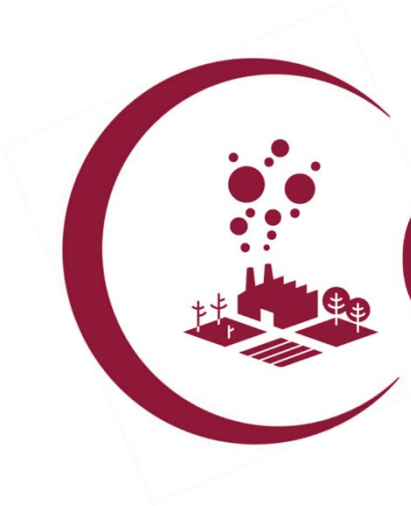

CONFESS

Consistent representation of temporal variations of boundary forcings in reanalyses and seasonal forecasts

Harmonized CAMS and CMIP6 datasets for aerosols

Tim Stockdale, Retish Senan, Roberto Bilbao

Co-ordinated by ECMWF

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Consistent representation of temporal variations of boundary forcings in reanalyses and seasonal forecasts

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1 Executive Summary

Two new HARMONIZED-CMIP6 aerosol datasets have been produced, representing different versions of a time-varying aerosol climatology driven by CMIP6-specified forcings, one based on raw model output and the other scaled to be consistent with usage in the latest version of the ECMWF Integrated Forecasting System (IFS) .

The data have been derived for the 1971-2019 period. Aerosol species which are dominated by natural sources are represented as a simple monthly mean climatology valid for the whole period. Aerosols which have substantial anthropogenic drivers are instead represented as a 9-year running mean climatology for each calendar month. The running mean is sampled at 5-year intervals to provide climatologies at specified epochs from 1975 to 2015. For use beyond 2015, extrapolation with constant values is recommended.

The first version of the dataset contains the aerosol mass loading of the 14 aerosol species as produced by the IFS-COMPO model configuration. Data are vertically resolved, and have been interpolated to 21 pressure levels from the surface to 1 hPa. The second version is based on exactly the same data, but has had certain scaling factors applied to ensure that the radiative impact of the aerosols in the current version of the IFS is acceptably realistic. The scalings are derived based on assessment of the mean model state in long model integrations. They have also been checked and confirmed as necessary in numerical weather prediction tests, which are very sensitive to initial temperature tendency biases. These scalings are specific to the optical properties of the aerosols as used in IFS CY47r3, and other models may need different adjustments, which is why the unadjusted values are also provided.

The HARMONIZED-CMIP6 aerosol dataset has been compared with aerosol from EC-Earth3-AerChem CMIP6 simulations, considering both mass loadings and optical depths. The levels of similarities and differences are in line with expectations, based on comparison studies of different CMIP6 aerosol models.

This method of creating time-varying aerosol climatologies has general applicability, both for other models and for future IFS model versions, and can be used to create new datasets as new IFS model versions become available. Lessons learned and recommendations for improvements in details of the method are provided.

The datasets are in netCDF format and are available from the CONFESS ftp site. Please contact the CONFESS Coordinator to confirm access details.

2 Introduction

2.1 Background

Tropospheric aerosols have a significant radiative impact on the atmosphere. However, aerosols are highly complex and the details of their global distribution and time history are at best only roughly known. When represented in models further simplifications are required (a limited number of species, approximate treatment of size distributions, simplifications in aerosol properties etc). The choices on how to represent aerosol within a given Earth System Model (ESM) are model-dependent, and thus it is not straightforward to transfer aerosol values from one model or dataset to another.

In recognition of these issues, the specification of historical aerosol values for CMIP6 focussed on the specification of emissions of aerosols and precursor species via agreed datasets, that could be used by ESMs to drive their own aerosol models and create a time-varying representation of aerosol which was compatible with each model's representation of aerosol and radiative properties. However, recognizing the cumbersome nature of a full aerosol treatment, and the difficulty of controlled experiments examining the impact of aerosol changes when such an approach is taken, CMIP6 also recommended the use of a specific aerosol dataset for use in a certain sub-project. This dataset, MACv2-SP (Stevens et al., 2017), is a very simplified representation of anthropogenic aerosols (simple patterns of spatial distribution, and a single aerosol species for each region with specified optical properties) and its time evolution . This dataset was intended purely for use in sensitivity studies, although some modelling groups without an aerosol model simply took it as a full if rather inaccurate representation of anthropogenic aerosols.

The standard configuration of the IFS uses a fixed aerosol climatology which was derived from Copernicus Atmosphere Monitoring Service (CAMS) analyses, based on the work by Bozzo et al. (2020). The climatology was created by running IFS Cycle 40r2 at TL159L60 to derive the partition of aerosol optical depth (AOD) between aerosol species. The total AOD was fixed to match that in the CAMS Interim Reanalysis over the same period, by simple scaling of all aerosol species at all levels by the same factor, chosen for each grid point and each month to ensure a match. In Cy43r3 the IFS radiation code was changed, and now requires aerosol to be input in terms of mass, from which the radiation code will calculate the optical properties. The Cy40r2 AOD-based climatology was translated to aerosol masses that gave an equivalent AOD when processed by the new radiation scheme. We will refer to this as the OLD climatology.

The aim of the work described here is to create an updated aerosol climatology, which captures the time-variation of anthropogenic aerosols as seen by CMIP6, but is also compatible with (or if possible better than) the existing aerosol climatology used by the IFS in Numerical Weather Prediction (NWP). The original intention had been to use the MACv2-SP to provide a time-scaling of the existing aerosol climatology, to allow consistency between representations of past and present and to ensure that the aerosol values were consistent with their use by the IFS. However, as explained in Section 2.2.3 below, it became apparent that a better choice was to instead re-derive the IFS climatology using the latest version of the aerosol model and the latest updated versions of the CMIP6 emissions forcings.

2.2 Scope of this deliverable

2.2.1 Objectives of this deliverable

The objectives of this deliverable were to produce a time-varying climatology of aerosol which could be used in WP3 to investigate the impact of long term changes in tropospheric aerosol on seasonal forecasts. The aim was to produce a dataset that was consistent with the (fixed) climatology currently used in the IFS, which is known to perform relatively well in terms of both mean-state in long model runs and impact on NWP scores, but that was also consistent with the time-variation as specified and used by CMIP6. This time-variation not only allows a more realistic representation of aerosol, but enables the influence of aerosol changes over time to be represented, necessary for accurate calibration of seasonal forecasts. The wider objective is not just to deliver data for scientific investigation within the CONFESS project, but to create data that is of value for future use by C3S in the production of seasonal forecasts and reanalyses.

2.2.2 Work performed in this deliverable

The forecast suite used at ECMWF to run the chemistry and aerosol model over extended periods of time with continual reinitialization of the meteorological fields was assessed and adapted to increase both efficiency and throughput speed, to allow timely production covering large periods of time. Software was added to the suite to calculate monthly means of the 3-hourly full model level data for aerosols at the end of each calendar month, before the data was then deleted from disk. This was necessary because the full 3-hourly 137-level 14-species aerosol dataset covering multiple decades was too large to be reasonably archived to tape.

To allow overlapping production periods, the spin-up properties of the aerosol and chemistry were investigated in a set of preliminary runs, with particular attention to spin-up of the stratosphere.

Assessments were made of the impact of variations in methane on the aerosols, to determine how much care was needed to ensure an accurate representation of methane.

The emissions specification of the chemistry and aerosol model was reviewed, to decide which emissions processes should be specified as a climatological or fixed seasonal cycle, and which should be provided from time-varying datasets, taking account of data availability and impact.

A first calibration production using CAMS-originated emissions was made for 2003-2020.

Software was written to process the resulting monthly-mean model-level data into a form suitable for use as a time-varying climatology for the IFS. This involved interpolating to lower resolution (both horizontal and vertical), a conversion from GRIB to netCDF, a separation between time-varying and fixed seasonal cycle species, choosing an appropriate time-smoothing of the data, and a reduction of the time-variation to “epochs”. Various diagnostic and validation products were also coded to allow an internal assessment of the product quality and ensure appropriate decisions were made.

The resulting aerosol climatology was assessed in a preliminary set of seasonal forecast integrations, looking at the impact on the model climate. Various sensitivity runs were made to find an appropriate set of scaling adjustments needed to ensure an acceptable radiative impact of the aerosol on the model.

Anthropogenic emissions data were downloaded from the Community Emissions Data System (CEDS), to allow creation of an aerosol climatology covering a much longer period than was possible with CAMS emissions, and that would be suitably linked to CMIP6. Global fire emissions data were similarly downloaded from the Global Fire Emissions Database (GFED). The production suite was configured to use these data, and preliminary experiments were made to test the compatibility of results. Scripts were adapted to ensure that suitable methane concentrations were used in the initial conditions of each 2.5 year production segment.

A full dataset was made from overlapping 2.5 year segments covering the period 1971-2019. Data from these segments were processed to produce a time-varying climatology for the period.

A second version of the time-varying climatology was produced which included the scaling adjustments needed for appropriate use by the IFS.

Monthly mean AOD files were created from 3-hourly AOD data archived by the runs, to allow further assessment and comparison with EC-EARTH aerosol used in CMIP. Various assessments were made, with a selection of results being reported here.

2.2.3 Deviations and counter measures

The original intention for this Deliverable was to use a different method. Many models in CMIP6 made use of MACv2-SP, a simplified representation of the time variation of aerosols derived by fitting a simple set of gaussian functions to anthropogenic aerosols produced by the MPI chemistry model with specified forcings. The resulting dataset was never intended to be a realistic representation of aerosols, but was rather conceived of as a tool to be used for coordinated sensitivity studies. The original plan for this Deliverable was to take the time variation of MACv2-SP and apply it as a time-varying scaling to the existing much more detailed aerosol climatology used in the operational IFS. However, once work had started it became apparent that such an approach, while possible, had a number of significant drawbacks:

1. The existing IFS aerosol climatology, created using a significantly earlier version of the IFS chemistry model, is very outdated and in need of replacement.
2. The MACv2-SP approach allows a time-varying scaling to be derived for anthropogenic aerosols, but it cannot be applied to total aerosol mass loadings, since the natural aerosols are assumed not to change from their preindustrial state. To use MACv2-SP scaling, it must be decided how much of the existing IFS aerosol climatology is anthropogenic, and how much natural. Although this seems obvious for some species (mineral dust, sea-salt), for most others there was no way to estimate this from the data at hand.
3. The whole MACv2-SP approach is in any case highly simplified, treating all anthropogenic aerosols as co-varying without distinction amongst species, and with only a very few spatial degrees of freedom. For example, no plume covers the Middle East, despite this being a major area of high aerosol concentrations in recent decades.

Given these drawbacks, which meant that at best a very ad-hoc and already outdated dataset could be produced, it was decided to develop a new, more comprehensive approach. It was recognised that this would take more effort and necessitate a delay in the originally-planned date of the Deliverable, but given the timescale of the whole project this was judged to be manageable, and the benefits to be sufficiently worthwhile given the wider aims of the CONFESS project.

