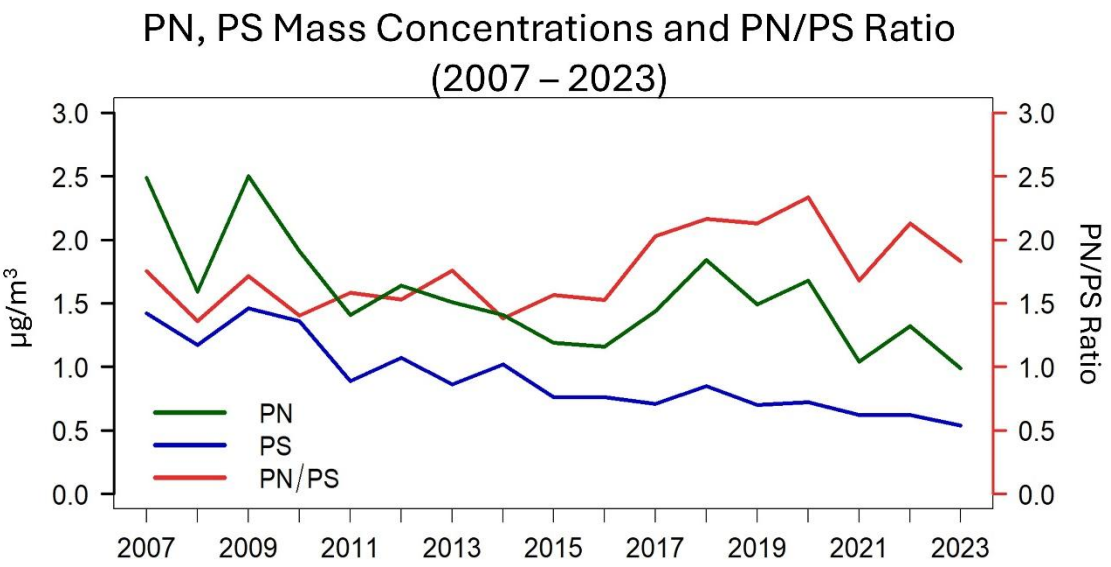

Figure S1: Wintertime PN mass concentrations (green), PS mass concentrations (blue), and PN/PS ratios (in red) over the MWUS from 2007 to 2023.

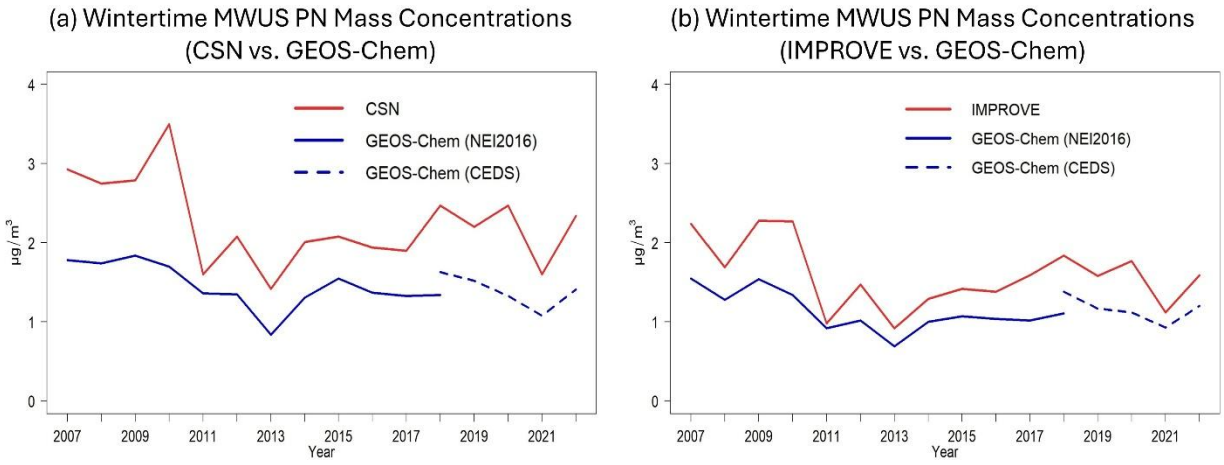

Figure S2: The comparisons in wintertime PN trends between ground monitoring observations, (a) CSN and (b) IMPROVE, and the Base simulation in GEOS-Chem. Solid red lines represent the wintertime PN mass concentrations from ground monitoring observations (IMPROVE and CSN). Solid blue lines represent the wintertime PN mass concentrations from GEOS-Chem using NEI2016 emissions inventory. Dashed blue lines represent the wintertime PN mass concentrations from GEOS-Chem using CEDS emissions inventory.

