



Supplement of

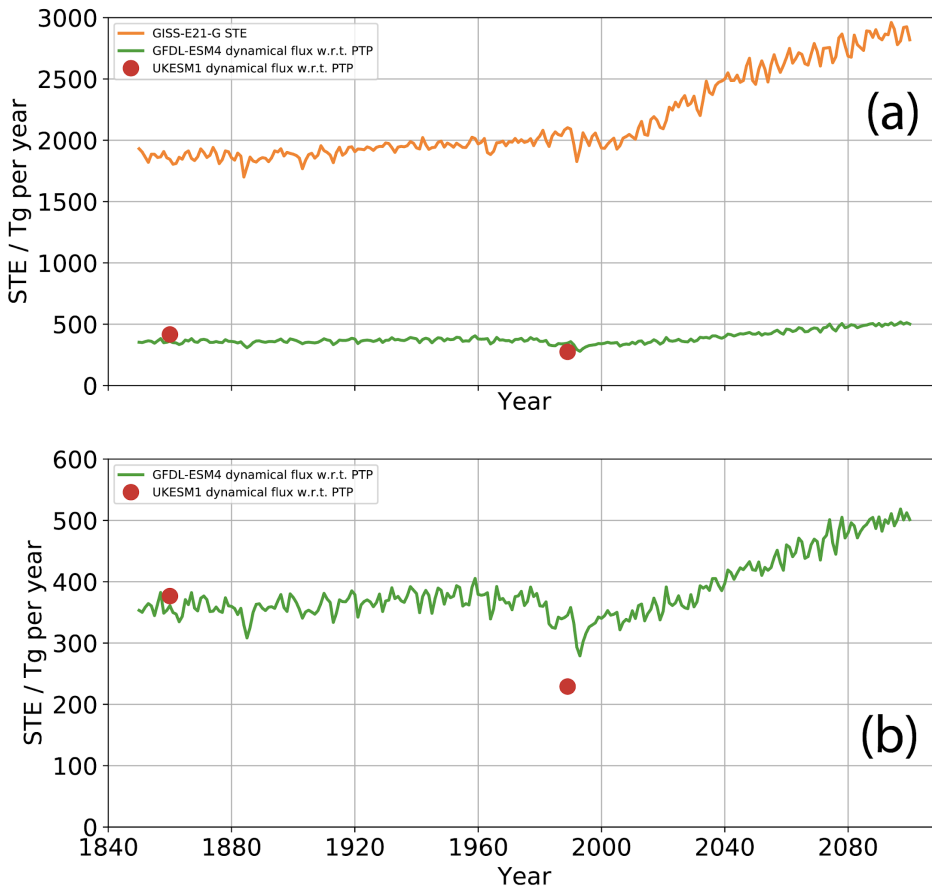
Tropospheric ozone in CMIP6 simulations

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S1 - Dynamically calculated stratosphere-to-troposphere transport of ozone



5 *Figure S1: dynamical net stratosphere-to-troposphere ozone flux calculated for GISS-E21-G (orange line), GFDL-ESM4 (green line) and UKESM1 (red circles). Figure S1(a): all data; (b) expanded view showing data for GFDL-ESM4 and UKESM1 on expanded scale*

Figure S1 shows the net stratosphere-to-troposphere ozone flux calculated for three of the models for which data
10 were available for calculation.

GFDL-ESM4 data were calculated using the ozone advective tendency diagnostic relative to the WMO definition
of tropopause (ptp) in the CMIP Historical and ScenarioMIP SSP370 simulations. The calculations used monthly
15 mean ozone tendency (in VMR/s) due to advection integrated over the region below the tropopause defined
using monthly mean PTP diagnostic.

UKESM1 data were calculated in a similar fashion using an ozone tendency due to dynamics and deriving transport
across the WMO PTP tropopause. Data are from repeat timeslice experiments from the histSST AerChemMIP
experiment which uses identical forcings as in the CMIP Historical experiment but in an atmosphere-only
20 configuration using sea-ice and SST fields taken from the coupled historical CMIP experiment.

GISS-E21-G data were similarly calculated online in the transient simulations as the vertical advective mass flux
of odd-oxygen across the WMO tropopause at each time step.