3 CAMS-FORCED – a 2003-20 climatology using CAMS emissions

3.1 Outline of method

To create a climatology of aerosol, we follow a similar approach to that of Bozzo et al. (2020), but with some important differences. We use a more recent aerosol model (the one included in the latest operational cycle, IFS Cy47r3) which has more aerosol species and is fully coupled to the tropospheric chemistry scheme. This necessitates running a much more comprehensive model than the previous aerosol scheme, which could be run in an aerosol-only configuration. To perform an initial test of the method and allow calibration, we created a prototype version using the latest versions of CAMS forcing datasets (see Section 4.1 for details), which are available only from 2003 onwards. We refer to this as the CAMS-FORCED climatology. Unlike Bozzo et al. 2020, we do not scale the aerosol mass concentrations to bring the AOD in the model simulation into line with analysed values of AOD. This is because we are not certain that such a calibration step is helpful. The latest model version produces total aerosol AODs which are much more in line with AERONET (https://aeronet.gsfc.nasa.gov) validation data. The CAMS analysis, perhaps because it is dominated by one type of satellite data, has AOD which is larger than the AERONET validation by about as much as the free-running model is smaller than AERONET, so adjusting AOD to match analysis may not result in being any closer to the truth. The previously used method of adjustment is moreover very uncertain – in Bozzo et al. 2020, all species at all levels were assumed to have the same relative error (diagnosed independently at each grid point), and were scaled in the same way. This led to artifacts in the stratosphere (imprinting of surface topographic features in the previously zonally uniform fields), and is hard to defend as a reasonable method of estimating relative aerosol bias for each species.

We produce a pseudo-reanalysis by specifying time-varying emissions needed to drive both the chemistry and aerosol models, and running the IFS for a series of 24-hour forecasts. At the end of each run, the resulting chemistry and aerosol fields are taken and provided as initial conditions for the next 24h forecast, while the meteorological initial conditions are taken anew from ERA5. This ensures that the meteorology (winds, precipitation, temperature) are always synoptically realistic, while the aerosol and chemistry fields can evolve freely over long periods of time.

The AER aerosol scheme used in the IFS is documented by Remy et al. (2019), used in its full chemistry coupling configuration, together with various subsequent changes. The aerosol types included are listed below in Table 1 (see Section 3.2).

3.1.1 Spin-up considerations

Early experimentation demonstrated that the tropospheric aerosol concentrations spin up from arbitrary initial conditions quite well within two months. In the stratosphere the spin up takes much longer, as might be expected. Our first experiments started from initial conditions which had relatively large amounts of aerosol in the stratosphere, which then reduced strongly over time. Although the model is not designed to simulate stratospheric aerosols, we at least want to avoid unrealistically large aerosol concentrations. We thus used a 2.5 year spin up run to enable stratospheric aerosol concentrations to reduce to lower, more realistic values. This partially-spun up state from mid-2005 was then used as the standard initial condition for all of our production runs.

In order to allow production of a multi-decadal dataset in a reasonable time, we ran the production of the aerosol data in a set of parallel streams. Each production run was 2.5 years long, with a 6 month individual spin-up starting from the previously mentioned standard initial condition. For example, data for 2000 and 2001 come from a run started on the 1st July 1999. The result is a six month spin-up period for the tropospheric aerosol (more than adequate), and an effective 3 year spin-up for the long-term mean state of the stratosphere. When we piece together the 2-year chunks to form the final dataset, there remains a very small discontinuity in the stratosphere once every 2 years. Since aerosol values are very low this is of no practical significance.

3.1.2 Definition of climatology

The full set of monthly mean model level mass loadings of aerosol are processed to form a climatology for use by the IFS or any other model.

Aerosols which are dominated by natural emissions processes were time averaged over the whole period to create a "fixed" monthly climatology. For these aerosols (three size bins of both salt and mineral dust, a total of 6 aerosol species) the variation from one year to the next is driven in our model system by changes in meteorology. This interannual variability can be large (especially for mineral dust) while longer term trends are assumed to be small, so averaging individual calendar months over the whole period is likely to be more appropriate than using sub-samples of the period which would suffer from residual sampling noise.

All other aerosols are processed by using a 9-year running mean applied separately to each calendar month and each grid point. Biomass burning is a major source of aerosols, and is a combination of human-induced and occasionally natural events. Given the existence of trends and changes in tropical biomass burning, and the possible non-stationarity over time of forest fires at high latitudes, it was decided to include burning-dominated species such as Organic Matter (OM) and Black Carbon (BC) in the time-varying part of the aerosol climatology. In mid-latitudes, it would not in any case be possible to separate which part of these species might be due to a hypothetical natural background, and which part was related to industrial-type sources.

The aerosol processes for which it is hardest to form a suitable climatology are episodic uncontrolled forest fires, for which emissions are very variable. It may be that there is just one major burn event in the whole period considered, and a single event can dominate the climatology of OM and BC in that area. These highly episodic emissions result in poorly sampled climatologies, even more so when a relatively short 9-year running mean is used. We considered methods to alleviate this, including smoothing the climatology between adjacent calendar months, either always or conditional on certain criteria such as the extent to which the mean and median differ, or the size of the standard deviation compared to the mean. Although such smoothing could be locally effective, it was found to introduce inaccuracies in other regions with higher aerosol burdens, and thus was not implemented. We also considered the more radical step of forming a climatology based on median rather than mean values. This would mean in effect totally removing the effect of major forest fires and a substantial reduction in overall biomass burning emissions, which is not desirable if we want to represent the mean impact of aerosol on climate. A major use of the time-varying aerosol climatology will be for re-forecasts used to calibrate seasonal forecasts, and that calibration is based on the mean of re-forecast values, not the median. Thus we choose to create our time-varying aerosol climatology to represent the **mean** level of aerosol mass loadings. The fact that a mean climate poorly represents the actual values of

aerosol loadings in areas with highly variable or episodic aerosol events is a good reason to pursue interactive aerosols, and cannot be solved by any sort of adjustment to the climatology used.

3.1.3 Interpolation

Our model runs are made at TL255L137 resolution, and so provide data on a reduced gaussian N128 grid (roughly 80km) and on 137 model levels in the vertical, extending up to 0.01 hPa. A climatology at this resolution would entail both large data files and excessive memory usage when used with the IFS. (On this latter point, some straightforward improvements are possible and are being made, but the problem does not go away without major restructuring in the distributed memory of the IFS code). For the first stage of processing we interpolate the data to a 3x3 degree grid, which reduces the data size by a factor of 11 and matches the grid of the existing IFS aerosol climatology. This low resolution is much more acceptable when representing a climatology which is already naturally smooth, than it would be for representing specific synoptic conditions. The only slight loss of precision is in regions of more complex orography, which are in any case not well resolved at T255. We further note that the purpose of the climatology is to calculate the radiative impact of aerosols within models, and such impact is most important at larger scales. The interpolation is done using the “interpw” software at ECMWF, a fast linear method which approximately conserves area-integrated values.

In the vertical, although a 137-level resolution is helpful in calculating aerosol transports, it is more than is needed to represent the climatology, especially considering that many of the 137 levels are in the stratosphere. We thus interpolate the 137-level data (as a function of pressure, assuming a seasonally varying climatological surface pressure field appropriate to the TL255 resolution model) in the vertical to 21 pressure levels, which correspond to the 19 levels used operationally at ECMWF around 1990, with two additional levels to extend the domain up to 1 hPa. When the 21-level data are read by the IFS, they are interpolated back to the resolution being used, typically 137 levels, so the use of 21 levels is simply a form of data compression. Tests show that not much information is lost. In some situations 33 levels allows slightly greater accuracy, but considering the overall uncertainties in aerosol any gains that could be obtained are considered negligible. The 137-level input data is interpolated with a cubic spline acting on the vertically integrated aerosol masses defined on half-pressure levels, and thus conserves total aerosol mass.

The aerosol climatology includes a 3-dimensional pressure level field for each month, giving the pressure coordinates of each level at each point. This can then be used within the IFS or any other model to interpolate the data in pressure and to map it back onto the model levels. It is important to use pressure rather than model level as the vertical coordinate because model levels can be very sensitive to orography - when used in a high resolution model (such as the TCo1279 9km resolution of the IFS), local orography can be very high, with mountains that may project above low-lying air pollution and smog. Pressure-level interpolation ensures that this situation is properly represented in the model using the climatology.

3.2 Comparison with OLD IFS climatology and choice of scaling parameters

The CAMS-FORCED 2003-20 climatology created with CAMS emissions and Cy47r3 was compared to the OLD operational climatology, which is based on that created by Bozzo et al (2020) using Cycle 40r2 and the CAMS Interim Re-analysis. The OLD climatology is known to give acceptably good results for

NWP, including short-term forecast temperature errors, suggesting that its radiative impact on the IFS is reasonably accurate.

Maps of vertically integrated aerosol mass for each aerosol species can be compared between the CAMS-FORCED and OLD climatologies. Spatial patterns are broadly similar, but there are also differences. For some species, notably mineral dust and sea-salt, the new climatology has a substantially different distribution of mass between the size bins.

Table 1: Aerosol species considered, showing also the total column burden in the CAMS-FORCED and OLD climatologies.

Aerosol type	OLD mg/m2	CAMS-FORCED mg/m2	CAMS-FORCED/OLD
Sea-salt 1 0.03-0.5 µm	1.19	4.02	338%
Sea-salt 2 0.5-5 µm	68.3	47.81	70%
Sea-salt 3 5-20 µm	82.86	15.75	19%
Mineral dust 1 0.03-0.55 µm	6.49	0.21	3%
Mineral dust 2 0.55-0.9 µm	19.3	2.10	11%
Mineral dust 3 0.9-20 µm	20.52	31.07	151%
Organic matter hydrophilic	4.0	6.50	162%
Organic matter hydrophobic	0.56	0.81	145%
Black carbon hydrophilic	0.36	0.37	101%
Black carbon hydrophobic	0.086	0.068	79%
Sulphates	3.21	2.85	89%
Nitrate fine		1.16	
Nitrate coarse		1.83	
Ammonium		0.36	

It is not possible to assess the net radiative effect of the changes just by looking at the changes in mass, given that there both increases and decreases and we also have new aerosol types that did not previously exist. A set of long-range forecasts was thus used to assess the impact of the CAMS-FORCED aerosol climatology on the model climate. These showed a substantial deterioration in 200 hPa winds in the tropical Atlantic region in JJA, with changes much larger than existing errors in the model climate. There were also large changes in 2-metre temperature (T2m), with many desert areas warming and central Africa cooling. These issues are not a complete surprise: the original aerosol climatology by Bozzo et al. (2020) had similar problems over central Africa, which were addressed by reducing aerosol concentrations in the region by a factor of 2 in the JJA period, to form the final version of the OLD climatology. We tested the impact of a similar reduction which only partially corrected the issues. We also noted (as seen in Table 1) that the size distribution of mineral dust has radically changed. This large change has been motivated by observations, but was made without corresponding adjustments in optical properties or checks on the radiative impact in the IFS. We made an ad hoc scaling of the dust aerosol to move it back towards the previous values, by scaling the amounts in the three dust bins by factors 20, 8 and 0.5. This resulted in dust amounts similar to the previous climatology in the desert regions. Once these two adjustments were made, impacts on T2m and especially 200 hPa wind were much reduced (Figure 1).

