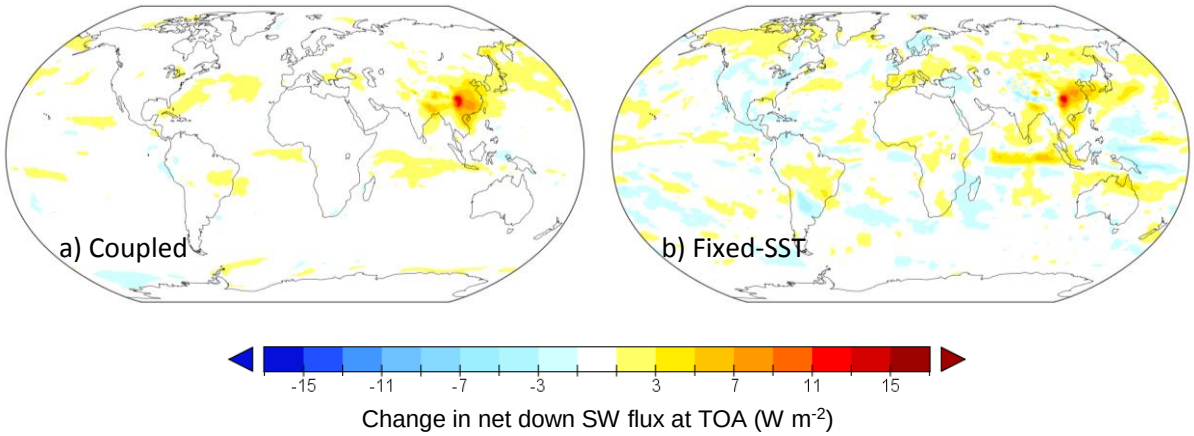
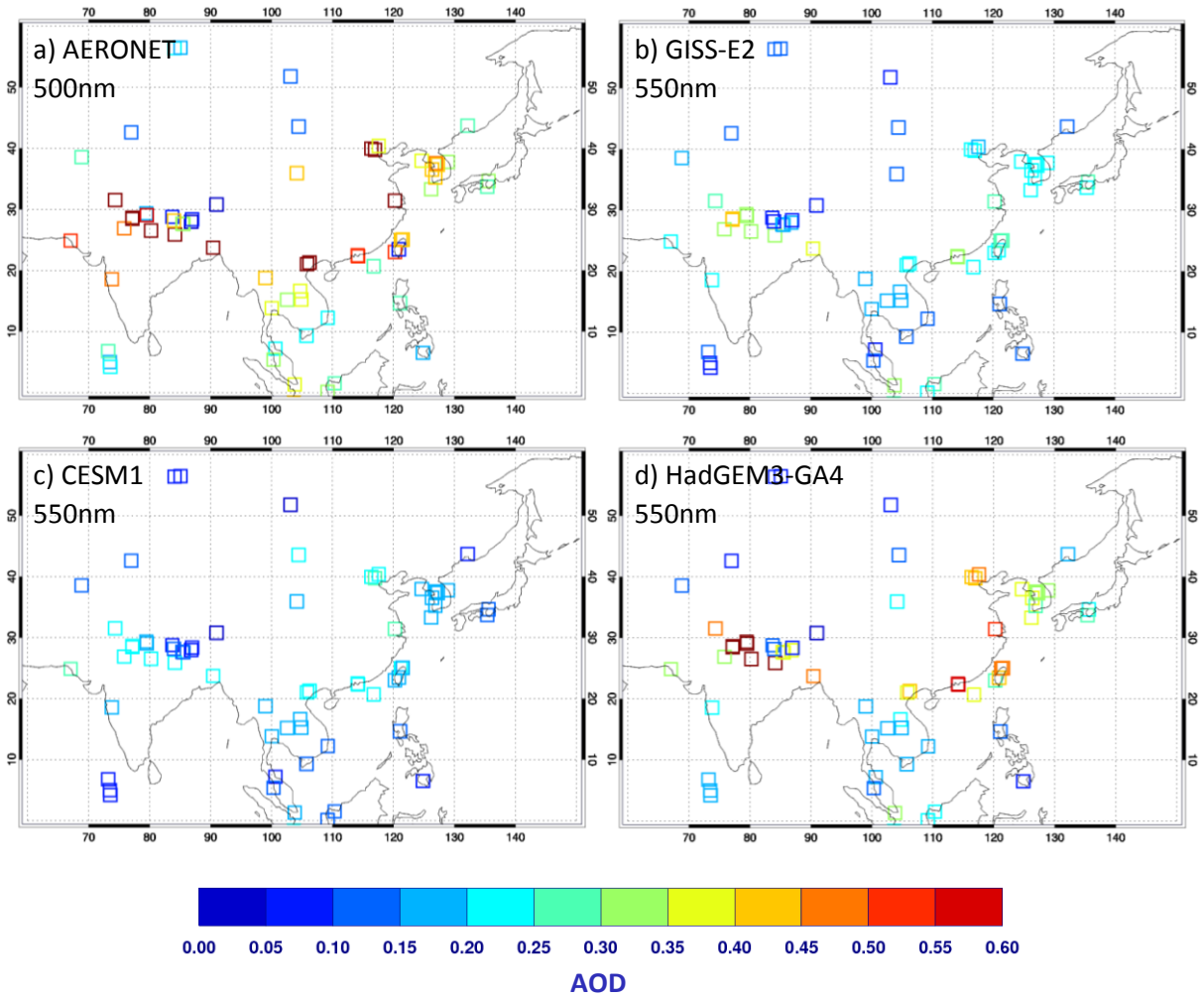
