

## **Supporting Information for Christiansen et al.**

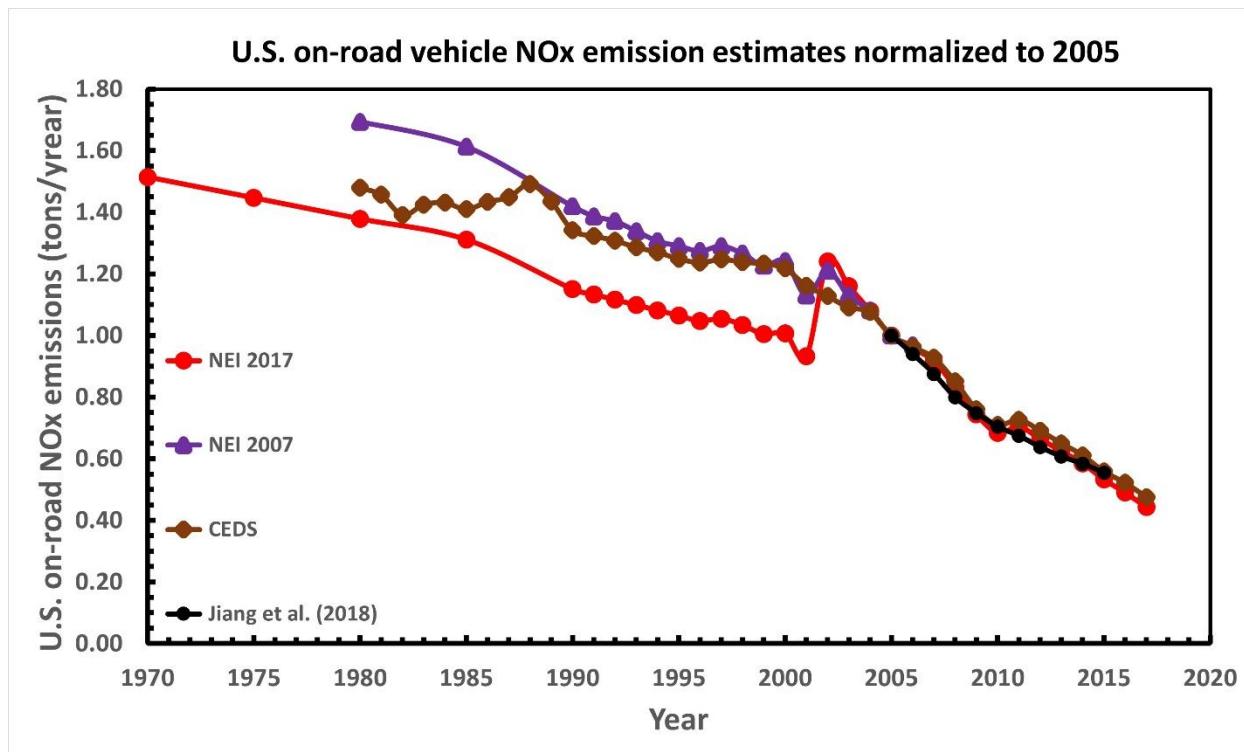
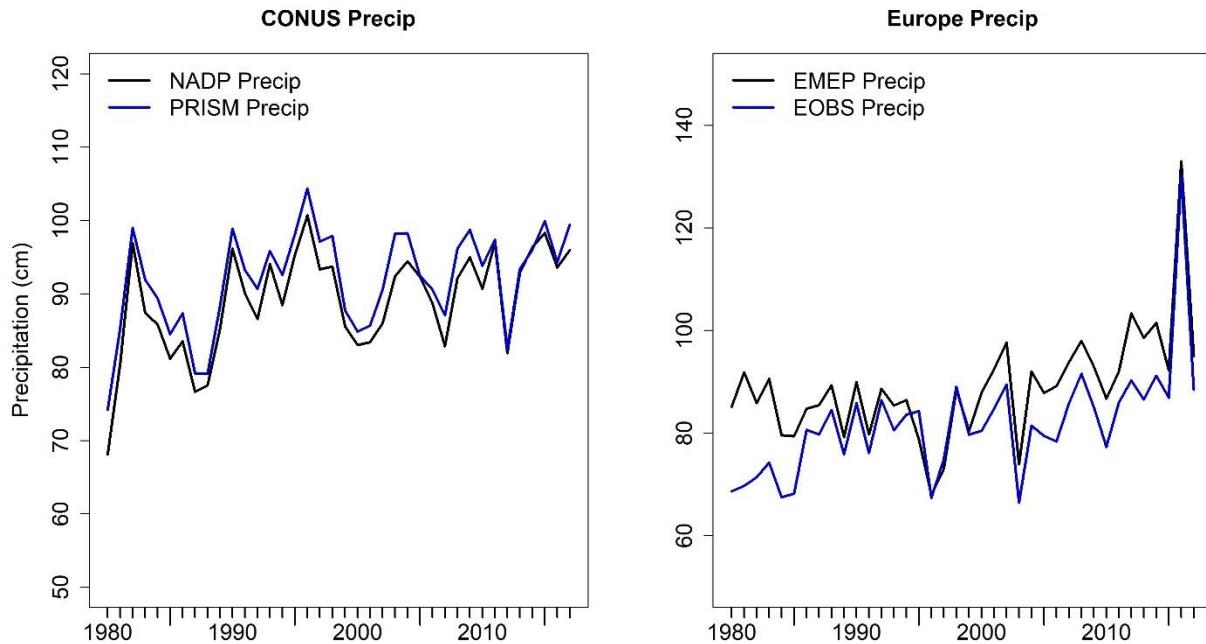


