Parse, simulation, and prediction of NOx emission across the Midwestern United States

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November 26, 2022

Abstract

Accurately constraining N emissions in space and time has been a challenge for atmospheric scientists. It has been suggested that 15N isotopes may be a way of tracking N emission sources across various spatial and temporal scales. However, the complexity of multiple N sources that can quickly change in intensity has made this a difficult problem. We have used a SMOKE emission model to parse NOx emission across the Midwestern United States for a one-year simulation. An isotope mass balance methods was used to assign 15N values to road, non-road, point, and area sources. The SMOKE emissions and isotope mass balance were then combined to predict the 15N of NOx emissions (Figure 1). This δ15N of NOx emissions model was then incorporated into CMAQ to assess the role of transport and chemistry would impact the 15N value of NOx due to mixing and removal processes. The predicted 15N value of NOx was compared to those in recent measurements of NOx and atmospheric nitrate.

Simulation of $\delta^{15}N$ and partition of NO_x emission across the Midwestern United States

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INTRODUCTION

1. Nitrogen oxides (NO_x) are important trace gases that affect atmospheric chemistry, air quality, and climate.
2. The importance of the sources of NO_x, both natural and anthropological are uncertain.
3. The nitrogen stable isotope ($\delta^{15}N$) has been proposed as a regional indicator for the partition NO_x emission sources.
4. The measurement of $\delta^{15}N$-NO_x is limited.
5. The emissions from neighborhoods potentially affects the fraction of different NO_x emission sources, due to atmospheric transport. Thus $\delta^{15}N$-NOx values will be changed.
6. Atmospheric mixing of 12, 36, 60, 108, 204 km around the site were assumed to represent the different levels of effects on $\delta^{15}N$-NOx values from neighborhood emissions.

METHOD

Determine $\delta^{15}N$ values of different categories

$$(\delta^{15}N - NO_x)_{total} = \sum f_{categorical\,(i)} \times \delta^{15}N_{categorical\,(i)}$$
$$(\delta^{15}N - NO_x)_{categorical} = \sum f_{source\,(i)} \times \delta^{15}N_{source\,(i)}$$

$(\delta^{15}N - NO_x)_{point}$: based on power plant type

f: county-level fraction of source
t: zip-code-based commute time

Fractions of sources

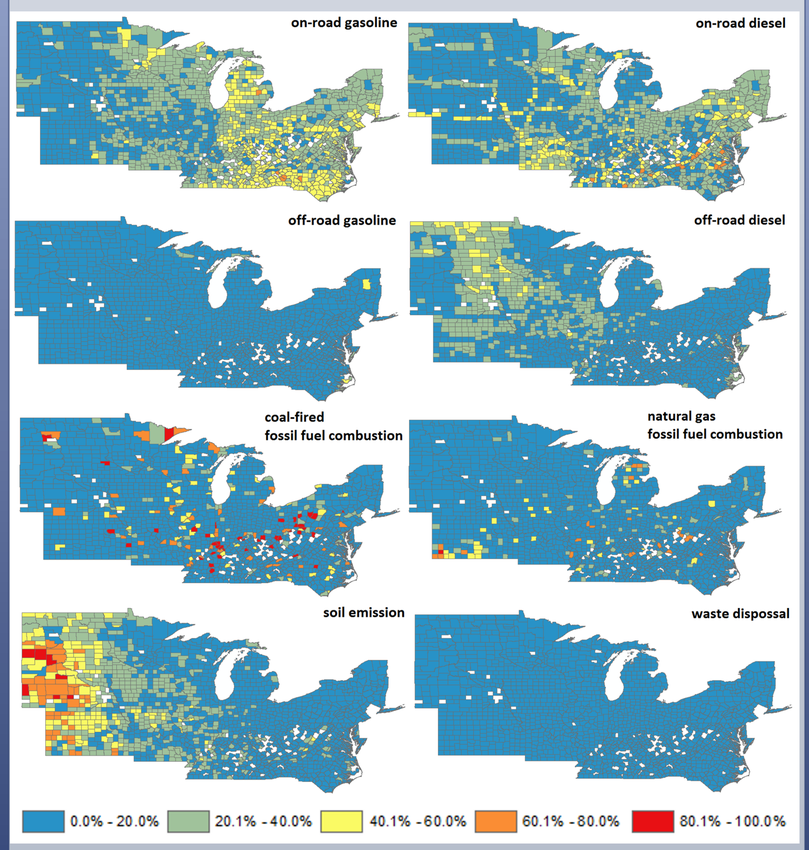

Figure 1

Method

Source categories

NOx Category	NOx Source		δ^{15}N-NOx(‰)
Area	Waste		-18.8 (Felix & Elliott, 2014)
	Off-Road	Gasoline	-11.5 (Walters et al., 2015)
		Diesel	-10.5 (Walters et al., 2015)
Biogenic	Soil		-34.3 (Felix & Elliott, 2014)
Mobile	On-Road Gasoline		ln (commute time + 0.455) * 3.02 – 12.35 (Walters et al., 2015)
	On-Road Diesel		-2.5 (Walters et al., 2015)
Point	Coal-fired Fossil Fuel Combustion		15 (Felix et al., 2012)
	Natural Gas Fossil Fuel Combustion		-16.5 (Walters et al., 2015)