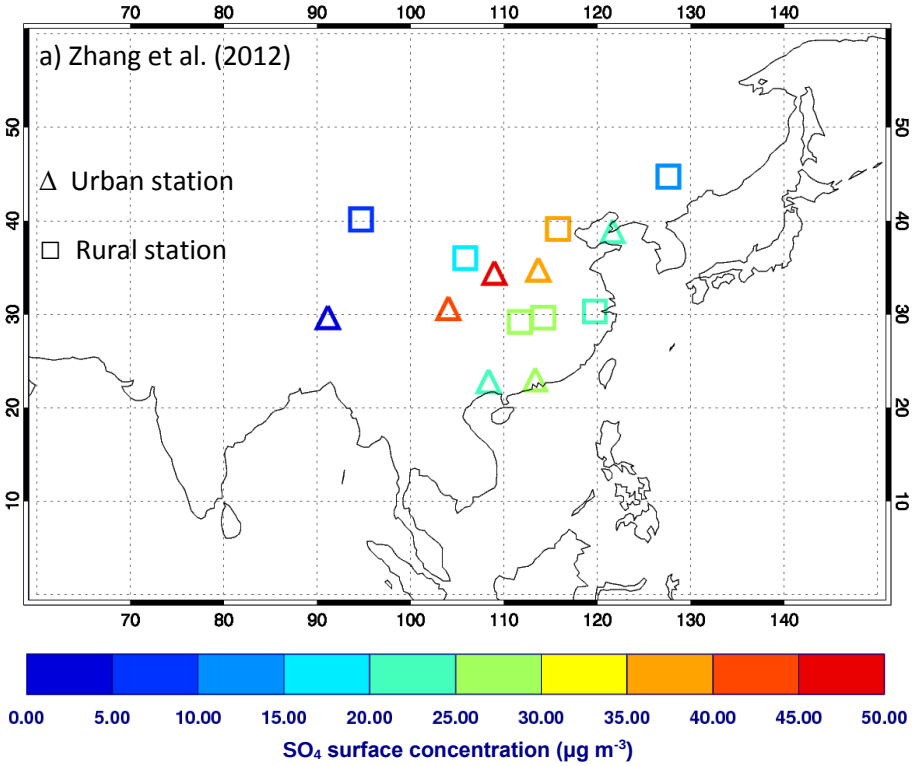


