Quantifying nitrous oxide emissions from agriculture in the Midwest of the U.S.

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PRESENTED AT:

AGU 100 FALL MEETING
San Francisco, CA | 9–13 December 2019
MOTIVATION

Nitrous oxide (N₂O):

- third most important long-lived anthropogenic greenhouse gas
- rising global concentrations
- major anthropogenic contribution: agriculture
- the U.S. is a hotspot of agricultural emissions
- inventories have high uncertainties (e.g., Fu et al., 2017)

N₂O in the atmosphere:

- third most important long-lived anthropogenic greenhouse gas in terms of radiative forcing
- accounts for ~ 7.5% of the total anthropogenic forcing (IPCC, AR5)
- Global Warming Potential on a 100 years horizon (GWP₁₀₀) is 265 (Myhre et al., 2013)
- nowadays the dominant ozone depleting species (Ravishankara et al., 2009)

Global Concentrations of N₂O:
Global concentrations are rising: (https://www.n2olevels.org/) (https://www.n2olevels.org/)

![Nitrous Oxide data from the NOAA/ESRL Global Monitoring Division](https://example.com/graph.png)

- **preindustrial era** (i.e. before 1750): 270 ppbv (MacFarling Meure et al., 2006)
- **August 2019**: 332 ppbv (Combined Nitrous Oxide data from the NOAA/ESRL Global Monitoring Division (ftp.cmdl.noaa.gov/hats/n2o/combined/HATS_global_N2O.txt); last accessed: 20 Nov 2019)
- **current growth**: ~ 0.8 ppbv year\(^{-1}\) (WMO, 2011)

Lifecycle of N\(_2\)O:

Most important anthropogenic contribution: **Agriculture**.
Sources

Anthropogenic - 39%
Natural - 61%

- Fossil fuel combustion and industrial processes 3.9%
- Biomass and biofuel burning 3.9%
- Rivers, estuaries, coastal zones 3.5%
- Atmospheric deposition on land 2.3%
- Human excreta 1.2%
- Atmospheric deposition on ocean 1.2%

Global N₂O budget for 2016; following IPCC ARS, 2013

Sinks

- Stratospheric loss (nearly exclusively) (Machida et al., 1995)
  - N₂O + hν → N₂ + O(¹D)
  - N₂O + O(¹D) → 2NO
  - N₂O + O(¹D) → N₂ + O₂

⇒ lifetime: 118 years (Prather and Hsu, 2010)

N₂O emissions in the U.S.:

(From the EDGARv4.3.2 (http://edgar.jrc.ec.europa.eu/overview.php?v=432&SECURE=123) dataset ranging from 1970 to 2012)

- approximately 9% of the global N₂O emissions in 2012 were emitted in the U.S.
- agricultural emissions are rising since 1970 (not shown)
- the dominant anthropogenic emission sector is 4D1 (direct agricultural soil emissions)
- in 2012 nearly 40% of the total emissions were 4D1 emissions
- **emission hotspots:** Cornbelt and Mississippi area, regions of high agricultural activity

EDGAR v4.3.2: Total Emissions in 2012

**High uncertainties in \( N_2O \) inventories:**

- limited amount of top-down studies
- most studies are based on tall tower measurements and Lagrangian models
- common inventories **significantly underestimate** anthropogenic agricultural emissions
**Table:** Correction factors for agricultural emissions in the U.S. Midwest for various emission inventories; parenthesis indicate the investigated time period.

<table>
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<th>Publication:</th>
<th>EDGAR32 FT2000</th>
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<td>(2008, June)</td>
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<td>1.9 - 4.6</td>
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<td>(2010-2011)</td>
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<td>19.0 - 28.1</td>
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<td>(2012, California)</td>
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METHOD

Comparable to Barkley et al., 2017:

Forward simulation with WRF-Chem + emission inventory

Compare simulated enhancements in the atmosphere with measurements

„Correction factor“ for inventory

Adjust inventory so that differences between simulation and measurements are minimal

Lifetime of N₂O: 118 years (Parthen and Hsu, 2010)

⇒ N₂O is handled as passive tracer

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Inventory:

Only incorporated emission inventory (so far): EDGAR v4.3.2
(H[https://data.europa.eu/val/10.2904/JRC_DATASET_EDGAR]; Lansens-Maenhout et al., 2017)

- spatial resolution: 0.1° x 0.1°
- coverage: global
EDGAR v4.3.2: Agricultural emissions in 2012

WRF-Chem setup:

- version: 4.0.2 (http://www2.mmm.ucar.edu/wrf/users/)
- initial conditions: ERA5 reanalysis (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5)
  - 30 x 30 km
  - 137 vertical layers
  - hourly
- FDDA:
  - D01: analysis nudging, surface analysis nudging, obs. nudging
  - D02: obs. nudging
  - observations: NCEP ADP global surface/upper air observations (https://rda.ucar.edu/) + OBSGRID (https://github.com/wrf-model/OBSGRID)
- Chemistry:
  - passive tracer (chem_opt = 14)
- Simulation performance: Comparison of in-flight measurements of meteorological parameters (wind in the first place) with corresponding simulated values
- Example domain setup for 10 Oct 2017:
two-way nesting

Δx = 15 km

Δx = 3 km

C130 flight track: 10 Oct 2017
CASE STUDIES: 2017 & 2019

10 Oct 2017: Strong N₂O enhancement (≤ 6 ppbv) in an area of high agricultural activity:

10 Oct. 2017 flight track (only in plume) over agricultural EDGARv4.3.2 emissions (Oct. 2010)

⇒ Emitting agricultural EDGARv4.3.2 emissions (AGR emissions) in WRF:
Qualitatively:

- Simulated plumes spatially coincide with measured N$_2$O enhancements
- Simulated enhancements are much too low

Quantitatively:

Increasing strength of emissions by multiplying with factor:

- linear relationship between plume strength and correction factor (compare areas)
- estimated correction factor from linear fit: **14.9**

Simulation performance:
Good agreement between onboard wind measurements and model simulations ⇒ The N₂O transport is assumed to be well represented in the model.