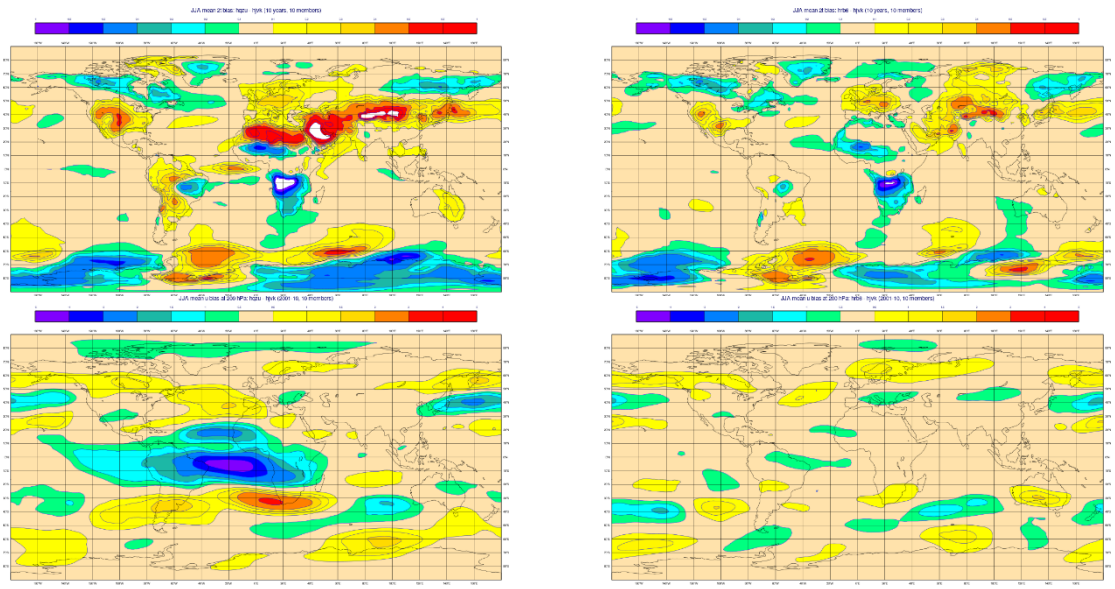

Figure 1: Impact of new aerosol climatology on the IFS model climate in JJA, as established from a 10-member ensemble run over the years 2001-10. Figures show the difference CAMS-FORCED – OLD for T2m (top) and U200 (bottom), for the raw version of CAMS-FORCED (left) and the scaled version (right).

This rescaled climatology was judged to be appropriate for use in the present cycle of the IFS (Cy47r3). Separate from the CONFESS project, this CAMS-FORCED-SCALED version has been tested in NWP experiments at ECMWF and found to perform comparably to the OLD climatology, while the unscaled CAMS-FORCED version led to a substantial deterioration in NWP scores. It is worth reiterating that the reason for the changes in the model giving less fine dust aerosol and more coarse dust were motivated by observations of dust size distribution, and noting that high levels of total aerosol over Africa are also observationally supported (though not necessarily speciation we use). The unscaled climatology is not necessarily wrong, but when used with the optical properties in 47r3 it gives a poor representation of the radiative impact. It is planned to revise these optical properties in a future cycle. We thus choose to present **two** versions of each of our aerosol climatologies, one being the raw output of the aerosol model, and one being a version scaled to suit the particular optical properties of Cycle 47r3 of the IFS.

4 HARMONIZED-CMIP6: a 1971-2019 climatology based on CMIP6 emissions

The CAMS-FORCED aerosol climatology can only be extended back to 2003 since that is the limit of the CAMS forcing datasets used to produce it. There is also only an indirect connection to CMIP6 specified data. In order to produce a time-varying aerosol climatology over a longer period, and that can be more directly tied to CMIP6, we create a new dataset based on CMIP6-related forcings. For practical reasons we restricted our production to data in the 1971-2019 period. It would be trivial to extend further back in time as far as the ERA5 meteorological data permit - the ERA5 back extension from 1959 has now been released, with 1940 to 1958 currently in production. We refer to the climatology based on the following forcings as HARMONIZED-CMIP6.

4.1 Choice of emissions datasets

4.1.1 Anthropogenic / industrial emissions

The Community Emissions Data System (CEDS, http://www.globalchange.umd.edu/ceds/) produces consistent estimates of global air emissions species over the industrial era (1750 - present, Hoesly et al., 2018). Historical gridded emissions data from CEDS was used in the Coupled Model Inter-comparison Project Phase 6 (CMIP6, Feng et al., 2020). Here we use an updated gridded emissions dataset based on the most recent version of CEDS (O'Rourke et al., 2021) which incorporates several updates and improvements (McDuffie et al., 2020) since the original CMIP6 version release, including updated emission estimates through 2019. CEDS provide emissions aggregated to nine sectors: agriculture, energy, industrial, transportation, residential–commercial–other, solvents, waste, international shipping, and aircraft, on a 0.5×0.5◦ grid.

Both CEDS and the CAMS anthropogenic emissions currently used in the IFS-COMPO configuration trace their origins to the EDGAR Emissions database (https://edgar.jrc.ec.europa.eu, Crippa et al. 2018). EDGAR emissions are used for country-wise scaling of the original CEDS inventory and further as a proxy data set for generation of spatial patterns in the gridded versions. In CAMS, CEDS-based trends are used to extrapolate EDGARv4.3.2 emissions beyond 2012 (see Granier et al., 2019).

Thus, the emissions data for the common period (2013-2019) are not identical but are related. It is also to be noted that the latest version of CEDS data used here has reduced emissions over China for recent years, removing erroneously high values which were used in the CMIP6 version of CEDS.

The CEDS emissions were processed for use in IFS-COMPO for the period 1950-2019. For compatibility with CAMS anthropogenic emissions, the same set of aerosols, aerosol precursors and reactive compounds (SO2, NOx, NH3, CO, volatile organic compounds (VOCs)) used in CAMS are specified.

CEDS does not provide information on diurnal variations in the emissions across sectors and we therefore use the CAMS diurnal cycle.

4.1.2 Biomass burning emissions

Historical global biomass burning emissions gridded dataset for CMIP6 (BB4CMIP6) were produced by the Vrije Universiteit Amsterdam (VUA, van Marle et al., 2017). The 0.25 x 0.25 dataset provides emission estimates back in time (to 1750) based on the satellite-derived Global Fire Emissions

Database version 4 with small fires (GFED4s) for 1997–2015 merged with several existing proxies (including sedimentary charcoal records, measurements of fire-emitted trace gases and black carbon stored in ice and firn, and visibility observations) and uses the average of six models from the Fire Model Intercomparison Project (FireMIP) protocol to estimate emissions when the available proxies had limited coverage.

For maintaining compatibility with the Global Fire Assimilation System (GFAS) biomass burning emissions that are currently used in the IFS-COMPO configuration, we use the same set of 19 emission species that is used from GFAS. While GFAS emissions are based on fire radiative power (FRP) retrievals from the MODIS instrument, GFED is based on burned area retrievals (Giglio et al, 2013). Nonetheless, the method used to retrieve GFAS values was calibrated against GFED data, so the two datasets are broadly comparable in terms of overall levels of emissions.

GFAS emissions use an injection height based on mean altitude of maximum injection and altitude of plume top information provided by a Plume Rise model (Remy et al, 2017) the latter calculated at very fine scale using data specific to each fire as well as the meteorological conditions at the time. No such data are available for GFED (nor could they be created, since the fire intensity is one of the inputs, which is measured by FRP but not by burnt area). We thus choose to provide all of the biomass burning emissions at ground level. Tests show that this results in approximately a 10% reduction in biomass burning aerosol loadings. It also gives a slightly different distribution (a bit more aerosol close by, a bit less aerosol at distance), but in terms of a climatology the impacts are small.

Note that IFS-COMPO, like other aerosol models, uses an emission factor to scale up the estimates of emissions given by the external datasets. The main justification for doing this is that the aerosol models only include a subset of the real aerosols – for example, Cy47r3 does not include secondary aerosols formed from VOCs, which are a significant source of aerosols from biomass burning. The value can also be used to tune the overall level of aerosol concentrations produced by the model. The emission factor used in the IFS for biomass-produced BC and OM is relatively large, at 3.4. This value was set for a much earlier version of the CAMS-COMPO system, one that used surface-only biomass burning emissions, and has not been revised since. Thus our specification of biomass burning emissions at the surface may be more consistent with the emissions factor than using injection heights would be, in terms of overall vertically integrated aerosol amounts.

Here, we use BB4CMIP6 emissions for the period up to 2014 and GFAS emissions from 2015-2019. The use of surface-only emission was used for all dates in the 1971-2019 period, regardless of the source of emissions.

4.1.3 Natural background emissions

Natural background emissions used include:

- Effusive volcanic SO2 emissions based on a satellite-derived climatology (Carn et al, 2017).
- Biogenic emissions of NH3, CO, NO and a selected set of VOCs based on MEGAN 2.1 modelling framework (Guenther et al., 2012)
- Dimethyl Sulphide (DMS) emissions from a TM5-based monthly climatology. (See Section 5 for details on TM5).

4.2 Consideration of chemistry

4.2.1 Role and evolution of methane

Methane (CH4) is a relatively long-lived gas, whose lifetime in the troposphere is usually regarded as around 10 years, although this is not fixed and depends on the concentrations of other chemicals, in particular OH, which provides a major sink of tropospheric CH4. Concentrations of OH are important for much atmospheric chemistry, and major changes in CH4 affect the loss rate and hence concentration of OH, and in turn the concentrations of other species. This in turn can affect nitrogen chemistry and the concentration of nitrate and ammonium aerosols in particular.

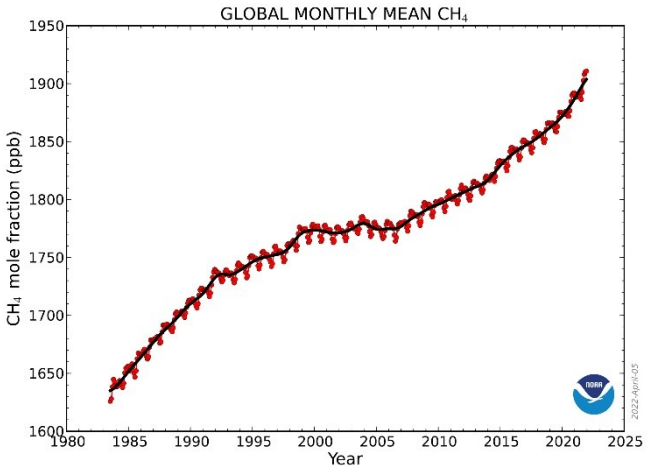

Figure 2: Time-evolution of globally-averaged methane from 1983 to present (NOAA Global Monitoring Laboratory, 2022).