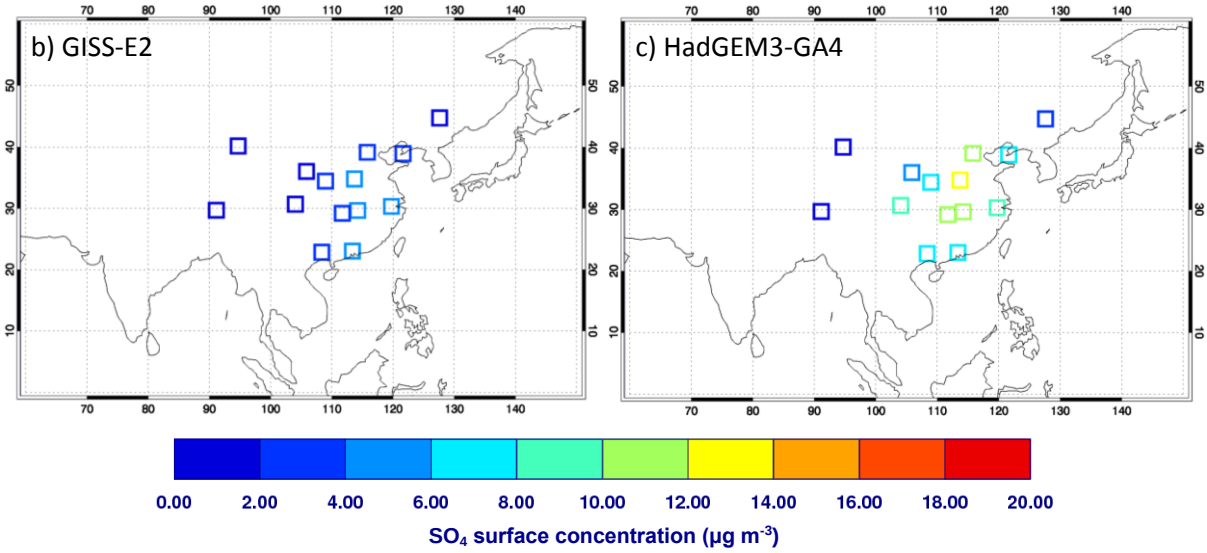
Supplementary Figure S1: Change in TOA SW flux due to removing SO_2 emissions from China in: a) fully-coupled simulations, and b) fixed-SST simulations, with HadGEM3-GA4. Both simulations are perturbation minus control; for the coupled simulation the plot shows an average over 150 years, with the first 50 years of the simulations discarded as spin-up. For the fixed-SST simulations, the model was run for 26 years with prescribed year 2000 SSTs and sea-ice concentrations, and the plot shows the average over the last 25 years, discarding the first year as spin-up.