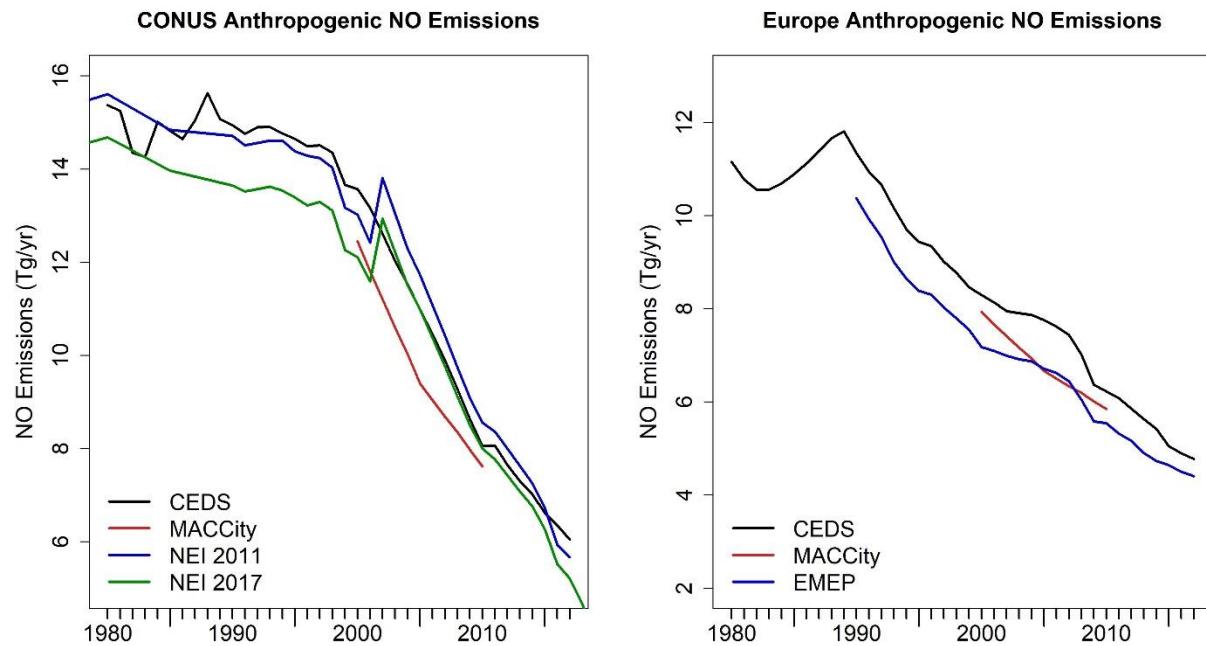
Figure S1. Comparison of US on-road mobile NO<sub>x</sub> emissions estimates from the NEI, the Community Emissions Data System (CEDS) inventory, and satellite measurements normalized to 2005 values. Many of these datasets are the same as in Figure 1, but we only include here datasets with estimates for the year 2005. Note that the jump in the NEI 2017 between 2001 and 2002 is due to a change in the emissions model used for mobile sources (from MOBILE6.2 to MOVES 2010), not from real increases in NO<sub>x</sub> emissions from mobile sources. Values from the NEI and CEDS inventories are taken directly from the on-road vehicle emissions sectors. Values from Jiang et al. (2018) were taken directly from that analysis.