25 It can be seen that the figure shows good agreement between the magnitude of this term as calculated in the GFDL-ESM4 and UKESM1 models, particularly in the period around 1850, of 300-400 Tg per year, consistent with earlier model estimates, and with the residual in this period, but that GISS-E21-G model, with its consistently higher tropopause, calculates an order of magnitude higher STE. In the present day, GFDL-ESM4 and UKESM1 diverge, largely as the result of higher ozone depletion calculated in UKESM1.

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S2 - Breakdown of tropospheric ozone burden and budget by hemisphere

35 Figure S2 shows the breakdown of the budget and burden data for the four models analysed in detail, by hemisphere.

For all four models, the NH DD is consistently higher than the SH DD due to a much larger landmass in the NH, and the difference between the NH and the SH increases substantially from 1950 onwards as the result of rapid increase in ozone. The NH and SH NCPs are quite close at the beginning of the simulation (1850) but NH NCPs increase much faster from the 1950s for all models due to the increase in ozone precursor emissions. The SH NCPs are close to zero or below zero (e.g., the MRI model) throughout the simulation period. The evolution of the residual shows a more complicated tendency; the SH residual is more or less flat before the 1980s for all models before a slight decrease towards the year 2000 and then increases again after year 2000. It coincides with the Antarctic ozone depletion which may be reflected in the SH residual in the models. The exception is the MRI-ESM2-0 model which has a shallower Antarctic ozone depletion (Figure 17). All models show a large decrease in the NH residual after ~1950s before recovering from the 2000-2020 period. The extent of the residual decrease varies among the models with the UKESM-0-LL showing the largest change, which determines the very low global residual. The evolution of ozone burden shows continuous increases in both the NH and the SH for all models, with a larger increase in the NH.

