| SMOKE Processing Category	Emission Source
Biogenic	By-products of microbial nitrification and denitrification occurring in soil
Area	Off-road vehicles Residential combustion: *anthracite coal, bituminous coal, distillate oil, residual oil, natural gas, liquified petroleum gas, and wood* Industrial processes: *chemical manufacturing, food, and kindred products, metal production, mineral processes, petroleum refining, wood products, construction, machinery, mining, and quarrying, etc.* Agriculture production: *crops, fertilizer application, livestock, animal waste, etc.* Solvent utilization Storage and transport Waste disposal, treatment, and recovery Forest wildfires Road dust and fugitive dust
Mobile	On-road vehicles
Point	Electric generating units (EGU) Commercial combustion and industrial combustion (non-EGU) Fugitive dust

Table S1: Categorization of emission sources
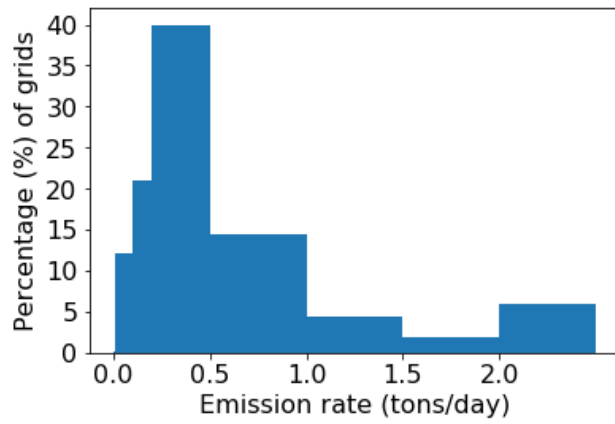
Figure S1: The histogram of total NO_x emission in the Midwest between April and June in tons/day.
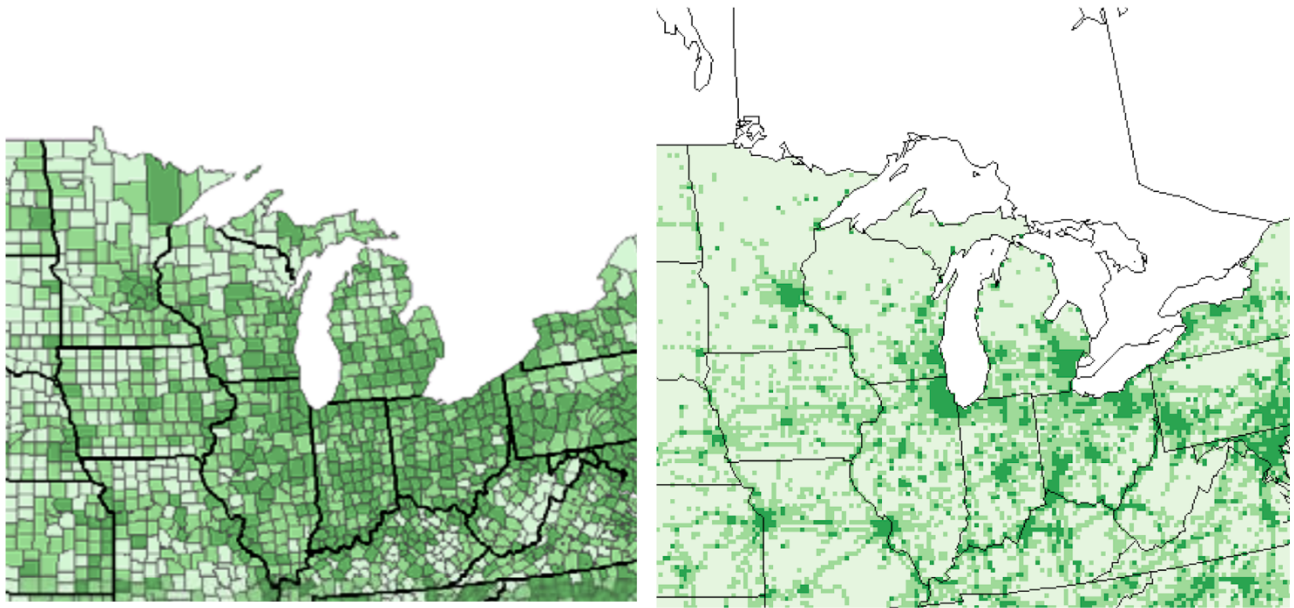
NEI county-level   **SMOKE grid-level**

tons/sq. mi
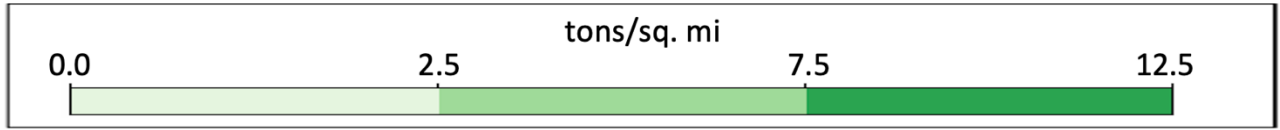
| 0.0 | 2.5 | 7.5 | 12.5 |

Figure S2: The geographical distribution of 2002 NO_x emission density estimated by NEI over each county (left) and simulated by SMOKE over each grid (right), throughout the Midwest.
