*Earth's Future*

Supporting Information for

# Isolating the meteorological impact of 21st century GHG warming on the removal and atmospheric loading of anthropogenic fine particulate matter pollution at global scale

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**Text S1**.

1. Evaluation of present-day model simulation

The simulation of major aerosol species (sulfate, black carbon, organic carbon) in this model configuration (CESM1/CAM5/MAM3) have been evaluated against with long-term surface concentration measurement from the Interagency Monitoring of Protected Visual Environment (IMPROVE) ground sites in the United States and the European Monitoring and Evaluation Program (EMEP) sites; field campaign measurement such as HIAPER Pole-to-Pole Observations campaign (HIPPO) and ARCTAS (NASA Arctic Research of the Composition of the Troposphere); remote sensing measurement of aerosol optical depth from several satellites; ground based measurement of aerosol optical depth from AERONET (Aerosol Robotic Network), and also with other chemistry model output from AeroCom model intercomparison project [*Liu et al.,* 2012]. The model performed reasonably well for most of NH continental regions. The simulated $SO_4$ agrees with observation within a factor of 2 for most of the US and Europe. But it was documented that $SO_4$ in the Western US is overestimated by the model, in particular during summer time (also seen here in Figure S2: SO4_JJA for the US).

One difference between our simulations here with those in *Liu et al.,* [2012] is that the meteorology is prescribed in *Liu et al.,* [2012], facilitating a more realistic simulation of chemical species distribution. In Figure S1, we compared the present-day (2006-2015) simulation of surface concentration and column burden of major aerosol species with a reanalysis product (Modern Era Retrospective analysis for Research and Applications Aerosol Reanalysis (MERRA2; *Buchard et al.,* [2016]). This reanalysis product assimilated aerosol optical measurements from satellite platform to provide a full global coverage and continuous record since 2002. The benefits of comparing model output with this reanalysis data set are to avoid the problem of inconsistent resolution of the model grid cell and observation sites, especially those close to source regions (e.g. cities).

The model performs well in capturing the present-day distribution of surface concentration as reported in previous studies, especially in the NH continental regions, our areas of focus (Figure S1). Notable low biases exist in the $SO_4$ simulation over the US and China, in particular during wintertime (December, January and February), which is partly compensated by an overestimation during summertime (Figure S2, blue shaded panels).

Figure S2 presents the probability density function of surface concentration (logarithmic scale) of POM and $SO_4$ at each grid boxes over major regions, with our simulation shown in blue line. POM simulation over the US exhibit major underestimation during summer time and the winter time simulation appears in reasonable agreement. The larger seasonal biases than annual average is an issue that regional chemical models and global models commonly suffer [*Simon et al.,* 2012]. The mean bias of present-day pollution level suggests the 21$^{st}$-century increase as calculated in this paper should be interpreted in a more relative sense (5% to 15%).

In Figure S1a, we also compared present-day rainfall with the Global Precipitation Climatology Project (GPCP) observation [*Adler et al.,* 2003]. The general agreement is not surprising as this model configuration (CESM1-CAM5-MAM3) is a stable version of CESM1 development and the set of 20th and 21st-century simulations [*Kay et al.,* 2015] have been extensively evaluated and utilized for studying many aspects of the climate system.

Reference:

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Kay, J. E. et al. (2015), The Community Earth System Model (CESM) Large Ensemble Project: A Community Resource for Studying Climate Change in the Presence of Internal Climate Variability, Bull. Am. Meteorol. Soc., 96(8), 1333–1349, doi:10.1175/BAMS-D-13-00255.1.

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Simon, H., K. R. Baker, and S. Phillips, 2012: Compilation and interpretation of photochemical model performance statistics published between 2006 and 2012. Atmos. Environ., 61, 124–139, doi:http://dx.doi.org/10.1016/j.atmosenv.2012.07.012.

