Electronic Supplementary Material (ESI) for Environmental Science: Atmospheres.

Impacts of a near-future supersonic aircraft fleet on atmospheric composition and climate: supplemental information

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Stratospheric age of air in GEOS-Chem

To evaluate the performance of the GEOS-Chem model in simulating stratospheric transport, an additional set of tracer transport simulations is performed at the same model resolution (4°×5°) and over the same time period (42 years), again using three repeats of the MERRA meteorological data for the period 2000-2014. In these simulations we use a “clock tracer”. This tracer is initialized to zero throughout the domain; at every subsequent time step, the mixing ratio throughout the lowest five model layers (nearest the surface) is re-set to a value proportional to the (simulated) time since the start of the simulation. The difference in mixing ratio between any point and the “current” surface mixing ratio then indicates the mean age of air at that location.

Figure S 1 compares the mean age of air at 20 km altitude to observations by Andrews et al. and model estimates by Chabrillat et al. using the BASCOE model and multiple different meteorological datasets[1,2]. We find that, consistent with Chabrillat et al., a model driven by MERRA meteorological data overestimates the age of air in the tropics by around 6 months. Outside of the tropics, the simulated age of air is within the uncertainty bounds of the results from Andrews et al., and matches observations well in the Northern Hemisphere. However, the simulated age of air in the Southern Hemisphere is near the lower limit of model estimates, suggesting that the model exhibits excessive horizontal mixing across subtropical transport barriers.

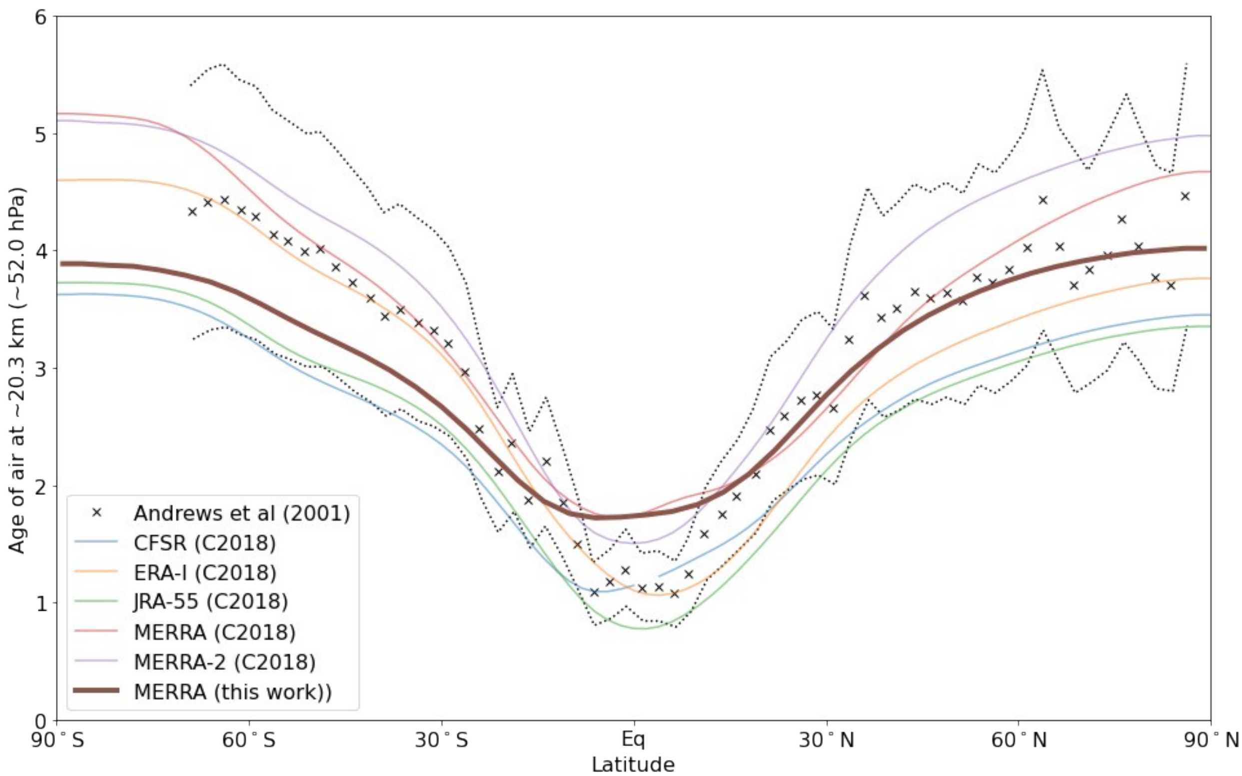

Figure S 1. Comparison of observed age of air from Andrews et al. (2001) to model-based estimates. Observations are shown as black crosses, with uncertainty bounds shown as dotted black lines. Model estimates from Chabrillat et al. (2018) produced using the BASCOE model are shown as faded, colored lines. Results from this work are shown as a thick brown line, and are based on the final 14 years of a 42 year simulation.