Methane was relatively stable in the first decade of the 2000's, having had substantial increases through the 20th century. It had been hoped that methane levels were stabilising, but in the last decade levels have been rising again, and particularly steeply in the last few years (Figure 2). The causes of both the slowdown in concentration in the early 2000's and the acceleration in the last few years are not well understood, and may involve natural feedbacks which are not well (or not at all) represented in models. It is possible that a chemistry/aerosol model, run for extended periods of time, might develop unrealistic values of CH4 which might impact aerosol concentrations. However, examination of CH4 in the 1993-2000 runs showed it to be well behaved. If we follow our standard procedure of starting all runs from 2005 initial conditions, we would not expect to capture the long term evolution of methane, due to its long lifetime. To decide whether this was important, the sensitivity of aerosol to realistic variations in CH4 was assessed.

4.2.2 Impact of methane on nitrate and ammonium aerosols

A pair of experiments were run from July 2002, identical apart from the specification of methane. In one experiment, methane was specified at a fixed level of 1264 ppb, representing values for 1960 as specified in CMIP6, the other using methane levels of 2002. Small differences were seen in species such as sea-salt, but the only two aerosols that changed non-negligibly were ammonium and nitrate, particularly coarse nitrate, with reductions of 5-10% in regions of high concentrations. Although this is within the overall level of uncertainty of these species, it was judged appropriate to try to represent

the time-variation of CH4, in particular for years before 1990 when methane levels were meaningfully lower.

The experiments run in the 2003-2020 had time-evolving methane, which was observed to behave sensibly in this period. It was thus considered enough to scale the initial conditions for methane appropriate to the year in question, using mid-year values from the CMIP6 specification. This was implemented for our 1971-2019 runs with CMIP6 forcing. Unfortunately, CH4 was seen to evolve rapidly in the first days of these forecasts, not preserving the values given in the initial conditions. This was traced to a subroutine in the chemistry model which applies a strong surface relaxation of CH4 to a specified time-varying climatology. The time variation is based on the linear trend for the period 2003-2014. This was a period of modest growth in methane, so although reasonably accurate for, say, 1990-2020, it overestimates methane levels in earlier years. Thus our attempt to account for the time evolution of methane on the aerosols was only partially successful - we can expect to capture some of the trend but not all. Since the effect is in any case small, this is not a serious problem. We are in any case very unsure how well the chemistry scheme we are using would represent the impact on nitrates even with the correct methane time history.

4.3 Resulting time-evolving aerosol climatology

Figures 3 and 4 show the aerosol column burden (that is the vertically integrated mass per square metre) of the various species and how it has changed over the last forty years. The scale differs for each plot.

Organic Matter has a wide range of sources, and is the dominant aerosol type by mass, excluding mineral dust and sea salt which are not plotted here. Black Carbon emissions from fossil fuel combustion are now very small in developed countries (see January values for the Northern Hemisphere), but biomass burning is still a major source in summer, and may be increasing at high latitudes (Canada, Siberia). Biomass-burning in Africa remains the dominant global source according to these calculations, although they do rely on a large emission factor (3.4), so may overestimate BC in place of other aerosol types such as secondary aerosols. We also remind the reader that for practical use in the IFS we need to scale the JJA combustion related aerosols over central Africa by a factor of 0.5 – these plots are of the unscaled aerosol loadings.

Fine nitrate aerosol shows a striking increase around the world. This aerosol is formed from chemical processes, and is very sensitive to chemical balances in the atmosphere. Increases in North America and Europe in JJA are related in part to the reduction of sulphate aerosol. Elsewhere, growth in nitrate aerosol may be more due to growth in precursor emissions. Sulphate aerosol has had strong reductions during the last 40 years over North America and Europe, primarily as a consequence of emissions controls to counter acid rain. Sulphates have grown strongly in the Middle East, India, and China, although over China levels are now falling (see later).

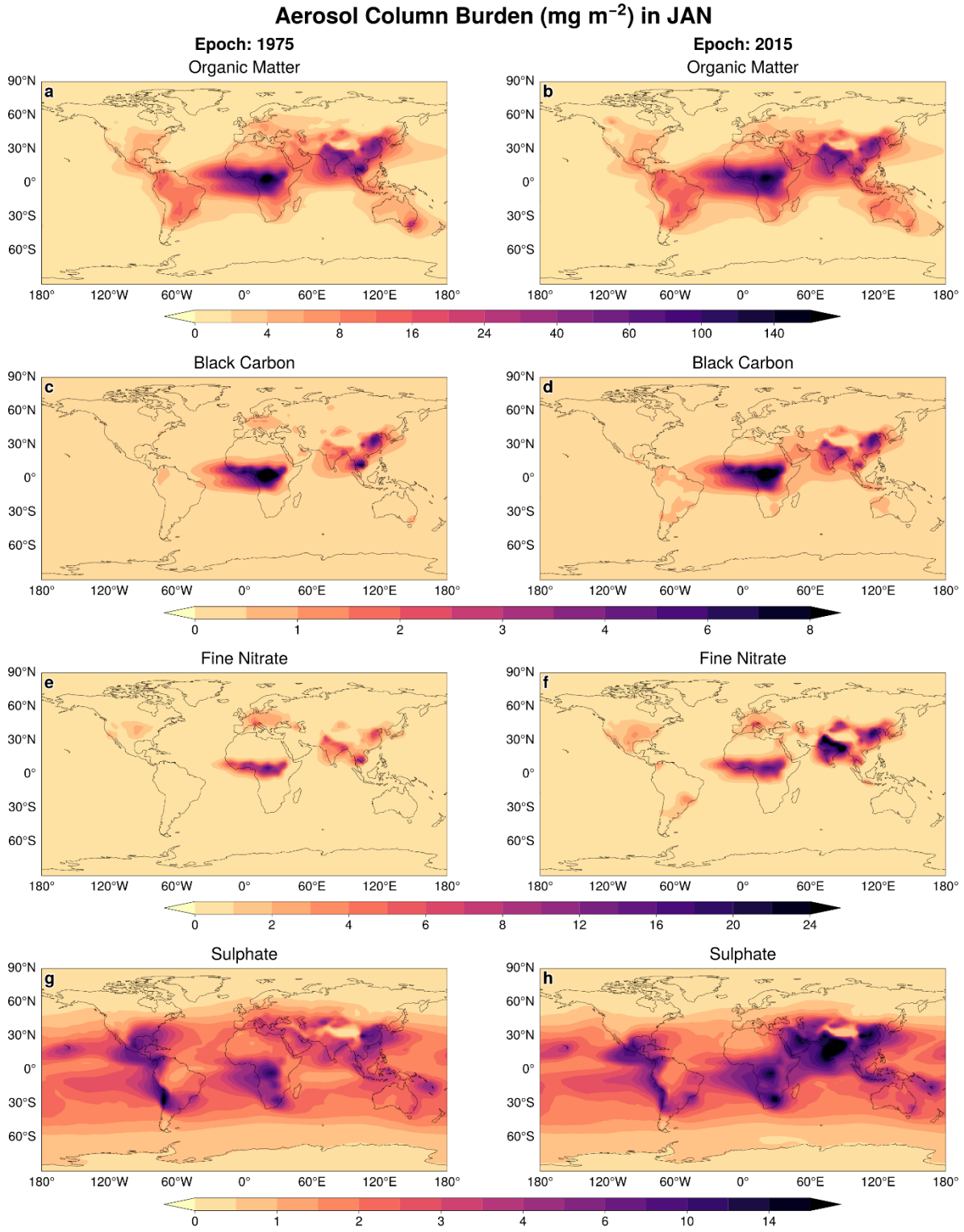

Figure 3: The time-varying vertically integrated January mass climatology shown for 1975 (left) and 2015 (right) for four epoch-varying species. OM and BC are the sum of hydrophilic and hydrophobic species.

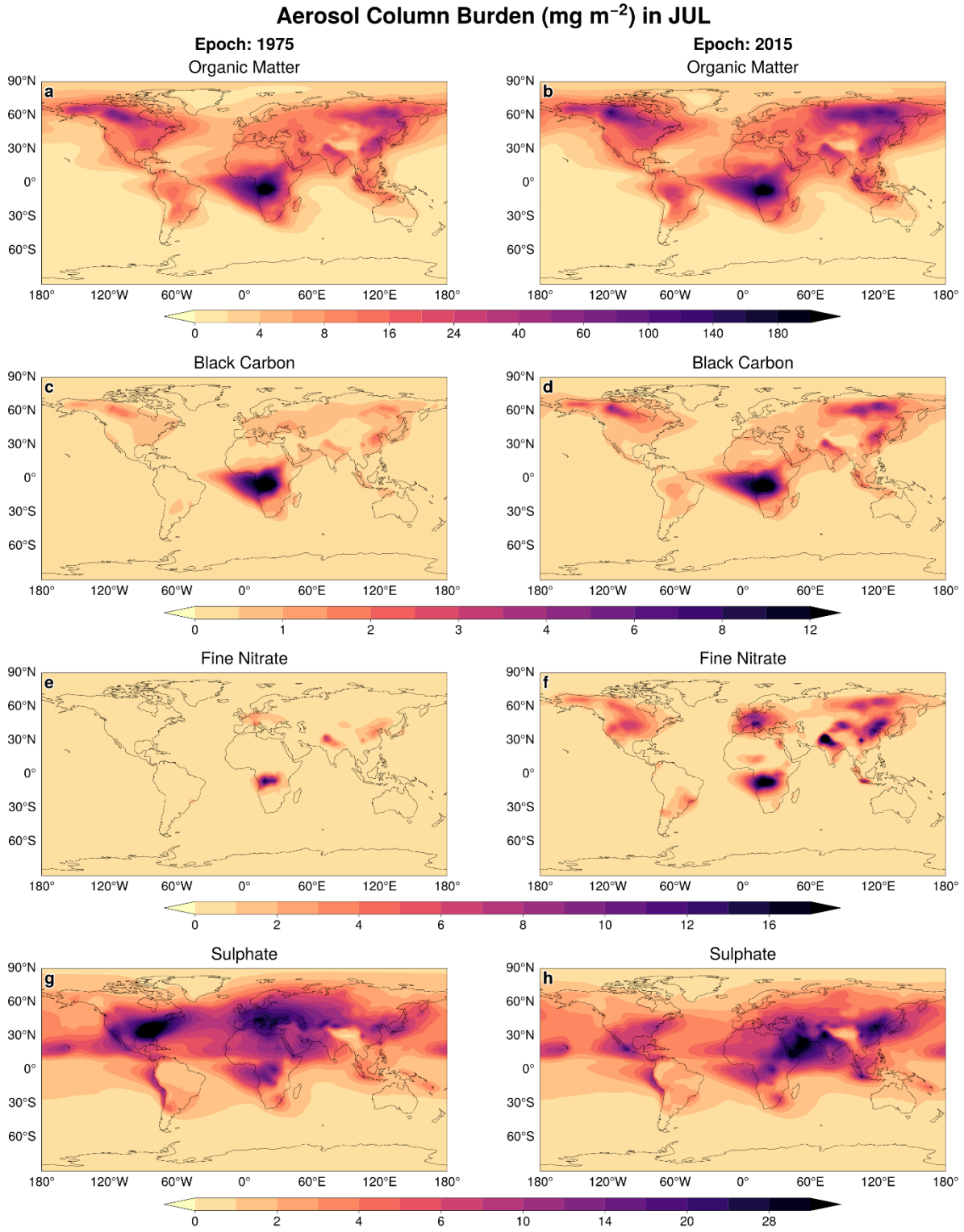

Figure 4: The time-varying vertically integrated July mass climatology shown for 1975 (left) and 2015 (right) for four epoch-varying species. OM and BC are the sum of hydrophilic and hydrophobic species.