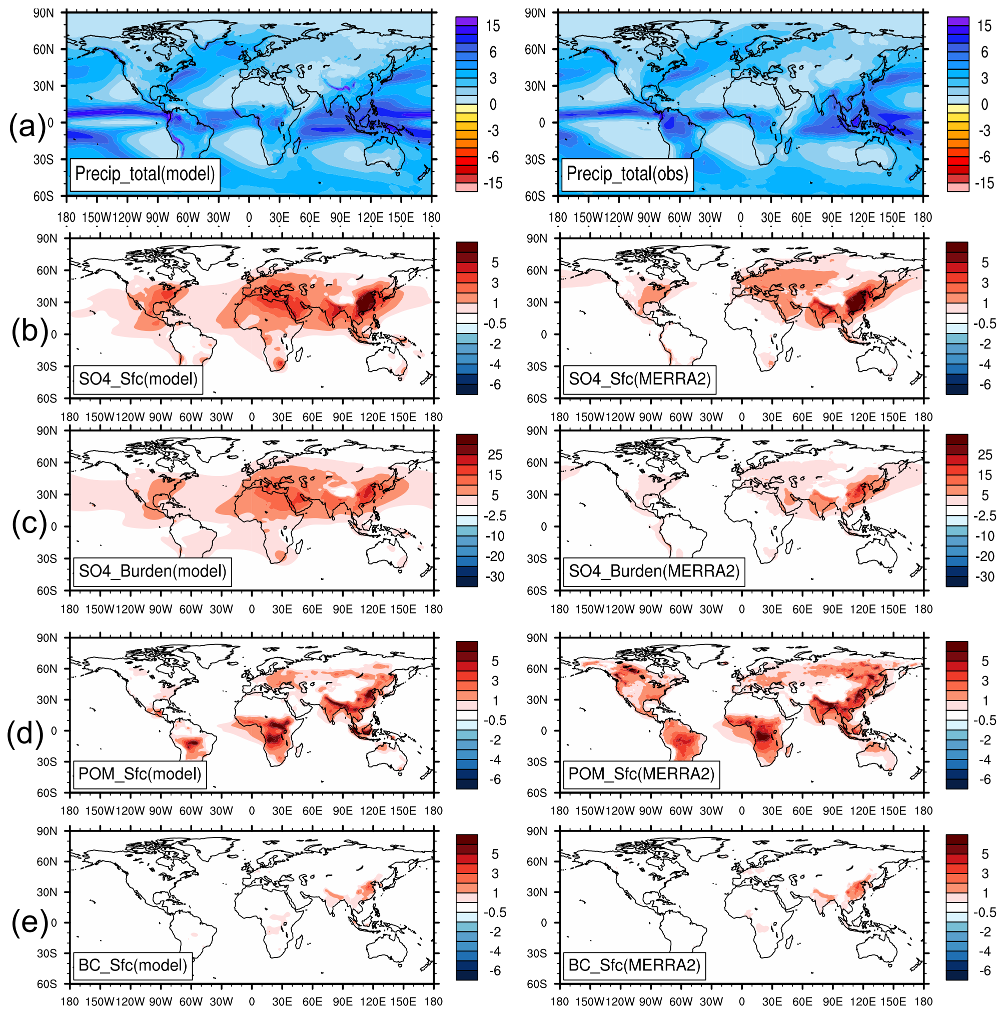


Figure S1. Comparison of various model output (left column) with observations and data products (right column). (a) Total precipitation (mm/day) as in Figure 4c compared with GPCP data set. (b) $SO_4$ surface concentration ($\mu g/m^3$) as in Figure 4a compared with the MERRA2 dataset. (c) $SO_4$ column burden ($mg/m^2$) as in Figure 3a compared with the MERRA2 dataset. (d-e) same as (b) but for POM and BC surface concentration.