Table 1

RESULTS

$\delta^{15}N$-NO_x over Midwest

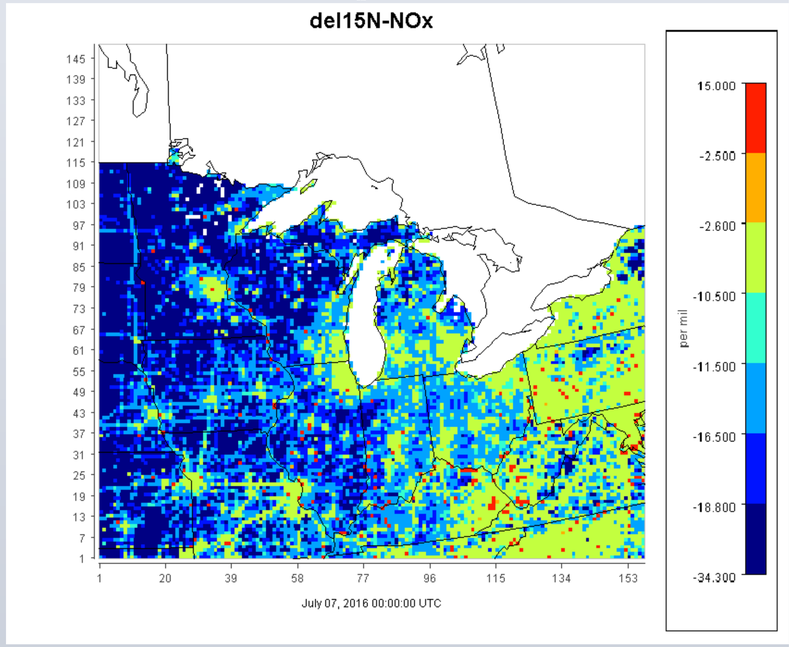

Figure 2

$\delta^{15}N$-NO_x around West Lafayette

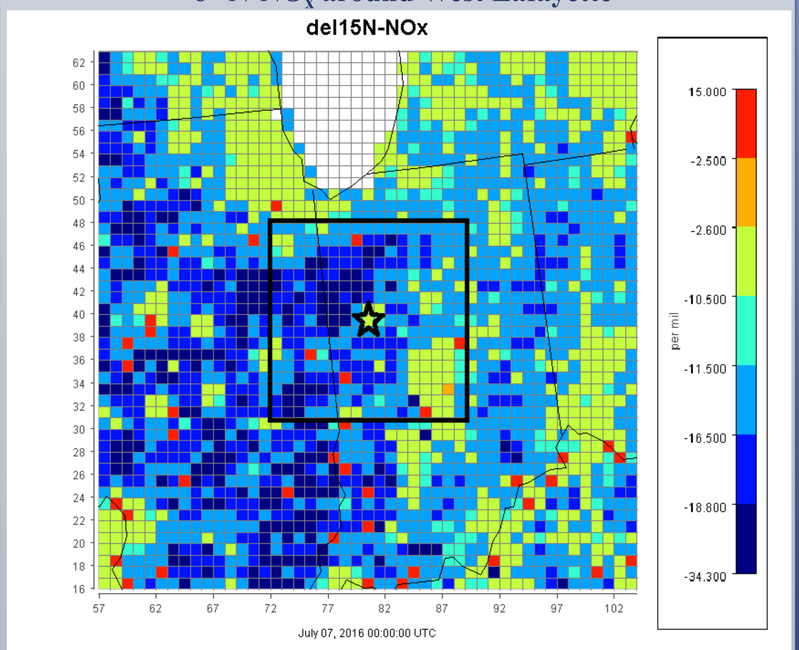

Figure 3

RESULTS

Estimation of $\delta^{15}N$-NO_x at West Lafayette

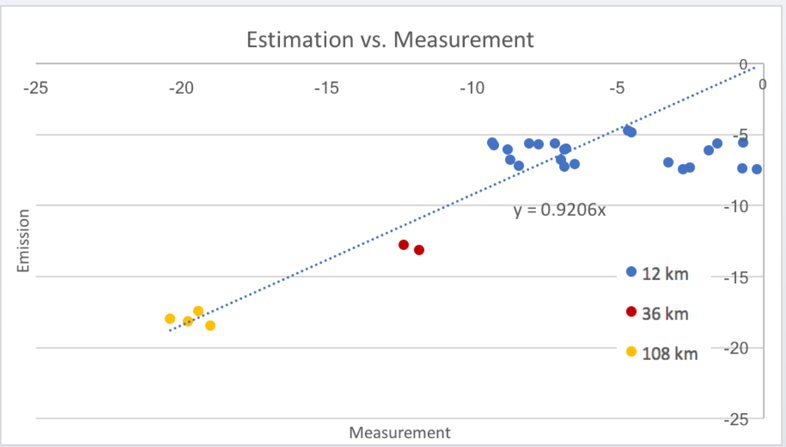

Figure 4

CONCLUSIONS

1. Considering emission from neighborhood surrounding grids can improve the accuracy of estimating $\delta^{15}N$-NO_x.
2. In which circumstances the neighborhood surrounding need to be considered is based on atmospheric dynamics, and atmospheric condition such as temperature, wind, humidity, precipitation, atmospheric stability, which potentially affect the atmospheric physical and chemical process, so that $\delta^{15}N$-NOx will change.
3. Four categories of emission files, incorporated with $\delta^{15}N$, will be used as input of CMAQ (Community Multiscale Air Quality) model to explore the atmospheric processes in detail, so that more accurate estimation of $\delta^{15}N$-NOx could be generated.

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ACKNOWLEDGEMENT

Funding was provided by Purdue University Department of Earth, Atmospheric, and Planetary Sciences, Purdue Climate Change Research Center, and US-AID Pakistan – U.S. S&T Cooperation Program.

Many thanks to Wendell Walters, Brown University for $^{15}NO_x$ measurements.