A possible cause of this discrepancy is the relatively coarse model resolution employed for this study (4°×5°), which was necessary to permit the large number of long-term simulations. We perform an additional sensitivity simulation at 2°×2.5°, in which only the clock tracer is simulated and non-transport processes are disabled.

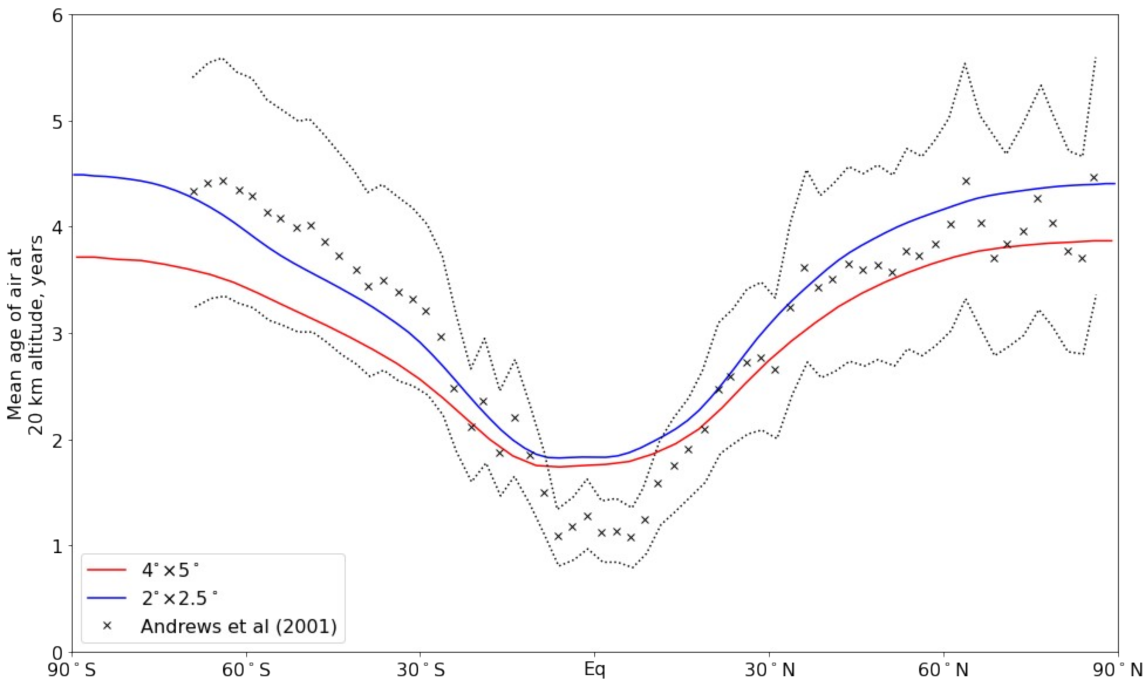

Figure S 2. Comparison of the simulated mean age of air at ~20 km altitude (lines) to observations by Andrews at al. (2001) (crosses). Data shown correspond to the 10th year of simulation at each resolution.

Figure S 2 compares the results of these two simulations to the same set of observations. We find that the higher-resolution simulation does increase the stratospheric age of air at all latitudes, with the greatest effect (a 4-6 month increase in age of air) at high Southern latitudes. This improves the agreement of the model with the observations in these regions. The age of air in the tropics remains around 2 years, meaning that the relative difference in age of air between the tropics and high latitudes is increased although the age of air in the tropics remains overestimated.

Methane feedbacks and equilibrium

Due to the long lifetime of methane in the atmosphere, simulations in which methane feedbacks are to be captured take several decades to reach near-equilibrium. Determining whether a simulation is “sufficiently” at equilibrium is subjective, but we here provide metrics of the degree of (dis-)equilibrium of our simulations with respect to methane.