Supplementary Figure S2: Annual mean AOD observed at 500nm by AERONET (a), and diagnosed at 550nm from GISS-E2 (b), CESM1 (c), and HadGEM3-GA4 (d). For AERONET the monthly mean climatology product was used, which for each station provides a value for each month averaged from all years that observations for that month were available. Only stations which could provide a climatological value for all 12 months were then used to calculate an annual mean climatology. For the models, the 150-year mean AOD from each model's control (2000) simulation is masked to the locations of the qualifying AERONET sites by taking the value in the gridbox that each AERONET station is located in. For HadGEM3-GA4 and GISS-E2 the AOD is diagnosed for clear-sky conditions, whereas in CESM1 all-sky AOD is diagnosed, which is expected to be higher than the equivalent clear-sky value. Because most AERONET stations in Asia stared observing more recently than year 2000 (the year the model simulations are based on), the observations may be skewed relative to the models. We assessed the severity of this issue by using MODIS and MISR to see how the same instruments observed AOD changing over China since 2000: For MODIS

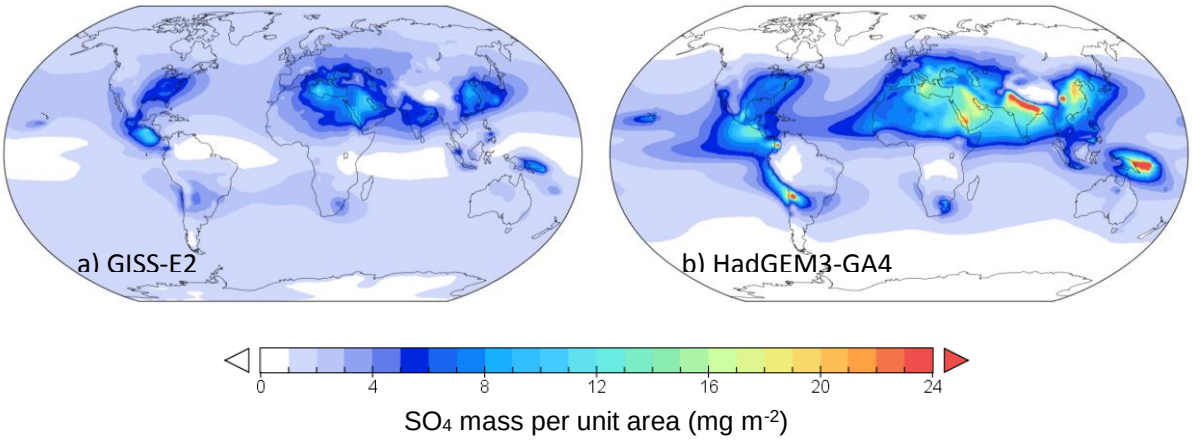
Terra the E. China average AOD at 550nm was 0.48 for 2001-2003, and 0.54 for 2010-2012. For MISR the E. China average was 0.29 for 2000-2002, and 0.31 for 2012-2014. So, we conclude that it does not appear that there has been a large trend in AOD in E. China over this period, and so the AERONET observations made over this period are likely still reasonably representative of year 2000 conditions.