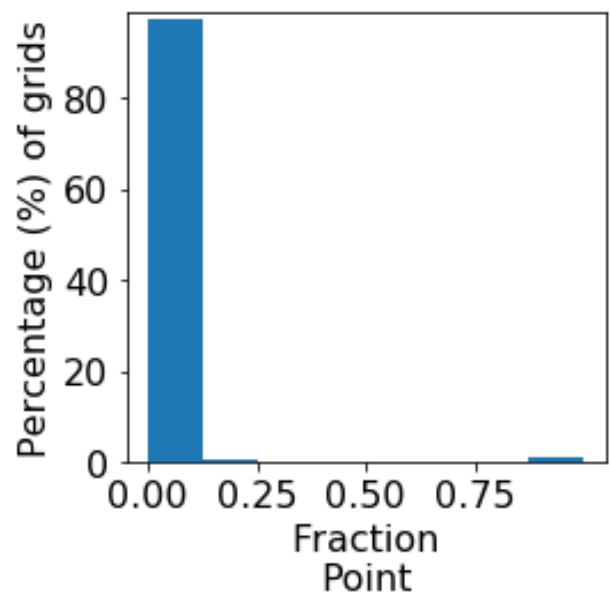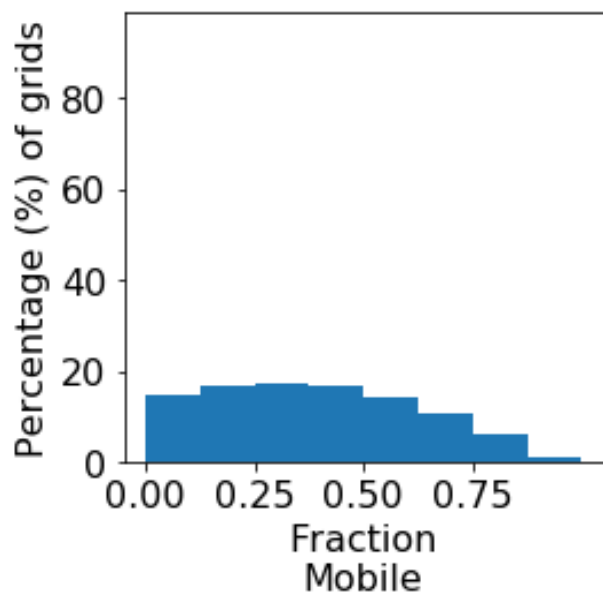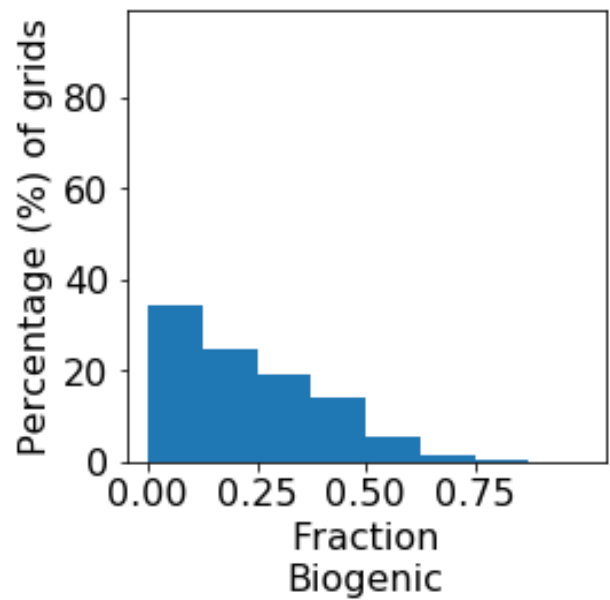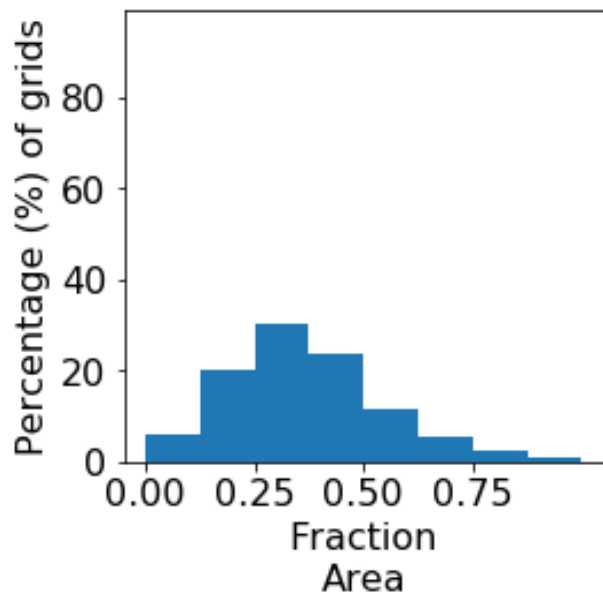
Figure S3: The histogram of the fraction of NO_x emission from each SMOKE processing category (area, biogenic, mobile, point) over each grid throughout the Midwest between April and June based on NEI-2002.