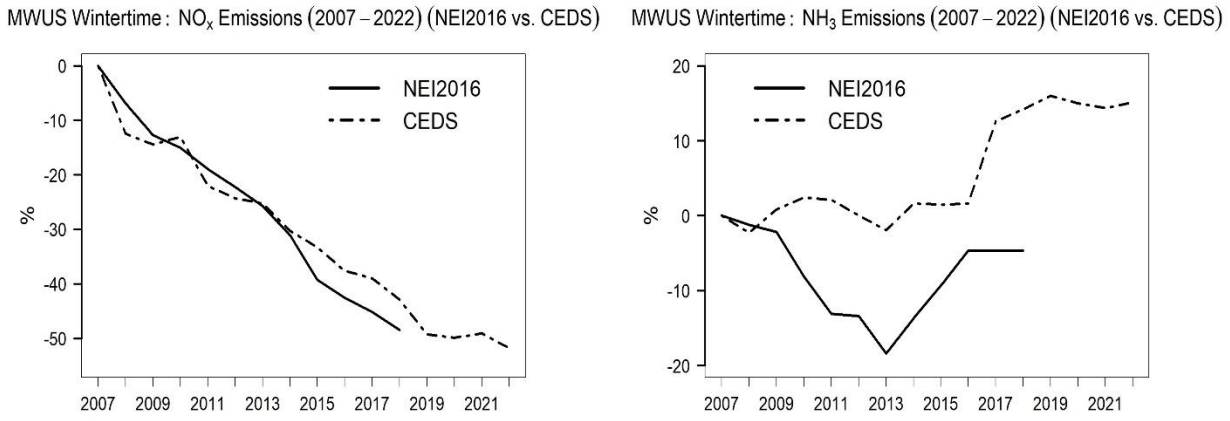

Figure S3: Wintertime NO_x emissions (left) and NH_3 emissions (right) using NEI2016 emissions inventory (solid lines) and CEDS emissions inventory (dashed lines and points) over the MWUS (2007–2022).

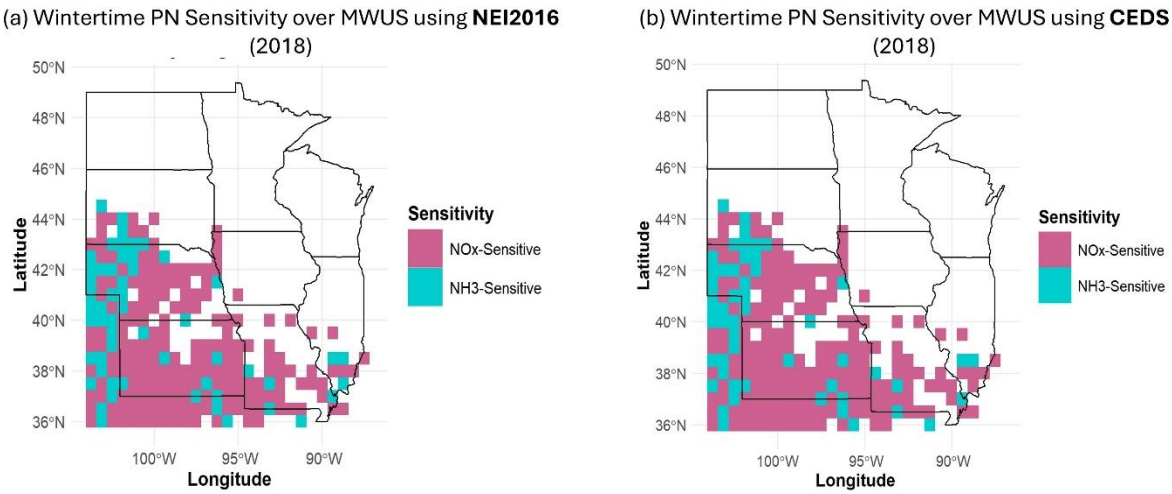

Figure S4: Wintertime PN sensitivity in winter 2018 (i.e., Jan 2019) using NEI2016 emissions inventory (left) and CEDS emissions inventory (right). Pink pixels indicate PN formation is sensitive to NO_X emissions (i.e., NOx-sensitive), and blue pixels indicate PN formation is sensitive to NH_3 emissions (i.e., NH_3-sensitive).