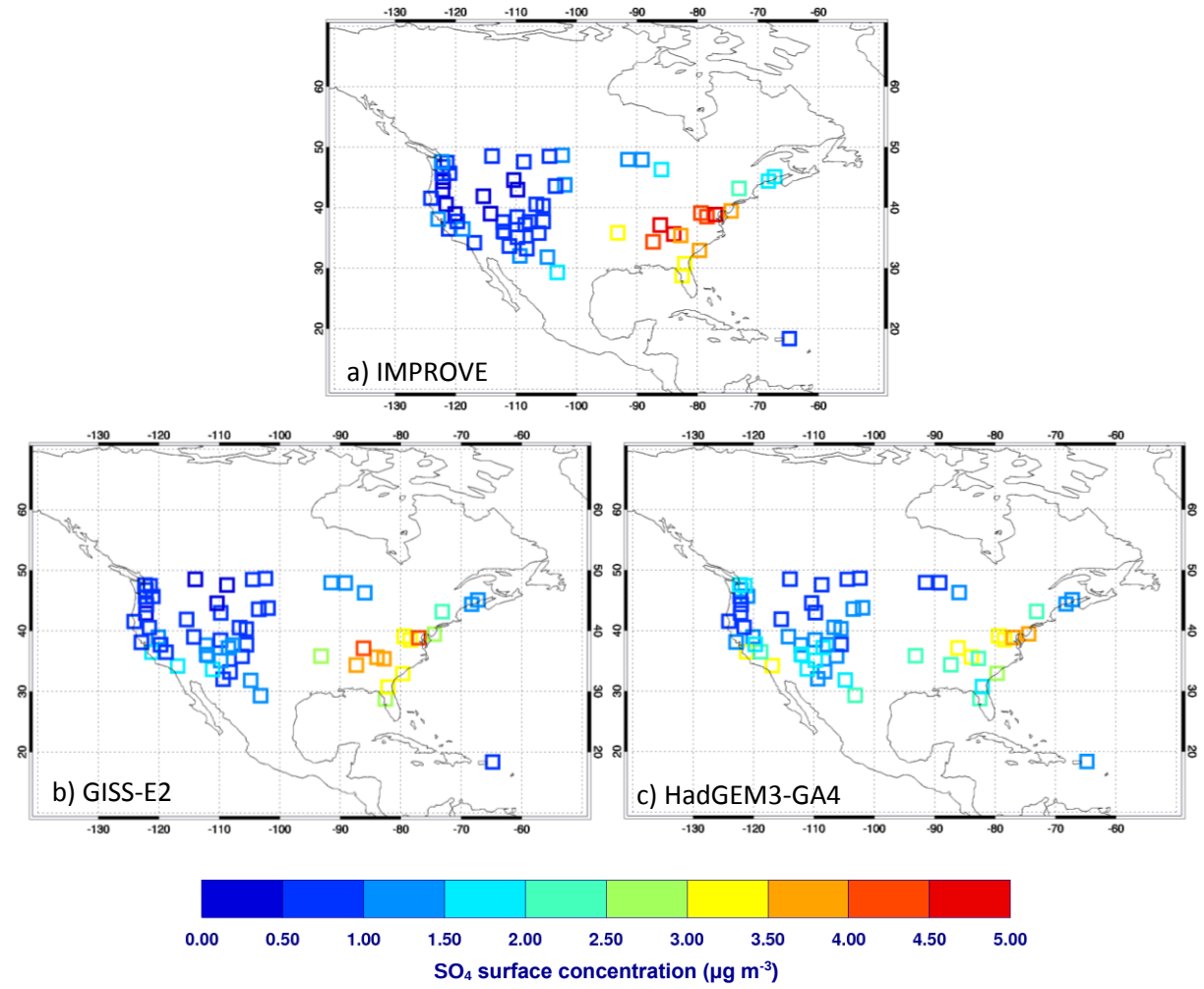


Supplementary Figure S3: Surface SO_4 concentrations in China as reported for 2006-2007 by Zhang et al. (2012) (a), and in GISS-E2 (b) and HadGEM3-GA4 (c). For the Zhang et al. dataset, a monthly climatology is calculated by averaging values for each month from both 2006 and 2007 (where available), and then all twelve months are averaged to calculate an annual mean. Stations located in urban areas are denoted by triangles. For the models, the 150-year mean from the control (2000) simulation of each model is used, and the data is masked to the locations of the observations by taking the value from the gridbox that each