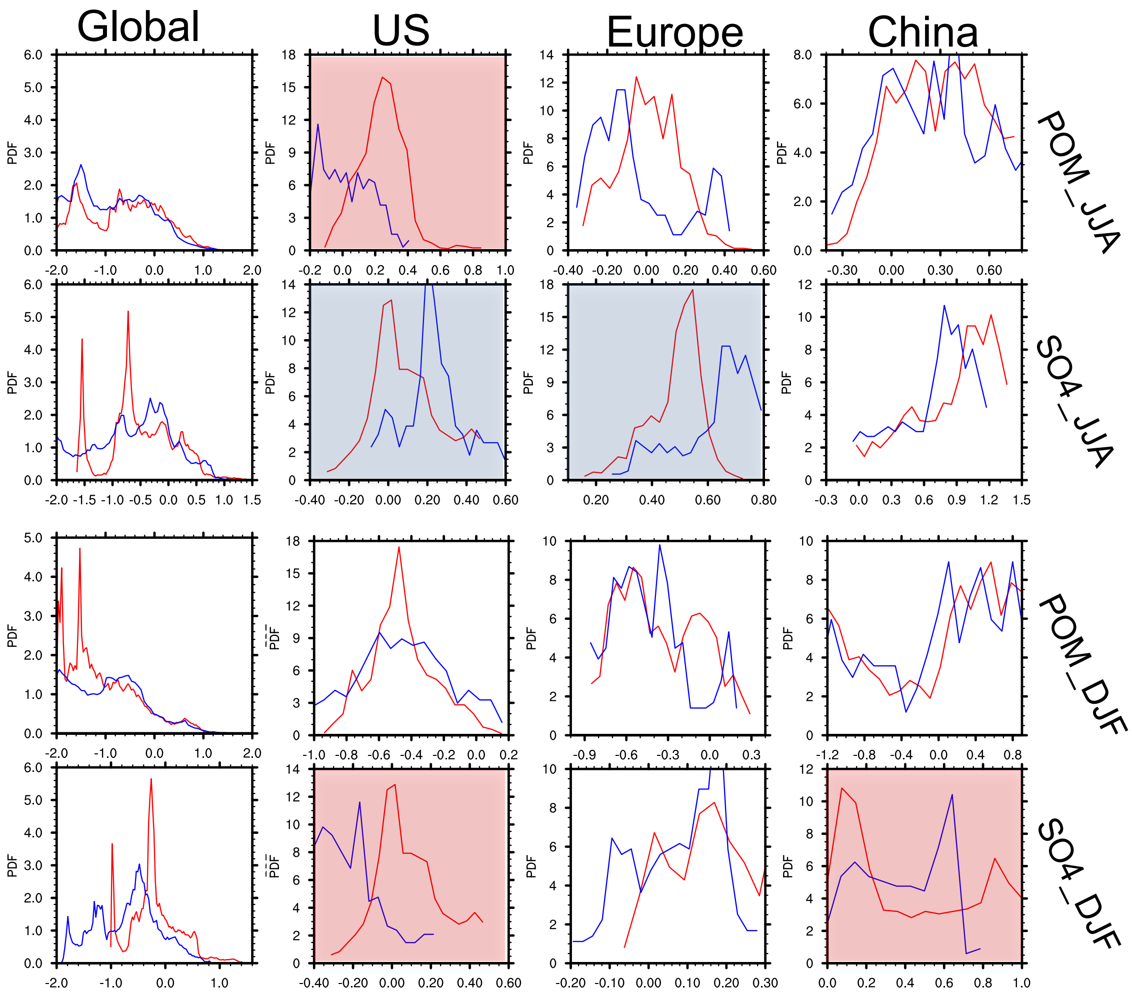


Figure S2. Comparison between simulation (blue) and MERRA2 reanalysis dataset (red) in the form of probability density function of the surface concentration of POM and $SO_4$ in logarithmic scale (X-axis: log(μg/m3)) over major regions (Global, US, Europe, and China). Both JJA and DJF results are shown in separate rows. Seasonal biases are identified in several regions (highlighted in color shaded panels), with blue panel indicating model overestimation, and red panel indicating model underestimation.

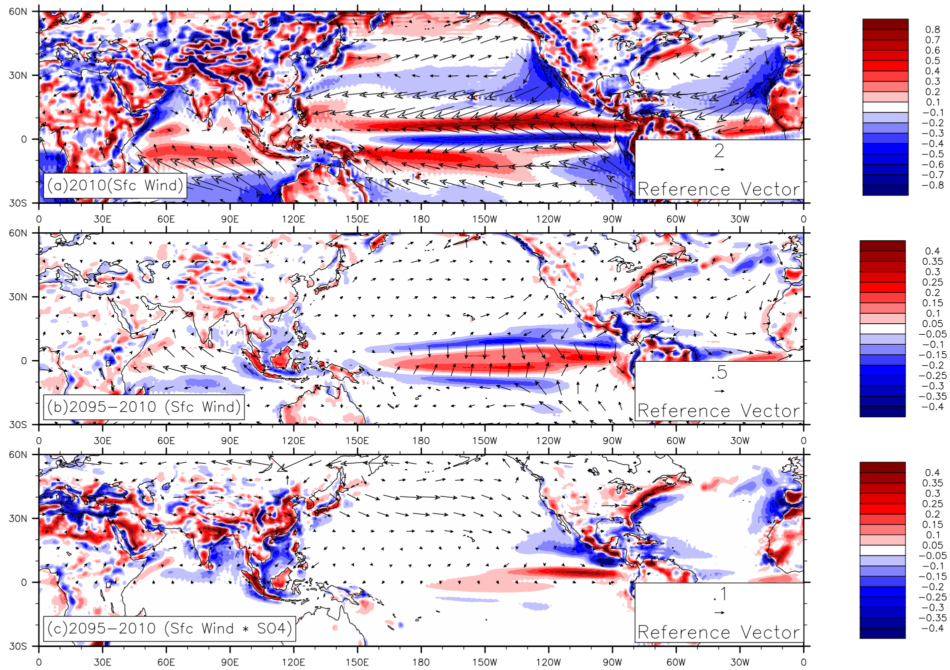


Figure S3. (a) Vector: present-day (2006-2015) surface wind (m/s). Color shading: the convergence of the wind field (unit: $10^{-5}$ 1/s). The positive (red) value in the convergence suggests inflow of air mass. (b) Same as (a) but for the changes at the end of the century (2090-2100) vs. present-day (2005-2015). (c) Same as (b) but for changes in surface wind weighted by $SO_4$ surface concentration as in Figure 4a, to approximate the mass flux of PM horizontal transport. Over the three main polluted regions focused in this study, there is a lack of consistent changes in the wind field (b) and in the PM transport (c). Over Mediterranean region and Eastern China, the changes flip in the sign and no consistent pattern emerges. While in the western US, changes in PM transport is in favor of PM increase (convergence, red) but the magnitude is small.