We can see the evolution of the time-varying climatology more clearly with time-series plots, which we produce as area-averaged quantities for the selected regions shown in Figure 5.

On the following plots, the values of the HARMONIZED-CMIP6 climatology over central Africa (orange) in July are scaled by a factor of 0.5. This is partly to make the figures easier to read, but also reflects the fact that in the scaled version used by the IFS, we reduce the values over Africa to approximately these levels. In January, no scaling is applied in the plots, and no scaling of these species is applied in the HARMONIZED-CMIP6-SCALED dataset used by the IFS.

Figures 6 and 7 show that in the broadest terms OM and BC vary only modestly over the 40 year period considered, while fine nitrates typically show large relative increases, and sulphates have substantial variations depending on the region. Over China, sulphate and to some extent BC, is seen to have peaked, while concentrations over India are still growing. Europe and the US now have low levels of aerosol loadings.

For comparison, we include in these plots values from CAMS-FORCED, shown as dashed lines for the last three data points of the time-series. (The first data point at 2005 is a 7-year average from 2003-2009 compared to the 9-year average 2001-2009 plotted for HARMONIZED-CMIP6, which may give slight deviations, but the last two data points are for identical 9-year averaging periods). Levels of aerosol are broadly similar, as expected, but there are some notable differences. Over Africa, HARMONIZED-CMIP6 aerosol loadings are slightly below CAMS-FORCED for the peak July fire season (both shown scaled by 0.5), but slightly above in January. For Sulphate, there is generally good agreement, apart from the Middle East in July, and the failure of CAMS-FORCED to capture the downturn over China in the most recent years, as expected from the differences in emissions used. Fine nitrate shows relatively large differences in all regions, with HARMONIZED-CMIP6 having more fine nitrate in January, and less in July. Fine nitrate is known to be a very sensitive aerosol due to its chemistry, so this is not as surprising as might be first thought.

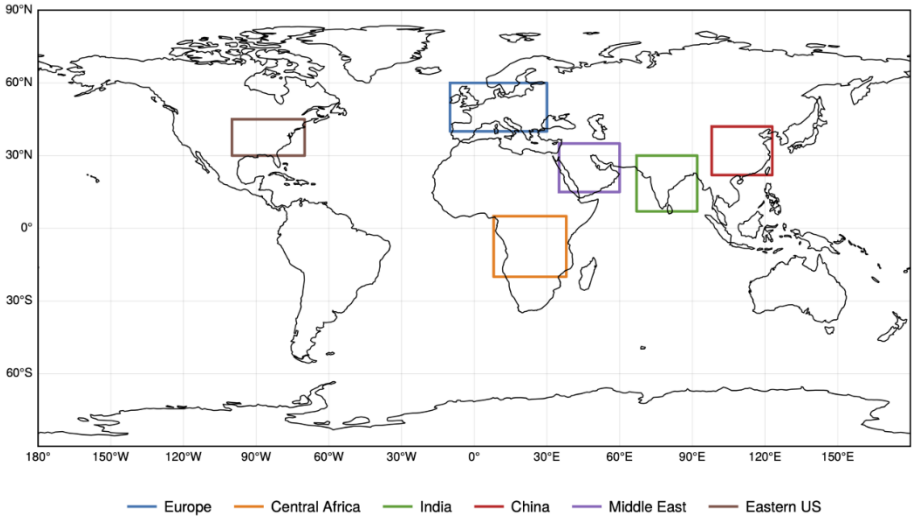

Figure 5: Map showing locations and boundaries of regions used for time-series plots

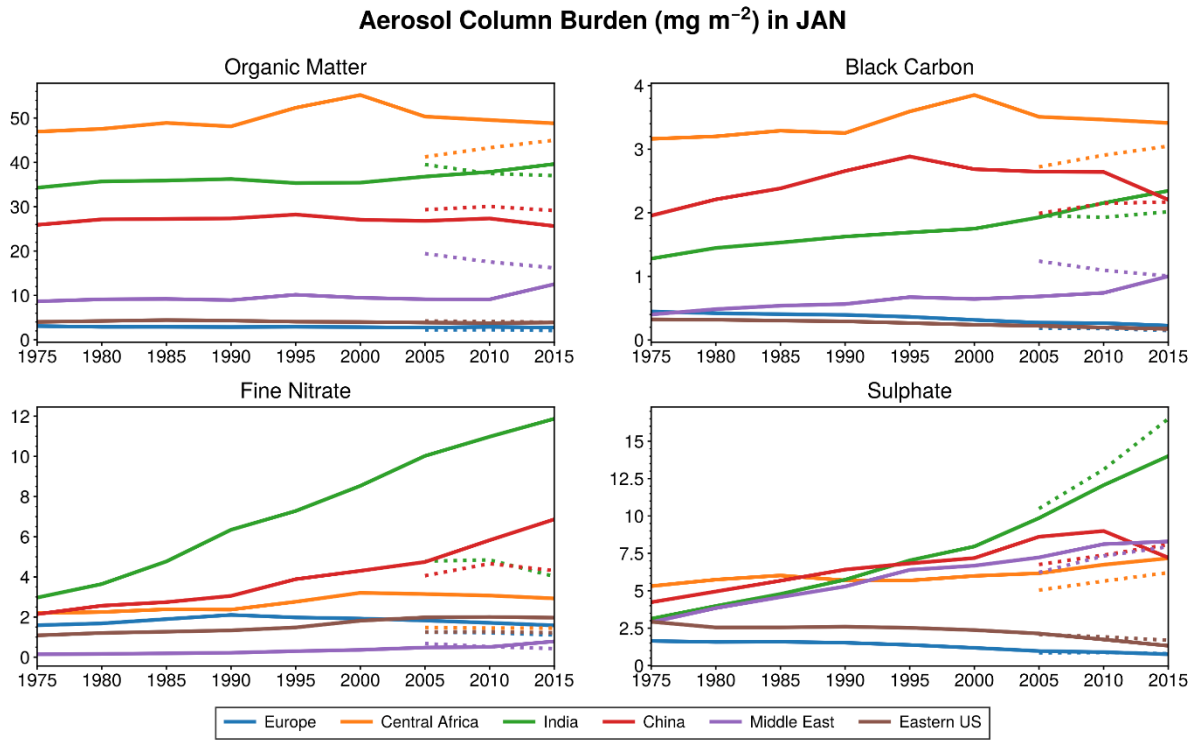

Figure 6: Time-variation of the HARMONIZED-CMIP6 climatology by epoch for January for four anthropogenically influenced aerosol species, area averaged over the stated regions. Dashed lines show for comparison the values from CAMS-FORCED.

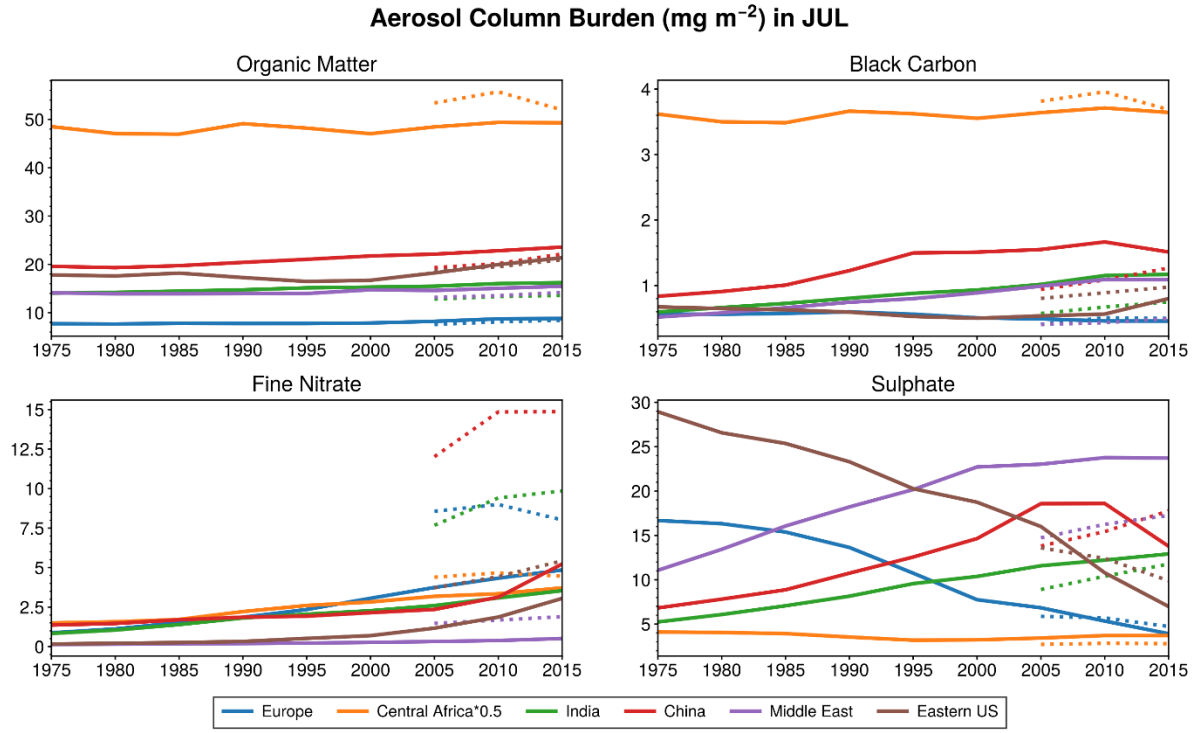

Figure 7: Time-variation of the HARMONIZED-CMIP6 climatology by epoch for July, area averaged for the stated regions. Values over Africa are multiplied by 0.5, reflecting the adjustments made in the scaled version of the datasets.