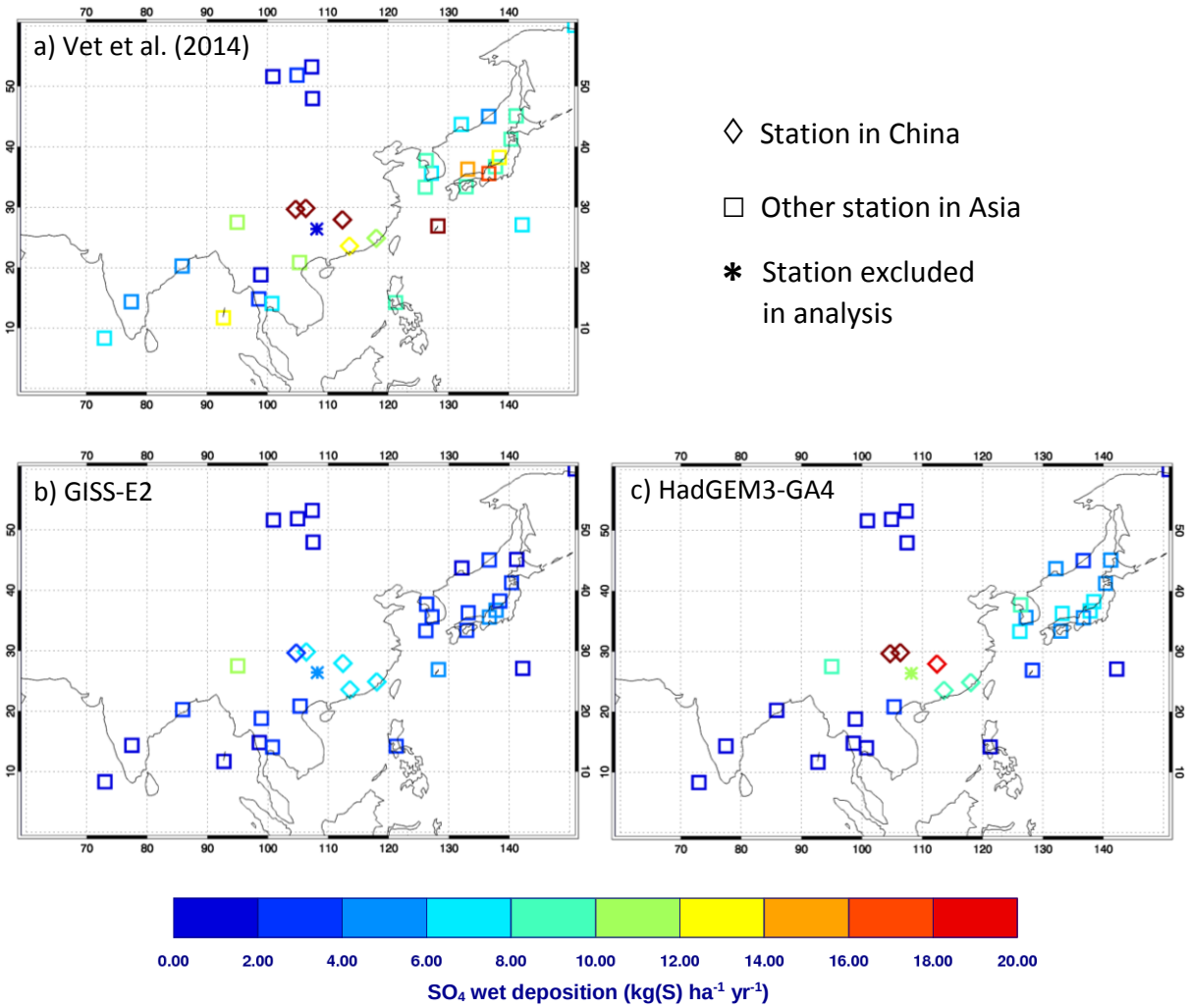
station is located in. Note the colour scale for the models is 2.5 times smaller than for the observations.