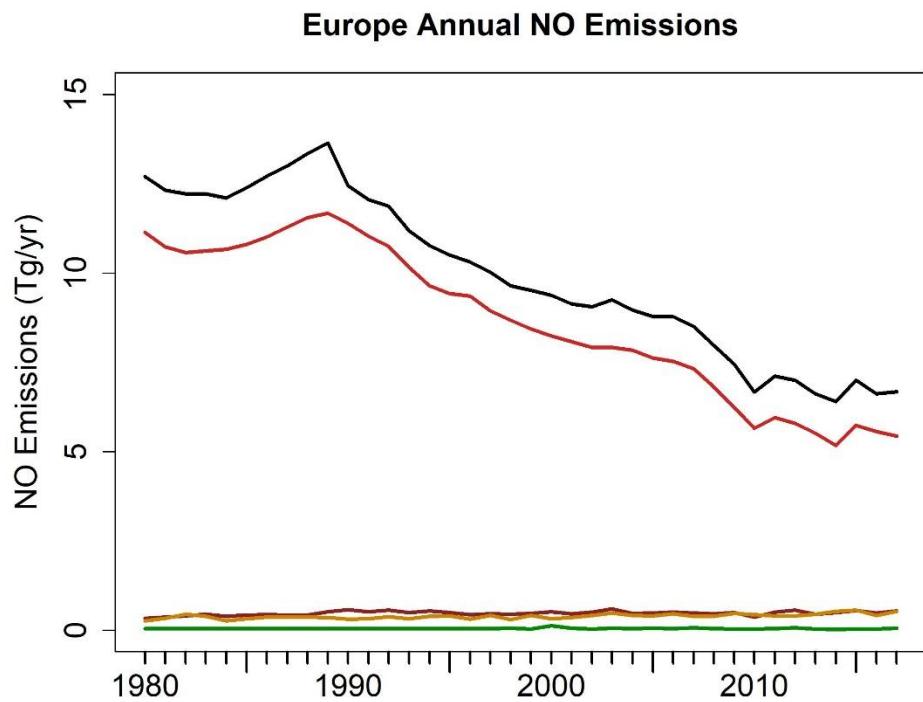
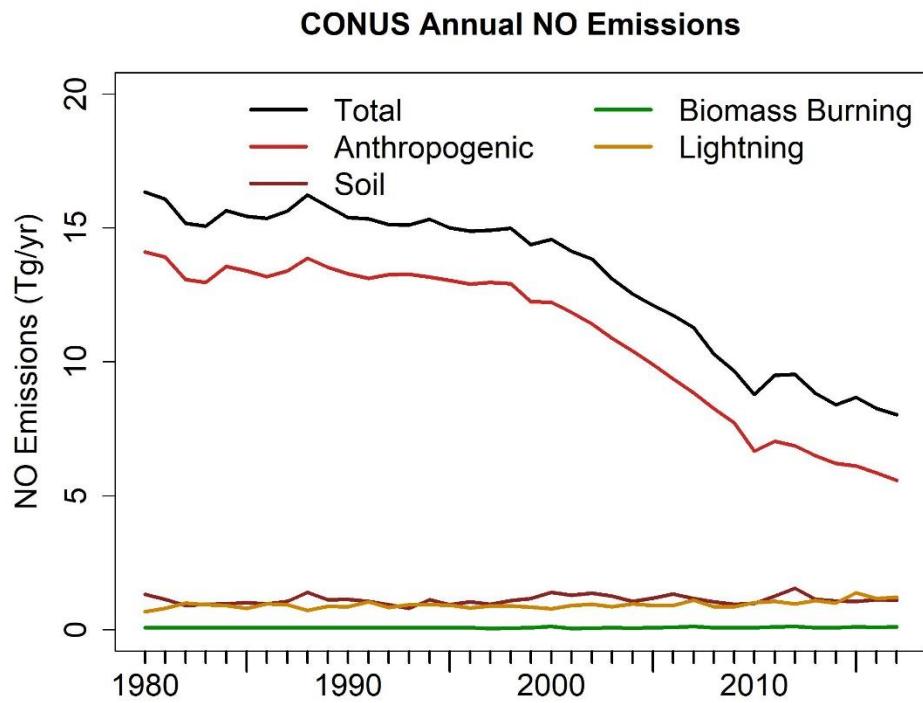
Figure S2. Locations of surface ozone monitoring sites (left, TOAR) and vertical profile ozone measurements (right, ozonesondes).