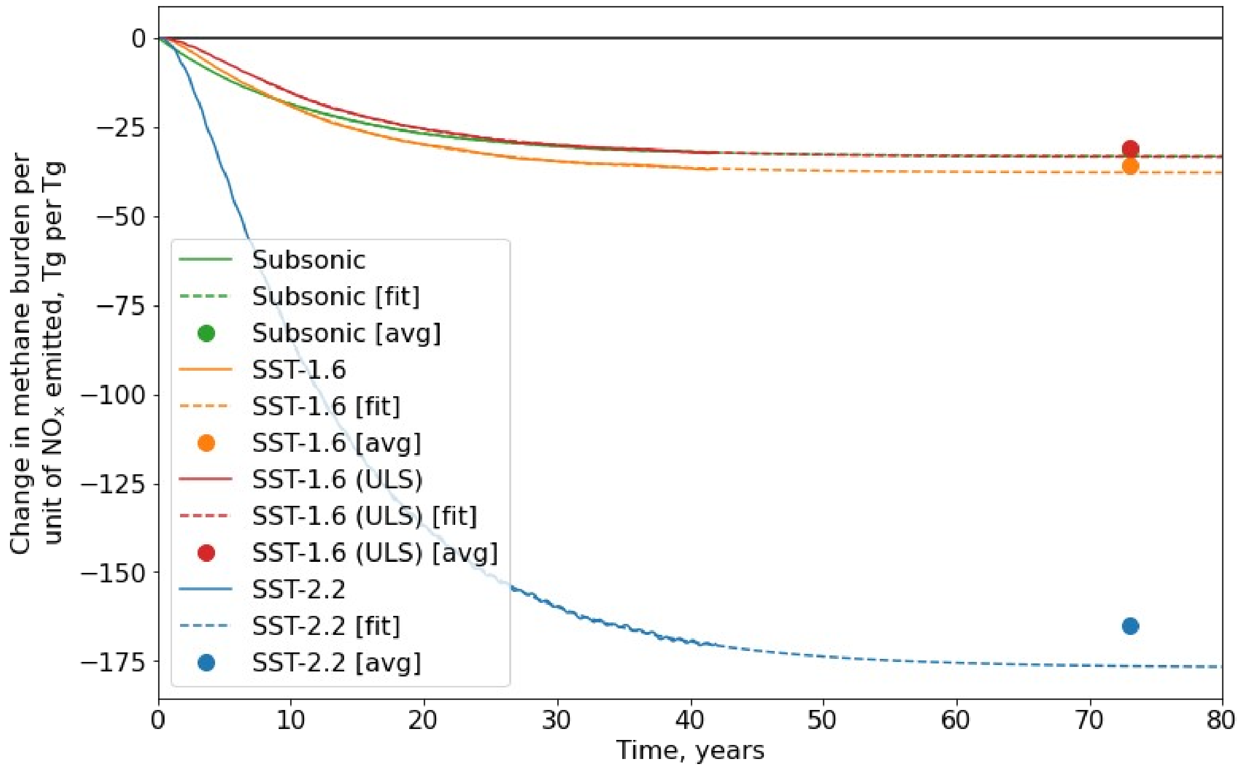

Figure S 3. Change in the global methane burden resulting from each simulation divided by the annual NO_x emission for that aircraft fleet. Solid lines show simulation data timeseries; dashed lines show an extrapolated fit ("[fit]"); filled circles ("[avg]") show the average of the last 14 years of simulation data (years 29-42).

Figure S 3 shows the change in global methane burden resulting from emissions by each fleet, divided by the annual NO_x emission for that fleet and plotted as a function of simulation time. Solid lines show simulation results, which dashed lines show an exponential fit. The fit is generated based on data from year eight onwards, to avoid influence from the initial transient behavior. Circles show the average change in methane burden over the 14-year averaging period used for the study's main results. In each case, 93-94% of the equilibrium change is captured by our approach. This implies that methane-related feedbacks, including on global distribution and concentrations of ozone and water vapor, may increase by a further 6-8% if longer simulations are performed.

Extended comparison of results to existing studies

A spreadsheet is attached to this supplemental information which compares the results of our work to that of previous studies. This table provides multiple normalizations of the data to facilitate this comparison, since not all studies report all relevant data. Where possible, we have attempted to infer quantities such as the ratio of ozone depletion between hemispheres, and the absolute or percentage depletion of ozone (using a reference value of 300 Dobson units for the conversion if one is not provided).

References

1 S. Chabrillat, C. Vigouroux, Y. Christophe, A. Engel, Q. Errera, D. Minganti, B. M. Monge-Sanz, A. Segers and E. Mahieu, Comparison of mean age of air in five reanalyses using the BASCOE transport model, *Atmos. Chem. Phys.*, 2018, **18**, 14715–14735.

2 A. E. Andrews, K. A. Boering, B. C. Daube, S. C. Wofsy, M. Loewenstein, H. Jost, J. R. Podolske, C. R. Webster, R. L. Herman, D. C. Scott, G. J. Flesch, E. J. Moyer, J. W. Elkins, G. S. Dutton, D. F. Hurst, F. L. Moore, E. A. Ray, P. A. Romashkin and S. E. Strahan, Mean ages of stratospheric air derived from in situ observations of CO2, CH4, and N2O, *J. Geophys. Res.*, 2001, **106**, 32295–32314.