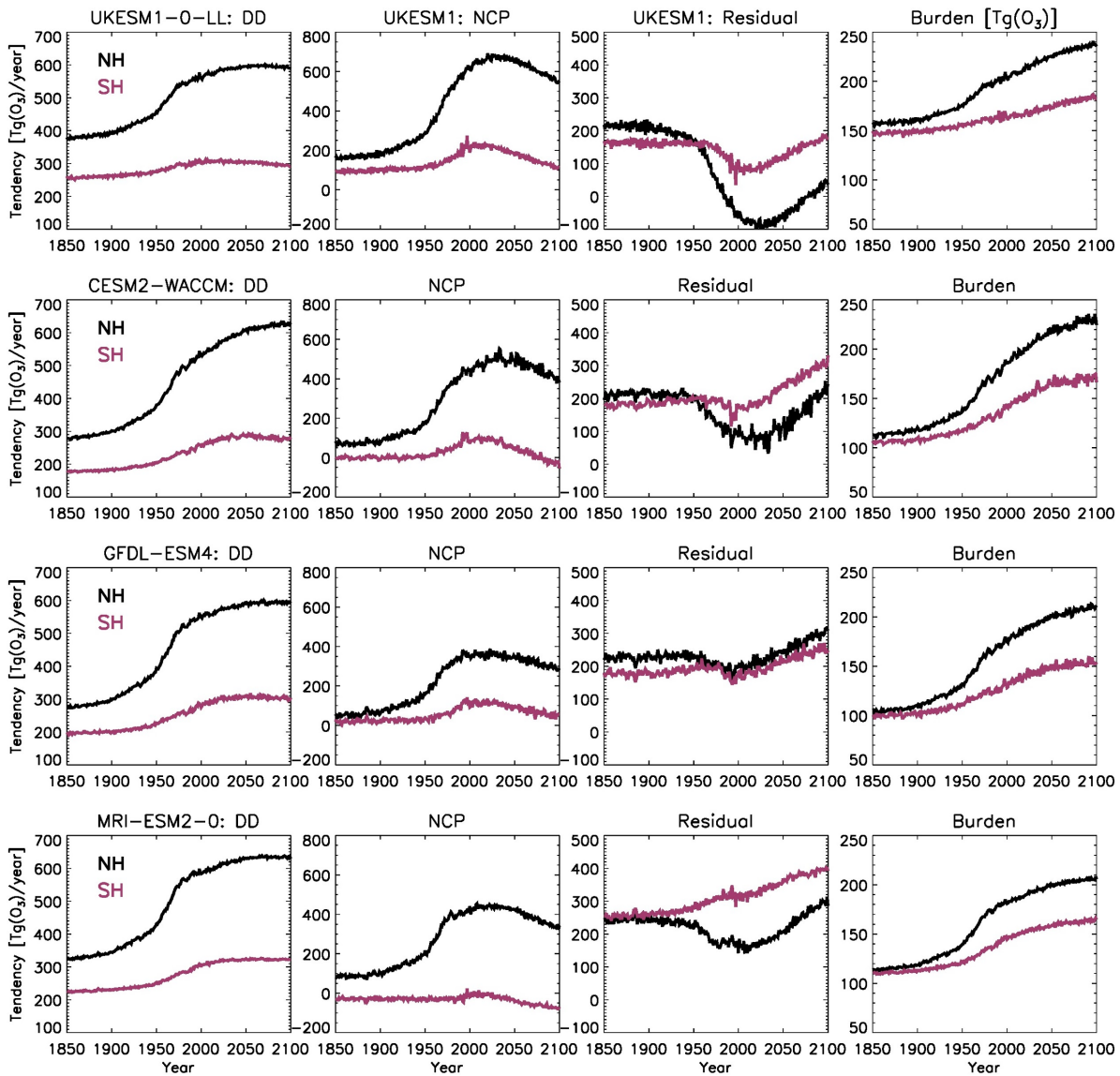


Figure S2

55 Evolution of integrated hemispheric ozone burden and the ozone budget terms of dry deposition (DD), net chemical production (NCP), and the “Residual” (i.e., residual = DD - NCP, and implies the net stratosphere-to-troposphere transport). The tropopause is defined using the WMO definition, the same as noted in the main text. The ozone burden and the budget terms are integrated over the Northern Hemisphere (NH) and the Southern Hemisphere (SH) respectively, assuming no net interhemispheric transport.

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Table S1 – Brief description of the chemistry and aerosol set up within CMIP6 models used in this study

CMIP6 Model	Horiz. Res.	Vert levels (top level)	Aerosol scheme	Aerosol Species	Natural Sources	Treatment of SOA	Chemistry Scheme	Chemistry reactions	BVOCs	Model Ref
CESM2-WACCM	0.9° x 1.25°	L70 (6x10 ⁻⁶ hPa)	MAM4 (modal scheme, simulating mass and number concentrations) with VBS-SOA	SO ₄ , BC, OM (both primary and secondary), sea salt, dust	Prescribed climatology of DMS seawater concentrations and emissions. Online emissions of sea-salt and dust aerosols. NOx calculated from lightning. Soil NOx and ocean CO, VOCs from POET	Explicit calculation of SOA using volatility basis set (VBS) where aromatic species, terpenes and isoprene are oxidised to produce a range of gas-phase SOA precursors with different volatilities. Formation of SOA linked to BVOCs emissions from interactive land surface scheme.	MOZART-TSMLT1 covering troposphere, stratosphere, mesosphere and lower thermosphere	231 gas-phase species, 150 photolytic reactions, 403 kinetic reactions and 30 heterogeneous reactions involving ClOx, BrOx, NOx-HOx-Ox, CO, CH ₄ and NMVOCs.	Online biogenic emissions (isoprene, monoterpenes, acetone, methanol, and other short and long-chained hydrocarbons) from dynamically evolving vegetation computed in the Community Land Model (CLM) using the MEGAN2.1 algorithm, which has dependence on light and temperature but also inhibits isoprene emissions based on CO ₂ .	(Gettelman et al., 2019; Tilmes et al., 2019; Emmons et al., 2020)
GFDL-ESM4	cubed-sphere (c96) grid, with ~100 km native resolution, regridded to 1.0° x 1.25°	L49 (0.01 hPa)	Bulk mass-based scheme. 5 size bins are used for sea salt and dust.	NH ₄ , SO ₄ , NO ₃ , NH ₄ , BC, OM, sea salt, dust	DMS and sea salt emissions calculated online as a function of wind speed (and a prescribed DMS seawater climatology). Dust emissions coupled to interactive vegetation. Lightning NOx calculated online as a function of convection. Natural emissions of NOx, CO, NMVOCs, H ₂ , and NH ₃ from POET. NH ₃ from seabird colonies. Two-way exchange of NH ₃ with ocean.	SOA formed simulated using an anthropogenic source from oxidation of C ₄ H ₁₀ tracer and a tracer representing BVOC emissions from vegetation	Interactive stratosphere-troposphere	43 photolysis reactions, 190 gas-phase kinetic reactions and 15 heterogeneous reactions. NOx-HOx-Ox- chemical cycles and CO, CH ₄ and NMVOC oxidation reactions	Online emissions of BVOCs (isoprene and monoterpenes) calculated from a prescribed vegetation cover using MEGAN2.1 algorithm, which has dependence on light and temperature but also inhibits isoprene emissions based on CO ₂ .	(Horowitz et al., 2019; Dunne et al., 2020)
GISS-E2-1-G	2° x 2.5°	L40 (0.1 hPa)	OMA (one moment aerosol scheme – mass based)	SO ₄ , NO ₃ , NH ₄ , BC, OM treated as externally mixed with prescribed and constant size distribution. Sea salt has two size classes. Sectional scheme for dust with 5 size bins that can be coated with SO ₄ and NO ₃ to increase solubility.	Sea salt, DMS, isoprene and dust emission fluxes are calculated interactively. Online NOx calculated from lightning. Soil NOx, ocean CO, VOCs from GEIA. NH ₃ from oceans. SO ₂ from volcanoes as in AeroCom.	Two-product model approximation to represent SOA formation from the oxidation of biogenic VOCs, including NOx dependent chemistry yields.	Coupled troposphere-stratosphere chemistry scheme. Modified Carbon Bond Mechanism 4 (CBM-4) chemical mechanism	inorganic chemistry of Ox, NOx, HOx, CO, and organic chemistry of CH ₄ and lumped higher hydrocarbons (only isoprene and terpenes are explicitly taken into account), along with Cl and Br stratospheric chemistry and heterogenous reactions on PSCs and SO ₄ aerosols.	Emissions of isoprene from dynamically evolving vegetation are calculated interactively using the algorithm of Guenther et al., (1995), which has dependence on light and temperature. Terpene emissions are prescribed.	(Bauer et al., 2020)