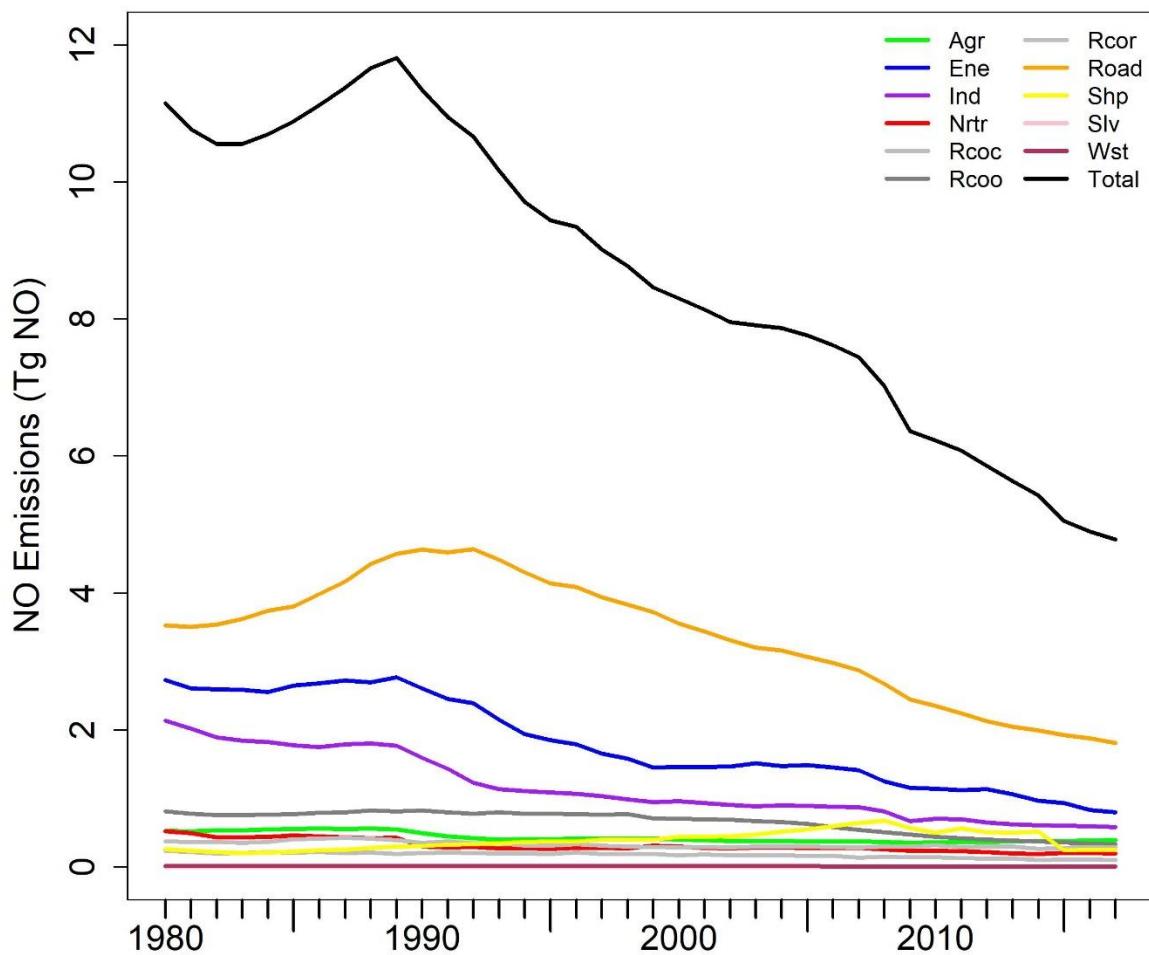
**Figure S3.** Annual average observed trends in precipitation over the CONUS and Europe. High-resolution precipitation data was taken from PRISM for the CONUS and E-OBS for Europe, both of which are shown with blue lines. Precipitation measured by the nitrate wet deposition networks (NADP and EMEP) is shown in the black lines.