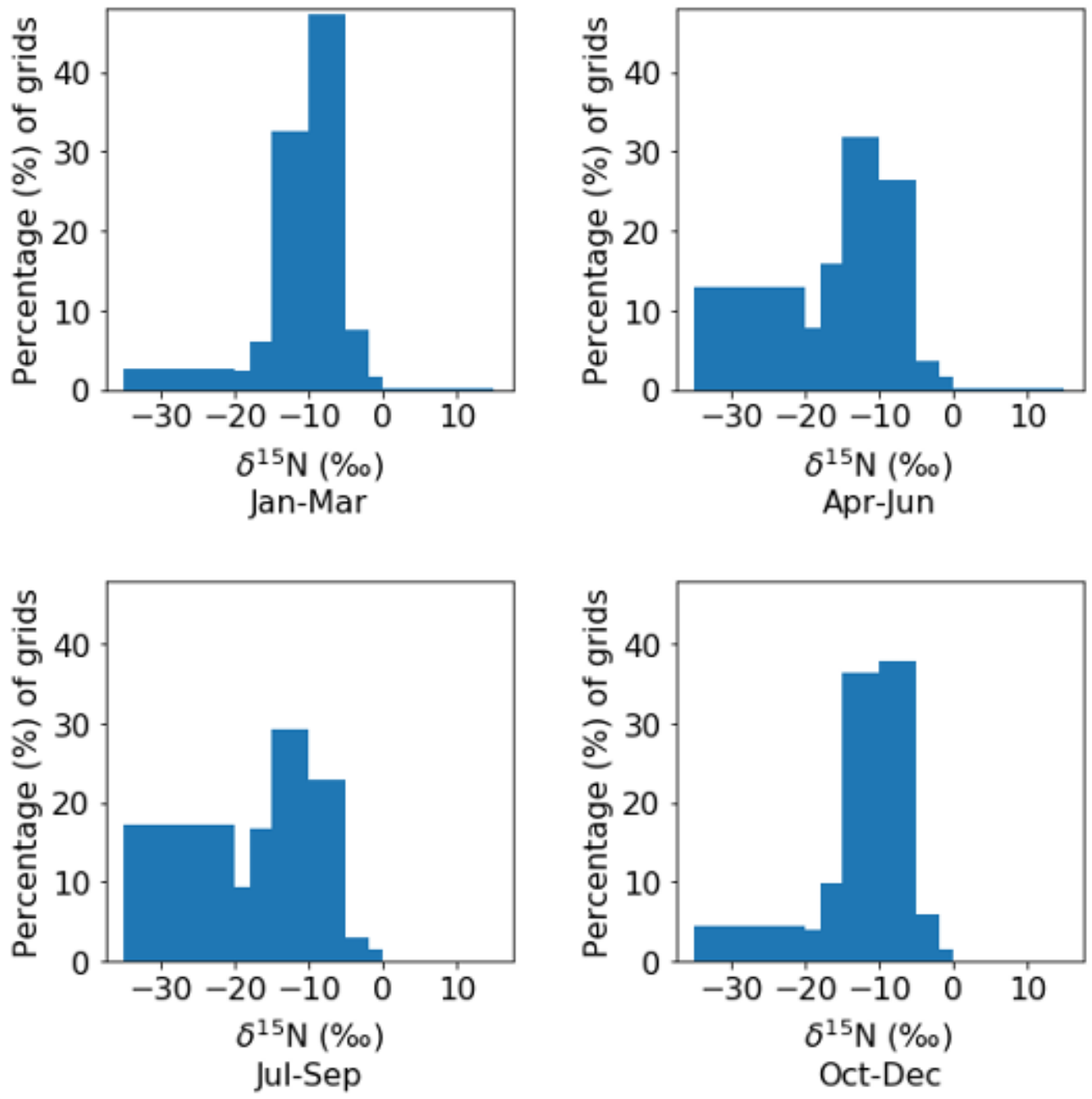
Figure S4: The histogram of the $\delta^{15}N$ of total NO_x emissions in each season (Winter: Jan-Mar; Spring: Apr-Jun; Summer: Jul-Sep; Fall: Oct-Dec) in per mil (‰) over the 12-km grids throughout the Midwest simulated by SMOKE, based on NEI-2002.