MRI-ESM2-0	MRI-AGCM3.5: 1.125° x 1.125°, MASINGAR mk-2r4c: 1.875° x 1.875°, MRI-CCM2.1: 2.813° x 2.813°	L80 (0.01 hPa)	MASINGAR mk-2r4c	Mass-based scheme with externally mixed size distributions. SO ₄ (three categories), BC (hydrophilic and hydrophobic), OM (hydrophilic and hydrophobic), sea salt (10 size bins), dust (10 size bins).	Interactive online emissions of DMS (using prescribed Climatological DMS sea water concentrations), sea-salt, and dust aerosols dependent on meteorology. Online NO _x calculated from lightning. Climatological soil NO _x and ocean CO, VOCs emissions.	No explicit calculation: 14% of prescribed monoterpene and 1.68 % of isoprene emissions are assumed to form SOA.	Chemistry Climate Model version 2.1 (MRI-CCM2.1) covering troposphere, stratosphere, and mesosphere	90 chemical species and 259 chemical reactions (184 gas-phase reactions, 59 photolysis reactions, and 16 heterogeneous reactions) involving HO _x -NO _x -CH ₄ -CO cycles and NMVOC oxidation reactions, and halogen chemistry (Cl and Br)	Climatological BVOCs emissions	(Deushi and Shibata, 2011; Yukimoto et al., 2019)
UKESM1-0-LL	1.25° x 1.875°	L85 (85km)	GLOMAP-Mode. (Modal scheme, mass and number). Mass based bin scheme used for dust.	SO ₄ , BC, OM, sea salt in 5 log-normal modes and dust in 6 bins	Dynamic vegetation and interactive ocean biogeochemistry used for online emissions of DMS, sea-salt and dust aerosols, as well as emissions of primary marine organics and biogenic organic compounds. Online NO _x calculated from lightning, soil NO _x and ocean CO, VOCs from POET	Fixed SOA yield of 26% from gas-phase oxidation reactions involving interactive land-based monoterpene sources.	UKCA coupled stratosphere-troposphere. Interactive photolysis	84 chemical tracers. Simulates chemical cycles of Ox, HO _x and NO _x , as well as oxidation reactions of CO, CH ₄ and NMVOCs. In addition, heterogeneous processes, Cl and Br chemistry are included.	Dynamic vegetation and land surface model used to calculate interactive emissions of Isoprene and monoterpenes using light and temperature, but isoprene emissions are inhibited based on CO ₂ . Isoprene emissions coupled to chemistry and affect tropospheric O ₃ and methane lifetime. Monoterpenes only affect SOA.	(Archibald et al., 2020; Mulcahy et al., 2020)