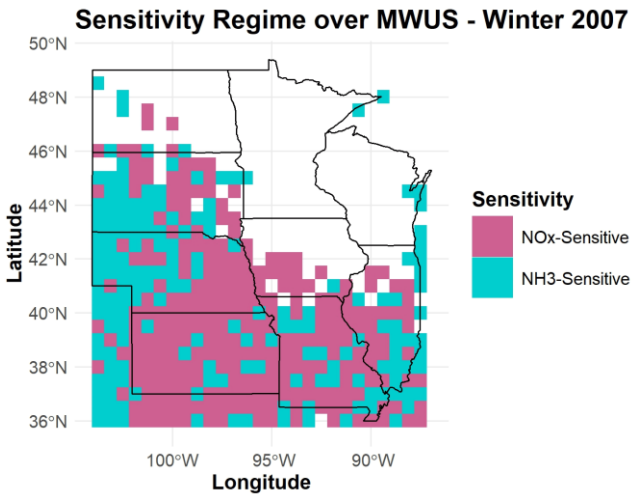

Figure S5: The distribution of wintertime PN sensitivity regime over MWUS (2007 – 2023). Pink pixels indicate PN formation is sensitive to NO_X emissions (i.e., NOx-sensitive), and blue pixels indicate PN formation is sensitive to NH_3 emissions (i.e., NH_3-sensitive).

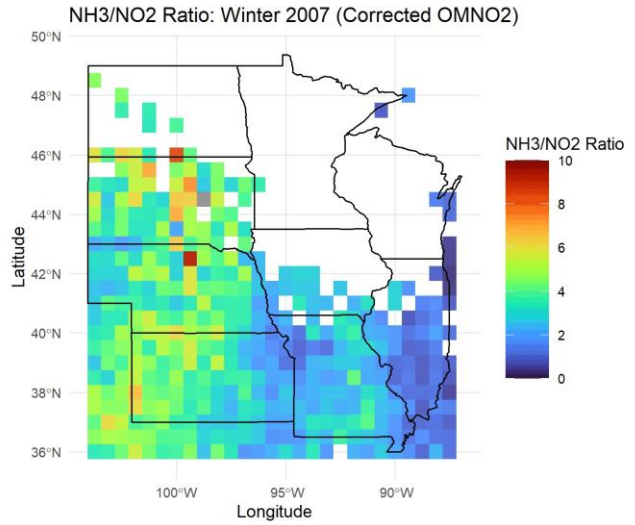

Figure S6: The distribution of wintertime satellite tropospheric column NH_3 /NO_2 ratios over MWUS (2007 – 2023). Grey pixels indicate NH_3/NO_2 ratios > 10.

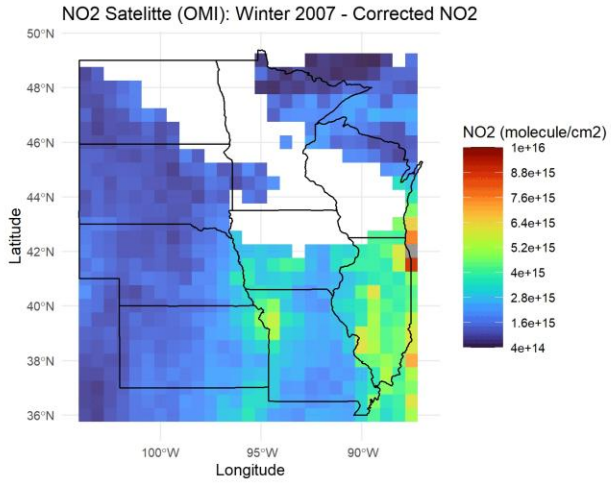

Figure S7: The distribution of wintertime NO_2 column density (2007 – 2023) over MWUS. Warmer color indicates higher NO_2 column densities, and colder color indicates lower NO_2 column densities. Grey pixels indicate NO_2 column density > 1×10^{16} molecules cm^{-2}.