5 Comparisons with EC-Earth3-AerChem CMIP6 results

For comparison purposes we use a historical simulation (r4i1p1f1) run with the EC-Earth3-AerChem (described in detail in van Noije et al., 2021) climate model which simulates interactive aerosols and atmospheric chemistry, and has contributed to the Coupled Model Intercomparison Project Phase 6 (CMIP6). The atmospheric component used is the IFS from the European Centre for Medium-range Weather Forecasts (ECMWF), cycle cy36r4, with a T255 horizontal resolution (grid size of approximately 80 km) and 91 vertical levels. Aerosols and atmospheric chemistry are simulated with the Tracer Model version 5 (TM5) with a horizontal resolution of 3º × 2º (longitude × latitude) with 34 layers in the vertical direction. TM5 simulates tropospheric aerosols, namely sulphate, black carbon, primary and secondary organic aerosol, sea salt, and mineral dust. Additionally it simulates the total mass of ammonium, nitrate, and methane sulfonic acid (MSA). The anthropogenic and natural emissions of reactive gases and aerosols applied in TM5 are described in detail in section 2.4 of van Noije et al. (2021). Of relevance here are the historical emissions from anthropogenic activities which are taken from the Community Emissions Data System (CEDS; Hoesly et al., 2018). The simulation run extends to the end of 2014.

Data from this run are processed so as to be comparable with the CEDS climatology described in Section 4. In particular, monthly means for January and July have been smoothed with a 9-year running mean, and plotted from 1975 (the mean of 1971-1979) to 2010 (the mean of 2006-2014). We refer to the resulting dataset as EC-EARTH3, and the results are plotted in Figures 8 and 9.

Several points stand out. The aerosol mass burden of OM and BC over Africa, India and to some extent the Middle East is only about half as much in EC-EARTH3 as in HARMONIZED-CMIP6. This may be related to the large emission factor used in the IFS, and will be compensated where it matters most (over Africa in the peak biomass burning season) in the scaled version of HARMONIZED-CMIP6. Sulphate burdens agree quite well in January, with bigger differences in July, particularly for Europe: HARMONIZED-CMIP6 decreases roughly from 15 to 5 in 2010, whereas EC-EARTH3 decreases from 25 to 10 over the same period. Over China, EC-EARTH3 is missing the recent reduction in sulphate aerosol seen in HARMONIZED-CMIP6, probably because it used the original CMIP6 forcings, which had erroneously large emissions in these years.

It is again for fine nitrate that we find the largest discrepancies. January values are comparable for Europe and the Eastern US, being stable at around 2 mg/m2, but while July values show sharp upwards trends in these regions in HARMONIZED-CMIP6, in EC-EARTH3 they remain at very low levels. In fact, fine nitrate is at very low levels in all regions in July in EC-EARTH3, suggesting a different chemical balance. HARMONIZED-CMIP6 remains at intermediate levels, with CAMS-FORCED showing the largest fine nitrate levels of the three modelling systems. Fine nitrate is higher over China than India in EC-EARTH3, while the opposite is true in HARMONIZED-CMIP6 in January.

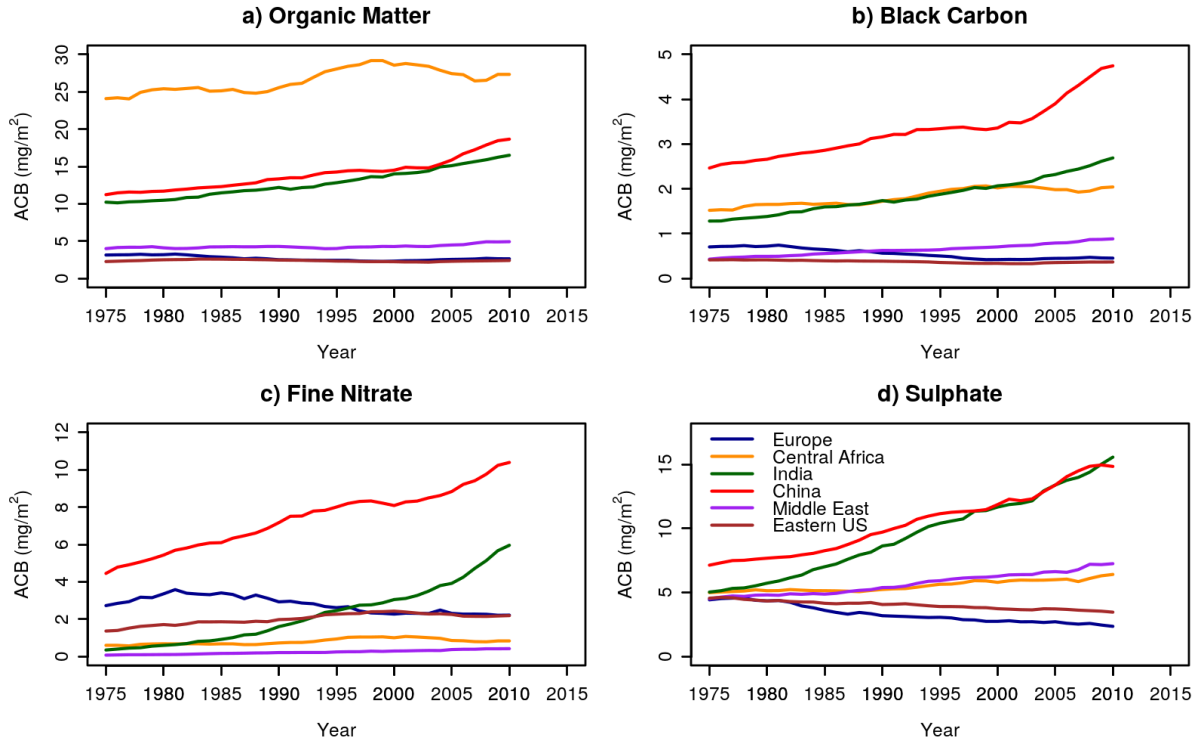

Figure 8: Time-variation of the January Aerosol Column Burden from EC-Earth3-AerChem, area averaged over the stated regions. For comparison with Figure 6.

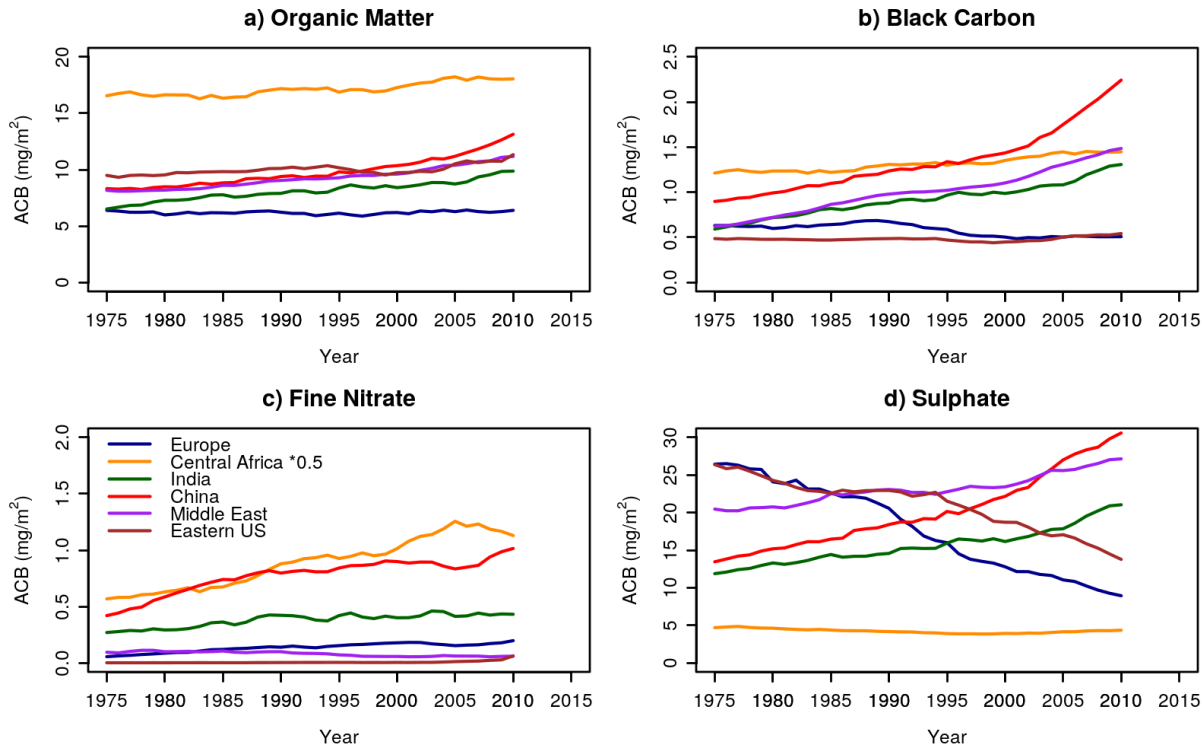

Figure 9: Time-variation of the July Aerosol Column Burden from EC-Earth3-AerChem, area averaged over the stated regions. For comparison with Figure 7.

To complement the comparison of aerosol mass burdens between the different analyses, Figures 10 and 11 show AOD per species. For these comparisons, monthly mean AOD fields for the IFS runs were created by taking 3-hourly values of AOD from the experiments used to calculate the aerosol masses.