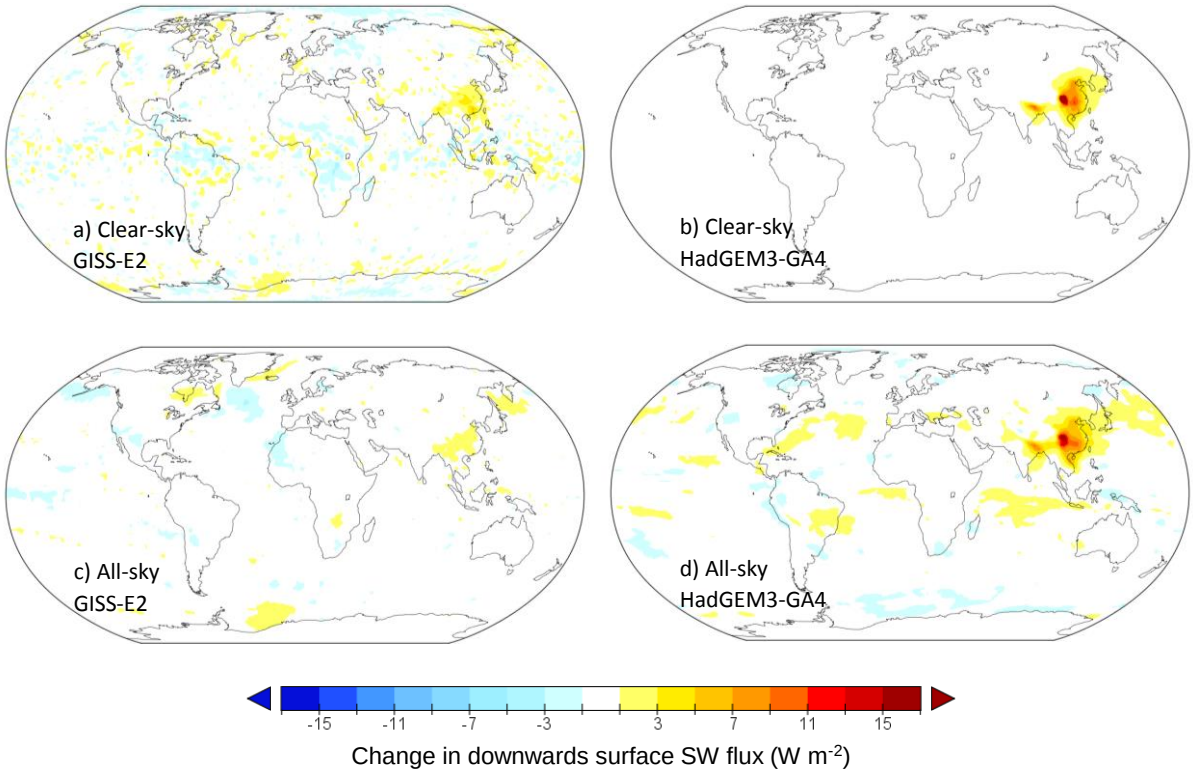
Supplementary Figure S4: Climatological column-integrated SO_4 burden in a) GISS-E2, and b) HadGEM3-GA4. In each case a 150-year average in taken from the control simulation of the model.



Supplementary Figure S5: Annual mean surface SO_4 concentrations in the US as observed by the IMPROVE monitoring network (a), and as diagnosed in GISS-E2 (b) and HadGEM3-GA4 (c). For the IMPROVE data we calculate monthly climatologies between 1995-2005 for all stations with data for each month from at least 6 years within this range, and average over months to calculate an annual mean climatology for each station. For the models, the 150-year mean surface SO_4 from each model's control (2000) simulations is masked to the location of the qualifying IMPROVE sites by using the value at the gridbox each site is located in.



Supplementary Figure S6: SO_4 wet deposition in Asia, reported for 2000-2002 in Vet et al. (2014) (a), and in GISS-E2 (b) and HadGEM3-GA4 (c). For the Vet et al. (2014) dataset, we use the 3-year mean values provided by the data product. For the models we use the 150-year mean from the models' control (2000) simulations, masked to the location of the observation stations by taking the value from the gridbox each station is located in. Diamonds indicate stations in China, and the asterisk indicates the station that was excluded from the analysis in the main manuscript, as both models had unusually large biases at this one station.



Supplementary Figure S7: Change in downwards surface SW flux in GISS-E2 and HadGEM3-GA4 respectively, for clear-sky (a, b) and all-sky (c, d) conditions. Each plot shows perturbation minus control simulation, averaged over 150 years.

References

Vet, et al.: A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmospheric Environment, 93, 3-100, doi:10.1016/j.atmosenv.2013.10.060, 2014.

Zhang, X. Y., Wang, Y. Q., Niu, T., Zhang, X. C., Gong, S. L., Zhang, Y. M., and Sun, J. Y.: Atmospheric aerosol compositions in China: spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols, Atmos. Chem. Phys., 12, 779-799, doi:10.5194/acp-12-779-2012, 2012.