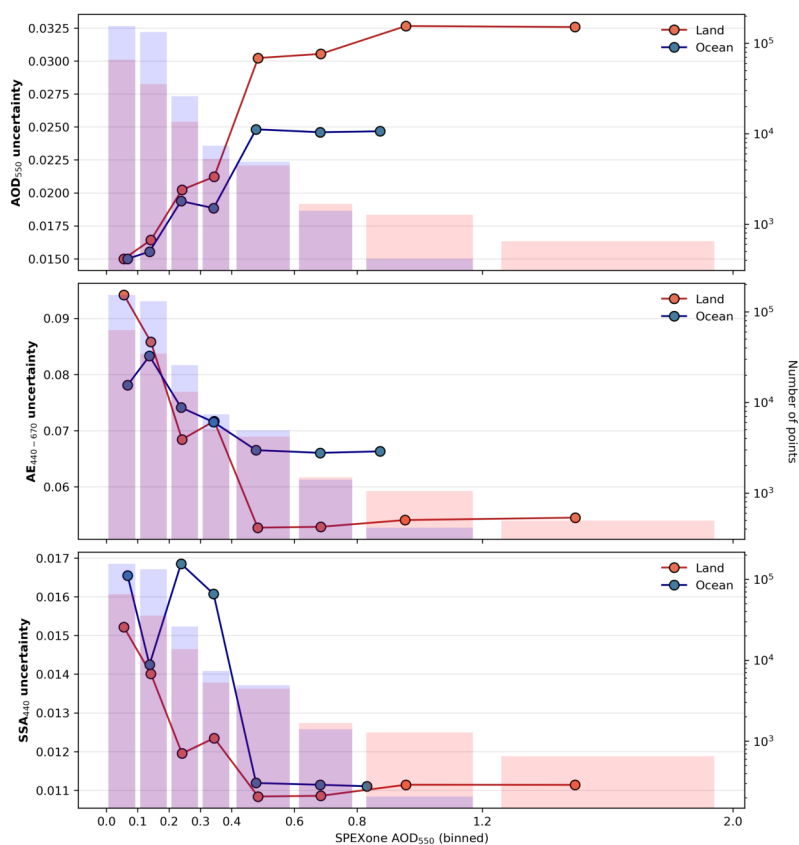


Figure C2. SPEXone absolute uncertainties on a $1^\circ \times 1^\circ$ grid for AOD₅₅₀, AE₄₄₀₋₆₇₀, and SSA₄₄₀, binned by AOD₅₅₀. Dots show mean uncertainties within each bin plotted against the mean AOD₅₅₀ in the bin; blue dots indicate ocean and red dots land. Bars show the number of points (logarithmic scale) used for aggregation. The uncertainties are based on the parameterization in Fig. C1, divided by the square root of the number of points used for aggregation onto the $1^\circ \times 1^\circ$ grid.



Appendix D: Region definitions

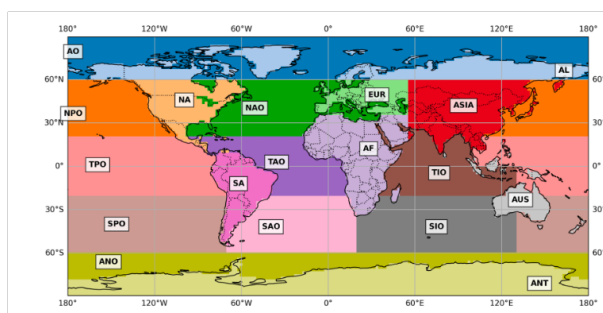


Figure D1. Map of the regions used for the analysis from the work of Jia et al. (2021). The regions are: AO (Arctic Ocean), AL (Arctic Land), NPO (North Pacific Ocean), NA (North America), NAO (North Atlantic Ocean), EUR (Europe), ASIA (Asia), TPO (Tropical Pacific Ocean), TAO (Tropical Atlantic Ocean), AF (Africa), TIO (Tropical Indian Ocean), SPO (South Pacific Ocean), SA (South America), SAO (South Atlantic Ocean), SIO (South Indian Ocean), AUS (Oceania), ANO (Antarctic Ocean), and ANT (Antarctica).

555 *Author contributions.* AA organized the experiments with the help of HJ and OH. AA prepared the experiments and performed the analysis. GF provided the SPEXone data and uncertainties. NS implemented much of the data assimilation code, with further contributions from AT. AK prepared the model input data. All co-authors contributed their expertise on the observations, model and data assimilation, specifically NS, AT and GF for the data assimilation setup.

560 *Competing interests.* At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics. The authors declare that they have no other conflict of interest.

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