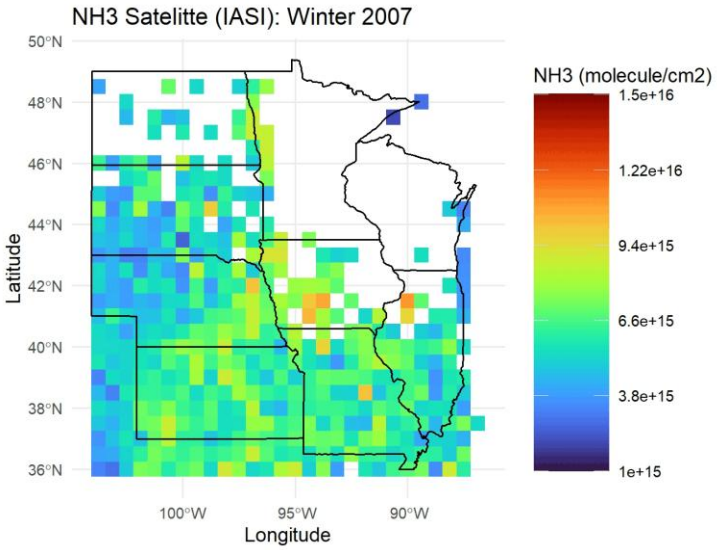

Figure S8: The distribution of wintertime NH_3 column density from IASI (2007–2023) over MWUS. Warmer color indicates higher NH_3 column densities, and colder color indicates lower NH_3 column densities. Grey pixels indicate NH_3 column density > 1.5×10^{16} molecules cm^{-2}.

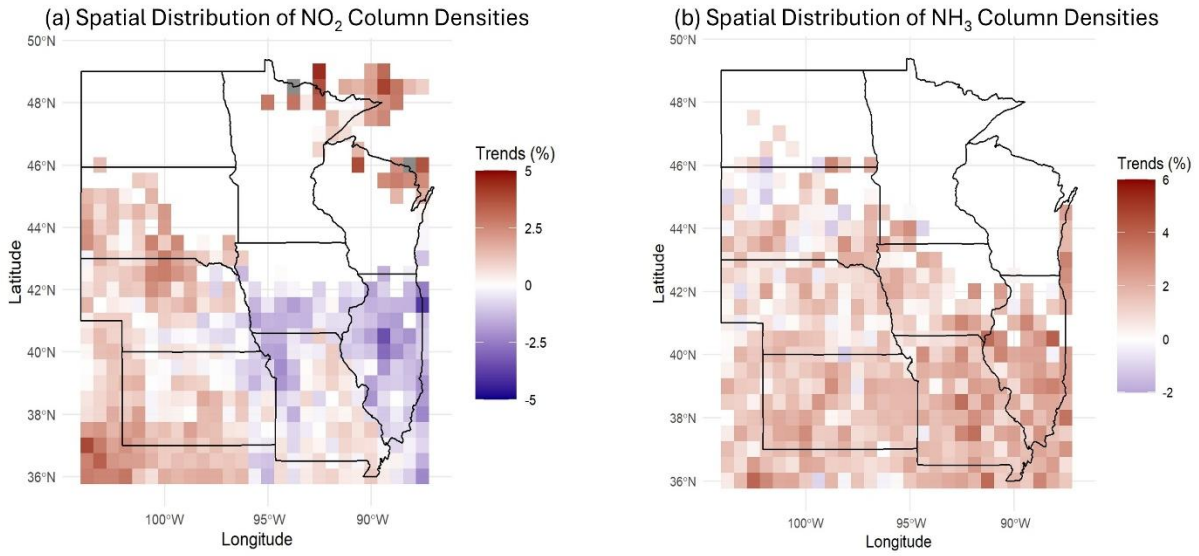

Figure S9: The spatial trends of wintertime (a) NO_2 and (b) NH_3 column density over MWUS (2007–2023). The increasing trend of NO_2 and NH_3 column densities is shown in red, and the decreasing trend in NO_2 and NH_3 column densities is shown in blue. Grey pixels indicate an increase > 5% in NO_2 column density for panel (a) only.

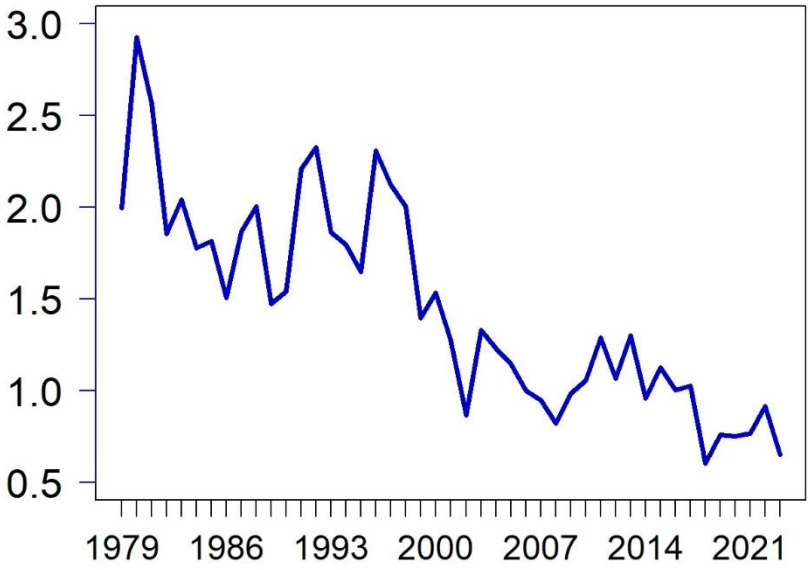

Figure S10: Wintertime nitrate wet deposition (NWD) trends over the MWUS from 1979 to 2023 using the National Trends Network.