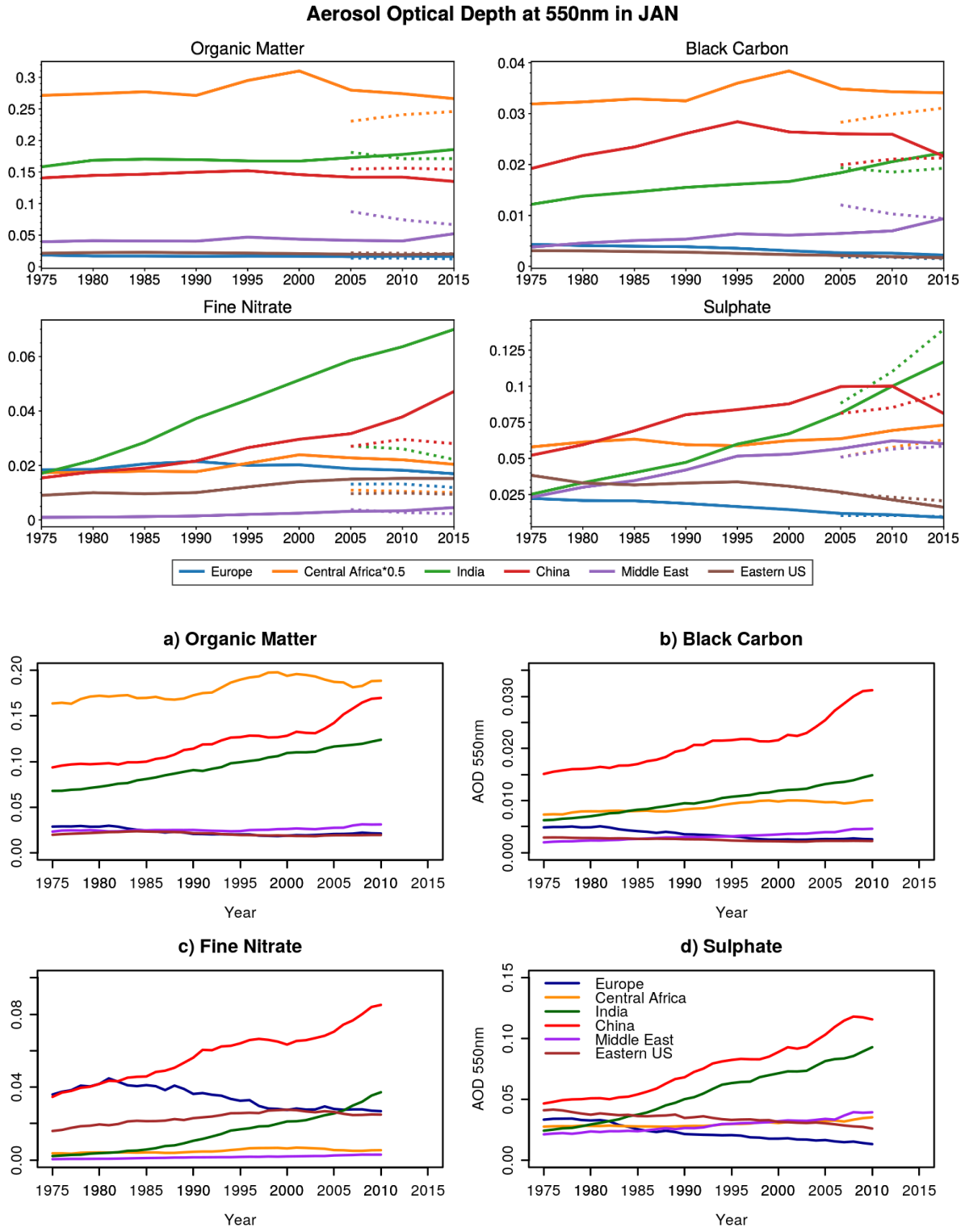

Figure 10: Time-variation of January AOD from HARMONIZED-CMIP6 and CAMS-FORCED (above) and EC-Earth3-AerChem (below), details as for previous figures.

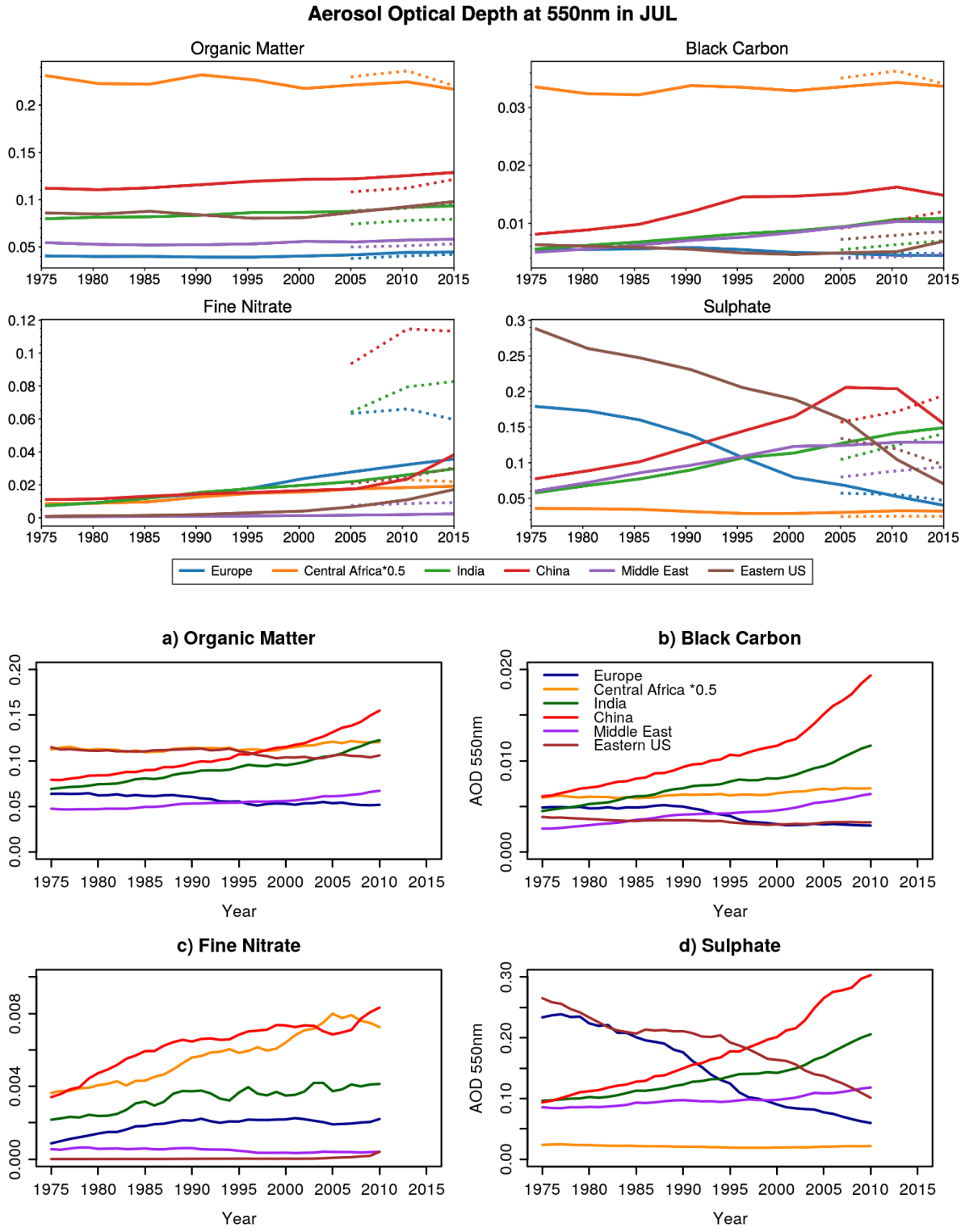

Figure 11: Time-variation of the July AOD from HARMONIZED-CMIP6 and CAMS-FORCED (above) and EC-Earth3-AerChem (below), details as for previous figures.

These plots tell a similar story to that of the aerosol mass comparison – areas of discrepancy in mass loadings translate into discrepancies in AOD.

Comparing per-species AOD between models is not sufficient, because of potential differences in species represented and optical properties. As a final comparison we present (Figure 12) the total AOD from all species (including dust and sea-salt, this is what can be most readily verified against observational data); and the absorptive AOD (Figure 13), representing the total clear sky aerosol absorption at 550nm (not easily verifiable by direct observations, but what matters for the direct radiative impact on the atmosphere).

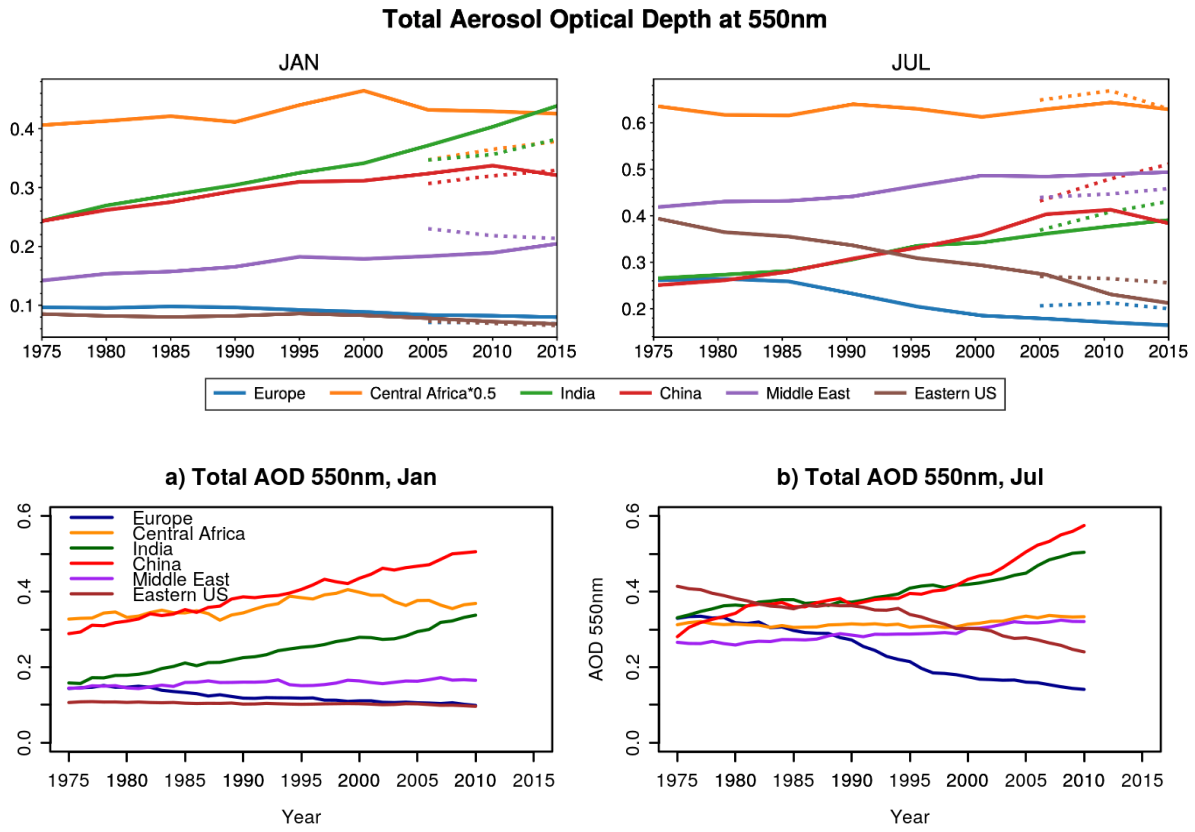

Figure 12: Time-variation of the total AOD from HARMONIZED-CMIP6 and CAMS-FORCED (above) and EC-Earth3-AerChem (below), details as for previous figures.

Again we see the relative excess of AOD in July over central Africa in the HARMONIZED-CMIP6 data compared to EC-EARTH3. HARMONIZED-CMIP6 also has moderately greater AOD over India in January, and slightly less in July, on the other hand it has increased AOD over the Middle East in July. EC-EARTH3 has moderately higher AOD over China for both seasons and throughout the whole period. Agreement over Europe and US tends to be good, although EC-EARTH3 has slightly higher AOD over Europe in the 1970s.