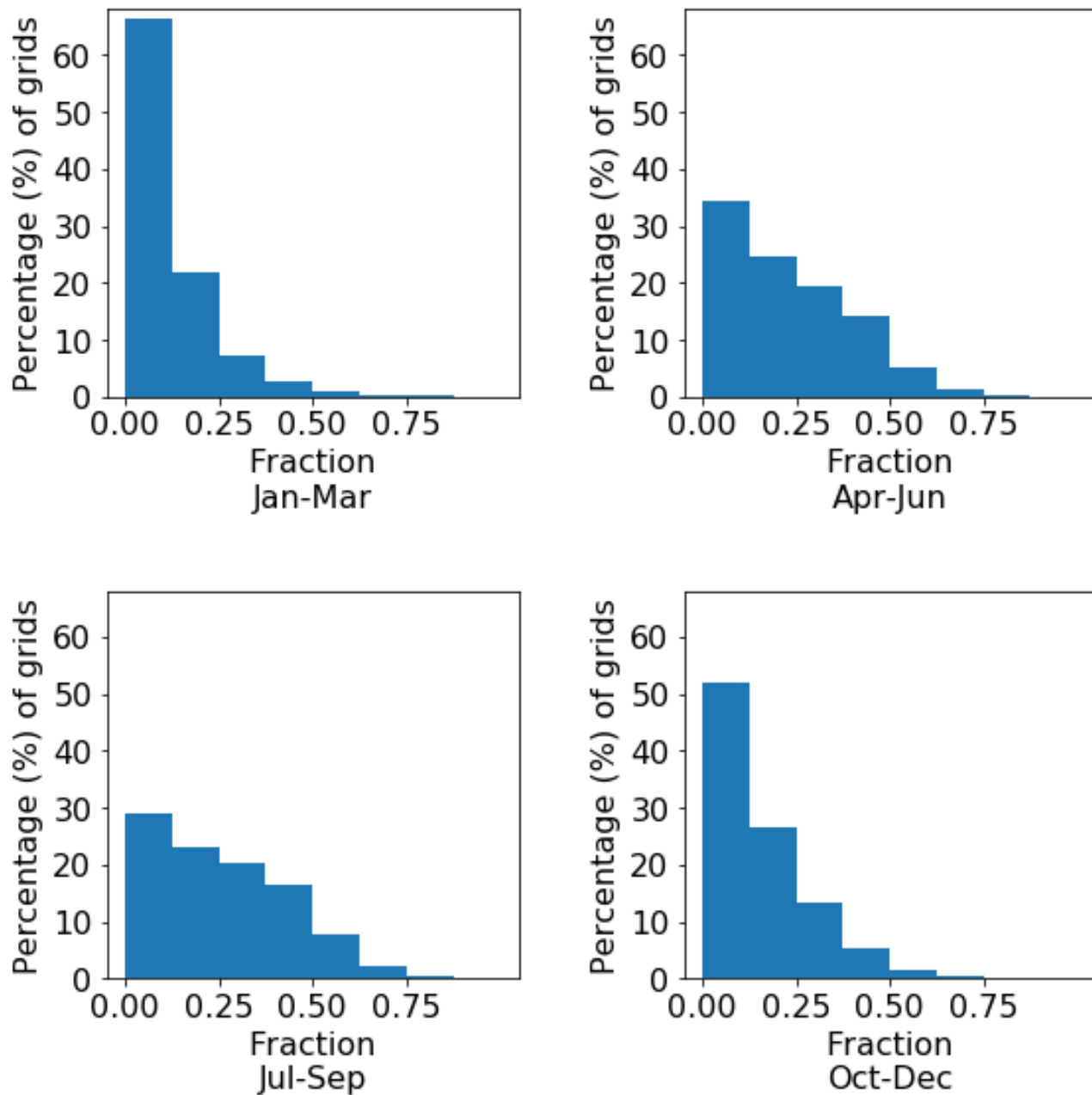
Figure S5: The histogram of the fraction of NO_x emission from biogenic sources over each grid in each season (Winter: Jan-Mar; Spring: Apr-Jun; Summer: Jul-Sep; Fall: Oct-Dec) throughout the Midwest simulated by SMOKE, based on NEI-2002.

Figure S6: The geographical distribution of the fraction of NO_x