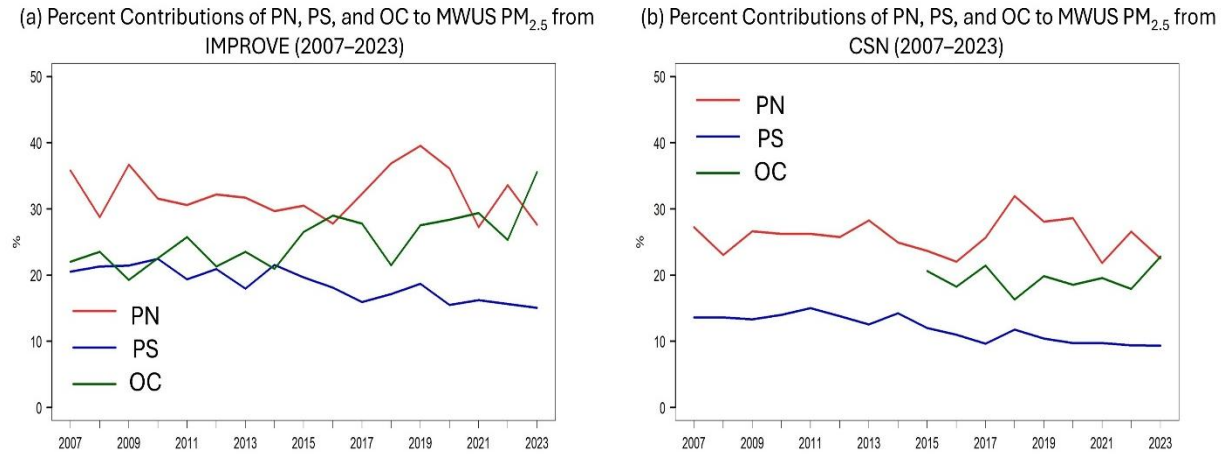

Figure S11: The contributions of PN (red), PS (blue), and total organic carbon (OC) (green) to total $PM_{2.5}$ trends over the MWUS (2007 – 2023) using (a) IMPROVE and (b) CSN ground monitoring observations.

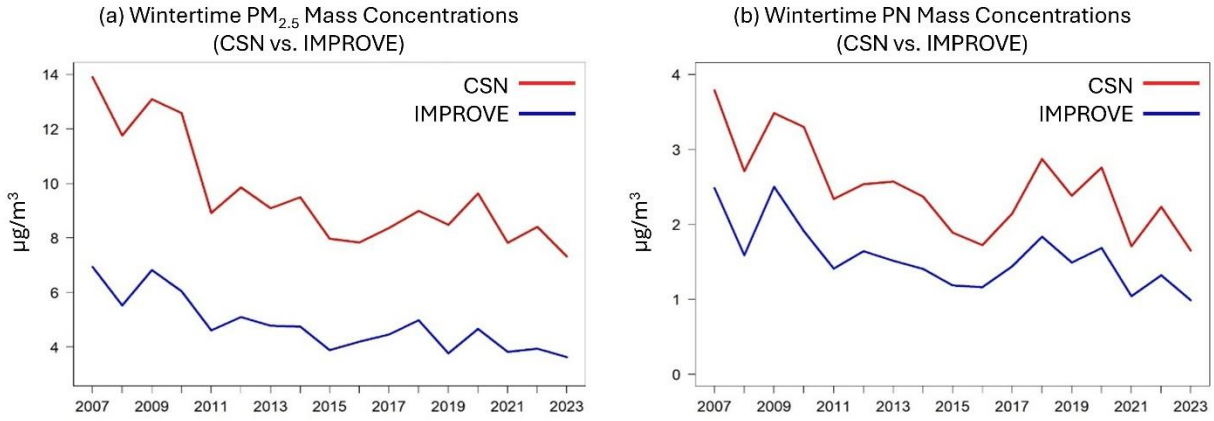

Figure S12: Wintertime $PM_{2.5}$ and PN trends over urban and rural areas in MWUS (2007 – 2023) using IMPROVE (blue) and CSN (red) ground monitoring observations.