**Figure S4.** Total anthropogenic NO<sub>x</sub> emissions estimates from various global and regional anthropogenic inventories over the CONUS and Europe. CEDS and MACCity are shown in the black and red lines, respectively. The EPA's National Emissions Inventory is shown in the blue (NEI 2011) and green lines (NEI 2017) in panel (a). The EMEP inventory over Europe is shown in the blue line in panel (b).



**Figure S5.** Magnitude and trends of NO<sub>x</sub> emissions sectors over the CONUS and Europe in the base simulation using CEDS as anthropogenic emissions.



**Figure S6. Individual anthropogenic sector contributions to total CEDS anthropogenic NO emissions over Europe. The primary contribution is road emissions. The sector contributions are agriculture (Agr), energy production (Ene), industry (Ind), non-road transportation (Nrtr), commercial (Rcoc), other (Rcoo), residential (Rcor), road transportation (Road), shipping (Shp), solvents (Slv), and waste (Wst).**

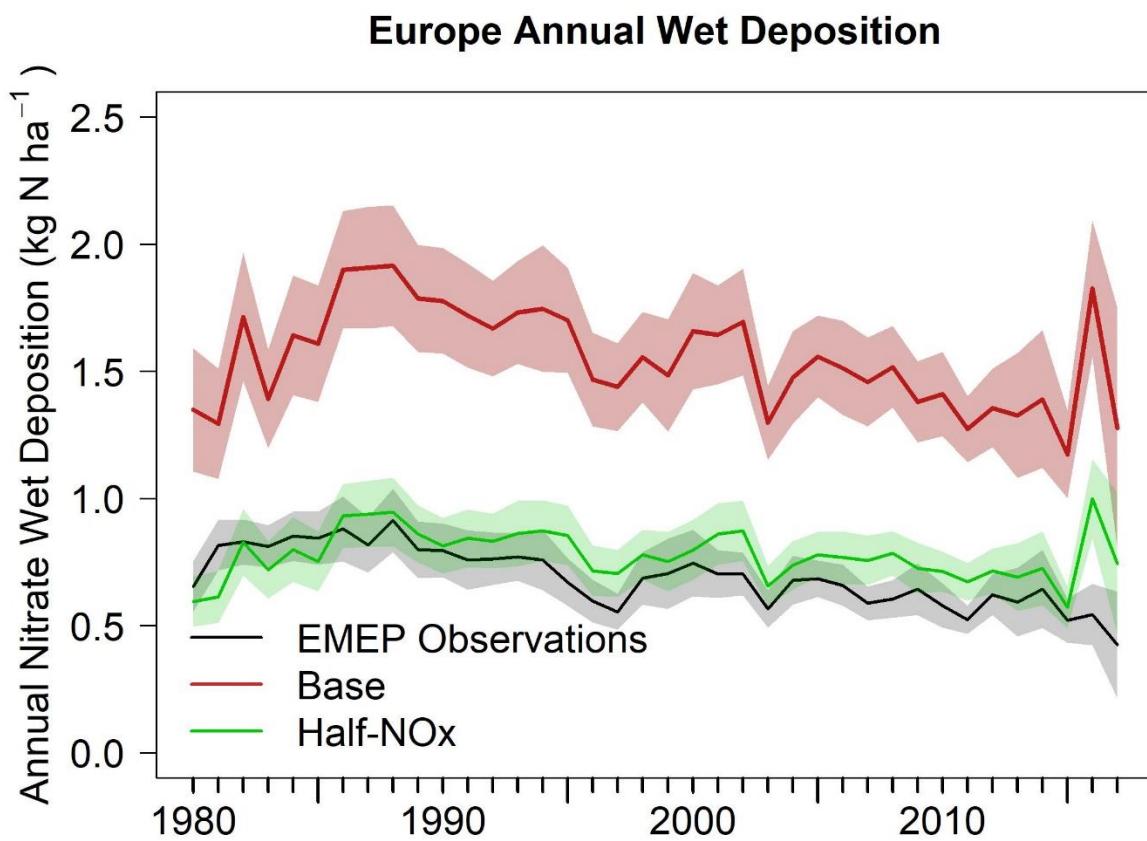


Figure S7. Annual NWD deposition (kg N/ha) over Europe in the base simulation (red) and the half NO<sub>x</sub> sensitivity (green). Observations from the EMEP network are shown in black.