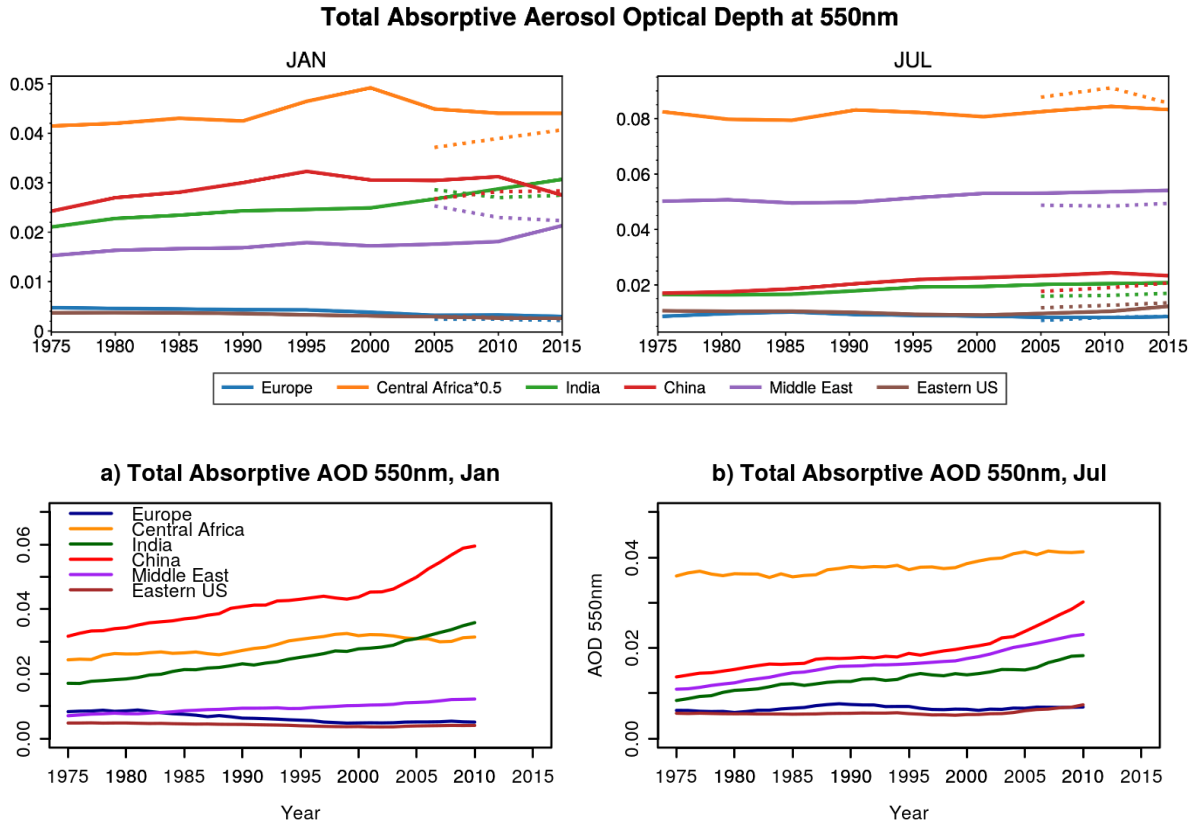

Figure 13: Time-variation of the total absorptive AOD from HARMONIZED-CMIP6 and CAMS-FORCED (above) and EC-Earth3-AerChem (below), details as for previous figures.

The HARMONIZED-CMIP6 aerosol has substantially more absorption than the EC-Earth3-AerChem aerosol throughout the tropics. The high values and relative lack of variation over time in the Middle East in July suggests that mineral dust may be playing a substantial role here. We note that these data are from the raw HARMONIZED-CMIP6 data, while in future CY47r3 IFS runs the scaled version would be recommended, which would have major differences in the mineral dust size distribution. Alternatively, the raw HARMONIZED-CMIP6 data might be used in future IFS versions where the mineral dust optical properties have been updated. In either case, the absorptive AOD is likely to differ from the values plotted here.

In summary, the comparisons of HARMONIZED-CMIP6 with EC-Earth3-AerChem show a broad level of comparability, and similar time evolution. Agreement is best where our knowledge of aerosols and emissions is best. Differences are in general typical of the differences found when comparing different aerosol modelling systems, a good summary of which is provided by Gliss et al. (2021). The largest and most consequential differences found (OM and BC over Africa in July, and total absorptive aerosol over dust affected regions) are supportive of the rescalings suggested for use in IFS CY47r3. More detailed differences between the datasets do not allow meaningful conclusions to be drawn because of the overall levels of uncertainty in state-of-the-art aerosol modelling.

6 Conclusions

6.1 Recommendations for future developments

We consider the approach we have taken to be fundamentally sound, and that the method is naturally extensible for future use. As aerosol modelling continues to develop, we expect that the accuracy of the model-generated aerosol fields will also tend to improve over time. Nonetheless, we have three specific recommendations that we hope will drive further improvements.

6.1.1 Improve the time-variability of CH4

As has been noted, CH4 interacts with OH, and changes in CH4 can drive changes in the atmospheric chemical balance, affecting nitrate aerosols in particular, with small knock-on effects on sea-salt aerosol. We do not have reliable information on the long-term history of highly reactive gases such as OH in the atmosphere, so we cannot be certain we have this right. However, we do know the time-history of methane, and we endeavoured to use that in our experiments. Unfortunately, although methane was initialised correctly in our runs, the CAMS chemistry includes a surface deposition module which strongly constrains surface methane to a specified climatology, which varies over time using linear trends based on 2003-2014. Although broadly acceptable over the period 1990-2020, for earlier dates this substantially overestimates the amount of CH4 present. CH4 in the years after 2020 has also been increasing at a rapid rate. These changes in CH4 are poorly understood and hence cannot be reliably modelled. However we recommend that the imposed surface values are specified in a more realistic manner. This will increase the accuracy of nitrate aerosols in the earlier years, with changes of order 5% or so. It is only a small improvement, but it is easy to implement.

6.1.2 Repeat with Cy48r1

Cycle 48r1 of the IFS will be released soon. Amongst many other changes it includes some substantial revisions to the aerosol model. A new secondary organic aerosol species will be introduced, with a significant indirect source from biomass burning. Alongside this, the emission factor for OM from biomass burning will be reduced from 3.4 to 1.5, and there will be changes in OM optical properties and a faster rate of ageing from hydrophobic to hydrophilic OM and BC. There is also a package of changes affecting mineral dust, including revised optical properties, which address biases seen after the recent change to the dust size distribution.

Cy48r1 thus addresses some of the major shortcomings that are suspected in the 47r3-derived climatology. The aerosol species in Cy49r1, planned for use by both ERA6 and SEAS6, are expected to remain the same, with possible further changes to optical properties (such as a reduction in BC absorption). This means that a climatology derived from Cy48r1 could be used directly in Cy49r1. This would be beneficial both to ERA6 and SEAS6, and also ease future experimentation with interactive aerosol, since both climatological and interactive aerosol would use the same set of species.

6.1.3 Coordinate the development of the aerosol model and optical properties

One of the limitations that became apparent during this work is that the development of the aerosol model and associated optical properties by external contractors to CAMS has not been coordinated with testing at ECMWF to establish the meteorological impact of changes. Rather, the focus of aerosol

development has been on fitting aerosol size data and aerosol optical depths, with decisions based on acceptance into each new cycle then based on the impact on the CAMS assimilation system.

To ensure that the radiative impact of the aerosols within the IFS is reasonable, it will be necessary to test this directly as part of the development and acceptance process. This will require potentially both an additional focus on NWP scores in CAMS when accepting new changes, and more direct contact and coordination between the CAMS aerosol model developers and those within ECMWF developing and testing the radiation scheme at both NWP and long-range timescales. Although such coordination involves a modest additional effort, it is likely to give large benefits in terms of a much more rapid convergence of the aerosol scheme to a position where it is beneficial to the model radiative behaviour.

6.2 Recommendations for usage

For seasonal forecast systems, the usage of a time-varying aerosol climatology is straightforward. Real-time forecasts and reforecasts should be made in a compatible way, and both will use a "time-averaged" estimate of aerosol loadings that is ignorant of the observed meteorological variations of a specific month. For the aerosol climatology in the near-term future, we suggest simple constant extrapolation of the most recent "epoch" available, on the grounds that we are unsure whether values will increase or decrease. The dataset is not designed to provide estimates many decades into the future.

For use in reanalysis, we recommend a similar approach, namely to represent the aerosols with a time-varying climatology. It would in principle be possible to create and use a version of the dataset for past dates without the 9-year smoothing, which would correspond to using monthly aerosol values specific to each month, thus capturing specific events such as forest fires. Although this approach can increase the realism of aerosol variability within the reanalysis, it has serious drawbacks. It cannot be implemented in a near-real-time re-analysis system, at least not in a monthly-mean form. And although it captures a part of aerosol variation, it still misses most of it, which happens on hourly or daily timescales. To build a reanalysis system which can accurately capture both the synoptic and long term variations of aerosol loadings in a realistic and traceable way, and run consistently both as a near-real-time system and over many decades of the past is a worthy scientific goal, but is a major undertaking which would require a dedicated approach.

We recommend that for experimentation involving Cy47r3 of the IFS, version HARMONIZED-CMIP6-SCALED should be used. For use in other models or with other IFS cycles, we suggest that it might be more appropriate to start with the raw HARMONIZED-CMIP6 dataset, and assess by experiment what scalings are most appropriate for that particular model and its particular aerosol optical properties. We further strongly recommend that users should contact ECMWF, to see if updated datasets are available, in particular updated datasets that are suitable for IFS model cycles beyond 47r3.

6.3 File details

Two versions of the time-varying aerosol climatology are available, `CONFESS_aerosol_1971-2019.nc` and `CONFESS_aerosol_1971-2019_scaled.nc`, corresponding to the HARMONIZED-CMIP6 and HARMONIZED-CMIP6-SCALED datasets described in this report. Each dataset consists of a

single netCDF file containing the 14 aerosol types, interpolated as described in Section 3.1.3. The netCDF metadata within the files are self-explanatory.

Additional files containing the vertically integrated aerosol masses (`VINT_aerosol_1971-2019.nc`) and the zonal mean mass mixing ratios (`ZONM_aerosol_1971-2019.nc`) are also provided for diagnostic purposes. Again, the netCDF metadata should be self-explanatory.

File listing:

```
-rwxr-xr-x   582910420 Jun 29 22:38 CONFESS_aerosol_1971-2019.nc
-rwxr-xr-x   582910440 Jun 29 22:42 CONFESS_aerosol_1971-2019_scaled.nc
-rwxr-xr-x    27409928 Jun 29 22:38 VINT_aerosol_1971-2019.nc
-rwxr-xr-x    27409948 Jun 29 22:42 VINT_aerosol_1971-2019_scaled.nc
-rwxr-xr-x     4860980 Jun 29 22:38 ZONM_aerosol_1971-2019.nc
-rwxr-xr-x     4861000 Jun 29 22:42 ZONM_aerosol_1971-2019_scaled.nc
```

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ECMWF	5 + 5 additional unfunded = 10
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