emission from biogenic sources over each grid in each season (Winter: Jan-Mar; Spring: Apr-Jun; Summer: Jul-Sep; Fall: Oct-Dec) throughout the Midwest simulated by SMOKE, based on NEI-2016.
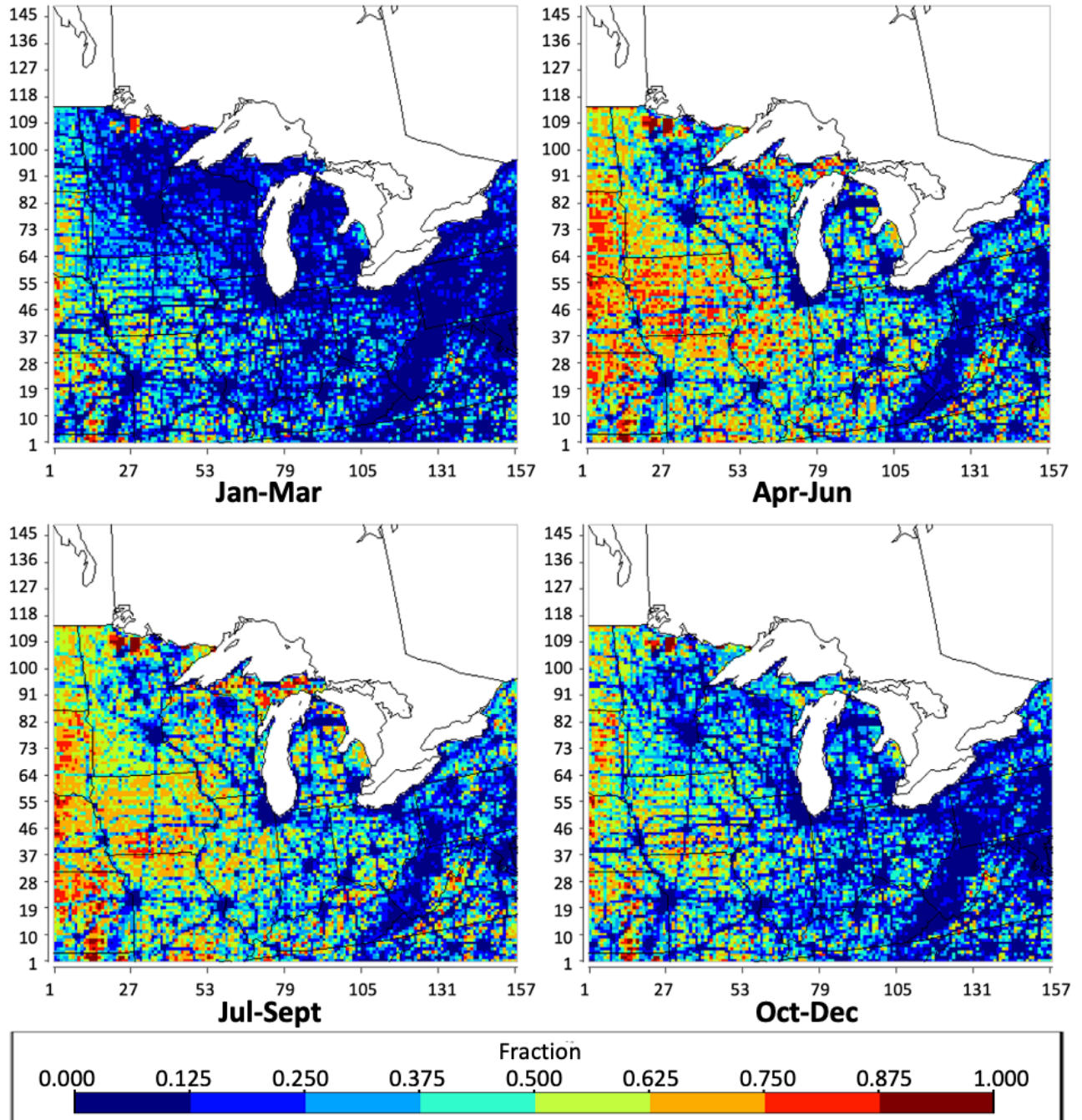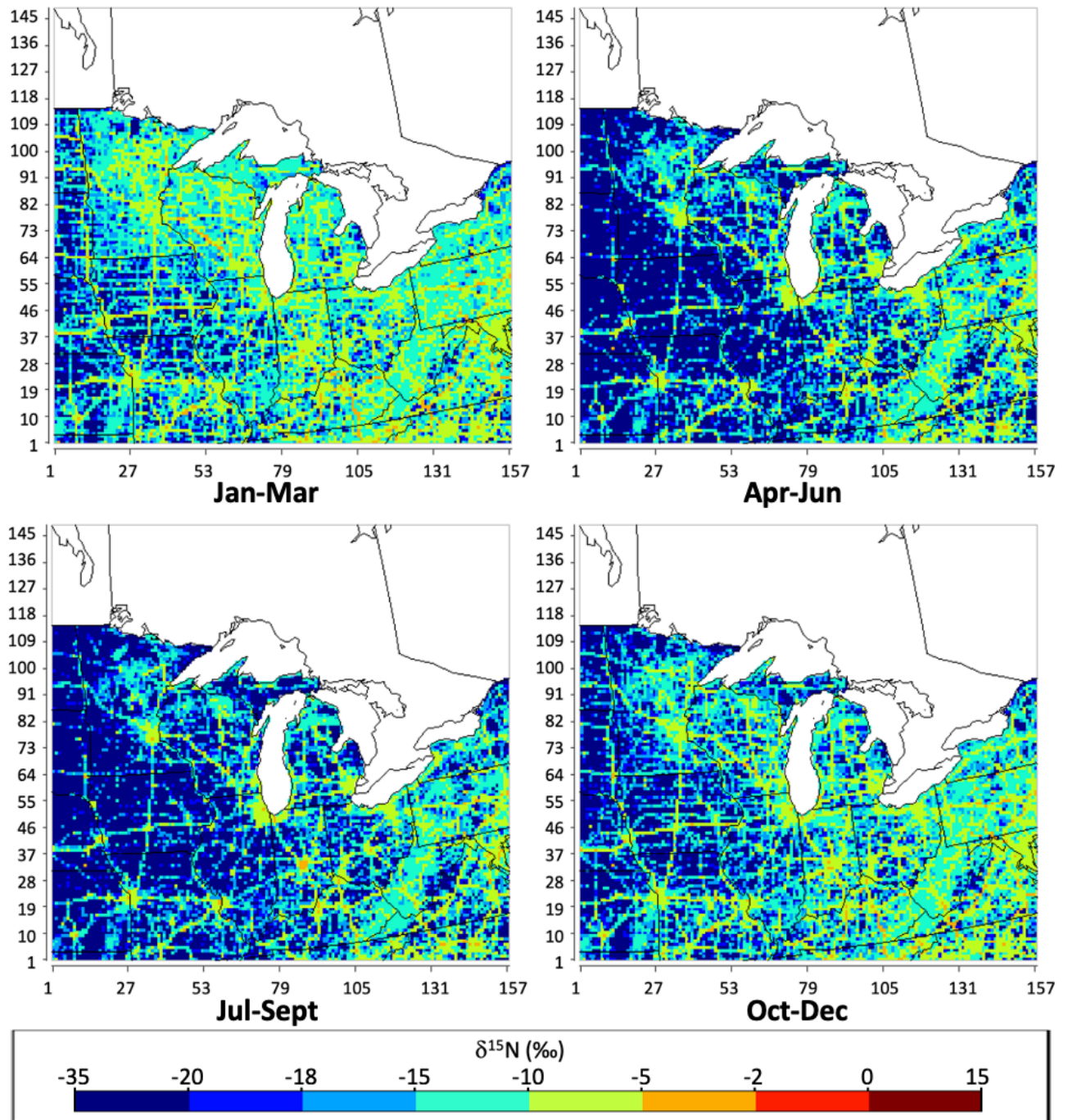
Figure S7: The geographical distribution of the $\delta^{15}N$ value of total NO_x emissions in each season (Winter: Jan-Mar; Spring: Apr-Jun; Summer: Jul-Sep; Fall: Oct-Dec) in per mil (‰) throughout the Midwest simulated by SMOKE, based on NEI-2016.

Figure S8: Locations of 82 NADP sites within Midwest © Google Maps
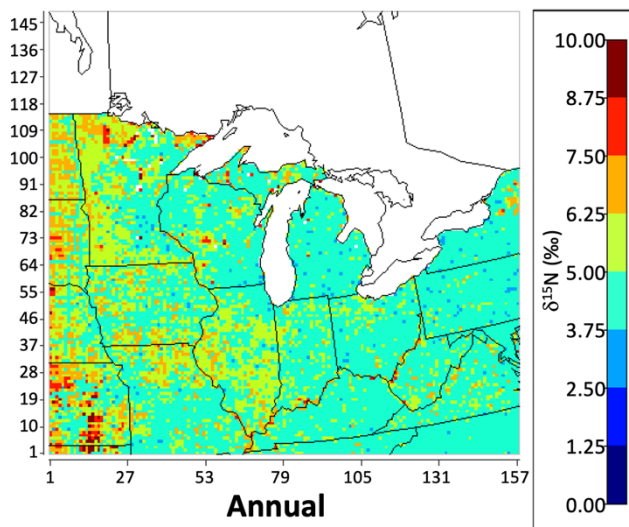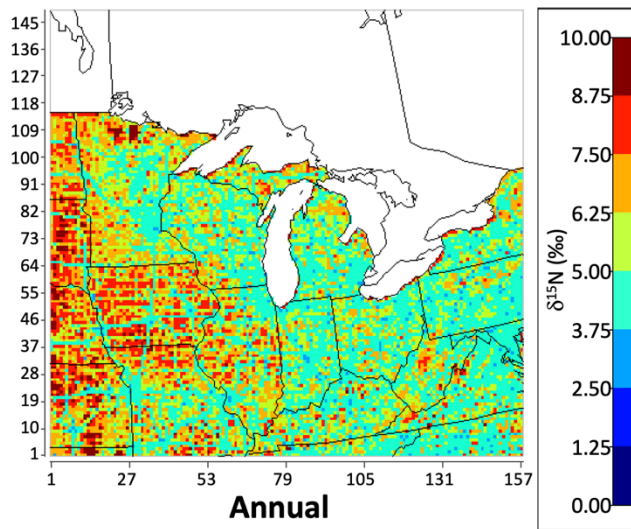
Figure S9: The uncertainties of δ^{15}N values based on NEI-2002 are presented by color in each grid. The warmer the color, the higher uncertainties of δ^{15}N values.

Figure S10: The uncertainties of δ¹⁵N values based on NEI-2016 are presented by color in each grid. The warmer the color, the higher uncertainties of δ¹⁵N values.