20 June 2019: Strong N₂O enhancement (≤ 10 ppbv) downwind of the Mississippi area:
06 June 2019 flight track (only in plume) over agricultural EDGARv4.3.2 emissions (June 2010)

⇒ Emitting agricultural EDGARv4.3.2 emissions (AGR emissions) in WRF:
Qualitatively (again, like 10 Oct 2017):

+ Simulated plumes spatially coincide with measured N₂O enhancements

- Simulated enhancements are much too low

Quantitatively:

Increasing strength of emissions by multiplying with factor:

- linear relationship between plume strength and correction factor (compare areas)
- estimated correction factor from linear fit: 46.5
- BUT: Flooded Mississippi area most probably influences N₂O emissions ⇒ Further analysis necessary!

Simulation performance:
Good agreement between onboard wind measurements and model simulations ⇒ The N₂O transport is assumed to be well represented in the model.
ACT-AMERICA

(Atmospheric Carbon & Transport - America)

- **2016-2019**: five campaigns and all four seasons
- two aircraft: NASA's C130 and B200
- more than 300 joint flight hours in the Midwest
- N\textsubscript{2}O *in-situ* instruments:
  - QCLS (DLR) (Kostinek et al., 2019): C130; fall 2017 and summer 2019 (~60 hours of data)
  - Flasks (NOAA): C130 and B200; all five campaigns

C130 and B200 flight tracks during fall 2017 and summer 2019 (continuous N\textsubscript{2}O data available (QCLS)):

During each campaign the team was stationed for two weeks in:

- **WFF**: Wallops Flight Facility, Virginia
- **LNK**: Lincoln, Nebraska
- **SHV**: Shreveport, Louisiana
Overview - $N_2O$ during ACTA 2017 & 2019:

- strong enhancements in the lower troposphere observed
- more and stronger enhancements in summer 2019 than in fall 2017

Expected emission strengths during ACTA 2017 & 2019: Throughout the year, anthropogenic $N_2O$ emissions in the U.S. are dominated by agricultural emissions:
SUMMARY & OUTLOOK

So far:

- Good agreement between measured and modelled N₂O plume structures
- Strong underestimation of agricultural N₂O emissions
- Estimated correction factors (so far):
  - 10 Oct 2017: 14.9
  - 06 June 2019: 46.5 (Flooding not taken into account!!!)

Next steps:

1. Calculate backward trajectories to clearly determine the origin of the measured air (partly done)
2. Apply framework on remaining days
3. Simulate remaining ACT-America campaigns (Summer 2016, Winter 2016, Spring 2018) with derived correction factors and compare results to flask measurements
4. Investigate different inventories (at best process-based like DAYCENT (https://www2.nrel.colostate.edu/projects/daycent/))
CV

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Every shown dataset of the QCLS is Revision RA.
ABSTRACT

Atmospheric nitrous oxide (N2O) is, after carbon dioxide and methane, the third most important long-lived anthropogenic greenhouse gas in terms of radiative forcing. Since preindustrial times a rising trend in the global N2O concentrations is observed. Anthropogenic emissions of N2O, mainly from agricultural activity, contribute considerably to this trend. Sparse observational constraints have made it difficult to quantify these emissions. The few studies on top-down approaches in the U.S. that exist are mainly based on Lagrangian models and ground-based measurements. They all propose a significant underestimation of anthropogenic N2O emission sources in established inventories, such as the Emissions Database for Global Atmospheric Research (EDGAR).

In this study we quantify anthropogenic N2O emissions in the Midwest of the U.S., an area of high agricultural activity. In the course of the Atmospheric Carbon and Transport – America (ACT-America) campaign spanning from summer 2016 to summer 2019, an extensive dataset over four seasons has been collected including in-situ N2O aircraft based measurements in the lower and middle troposphere onboard NASA's C-130 and B-200 aircraft. During fall 2017 and summer 2019 we conducted measurements onboard the NASA-C130 with a Quantum-Cascade-Laser-Spectrometer (QCLS) and on both aircraft over the whole campaign flask measurements (NOAA) were collected. More than 300 joint flight hours were conducted and more than 500 flask samples were collected over the U.S. Midwest. The QCLS system collected continuous N2O data for approximately 60 flight hours in this region. The Eulerian Weather Research and Forecasting model with chemistry enabled (WRF-Chem) is being used to quantify regional agricultural N2O emissions using the spatial characteristics of these atmospheric N2O mole fraction observations. The numerical simulations enable potential surface emission distributions to be compared to our airborne measurements, and source estimates can be adjusted to minimize the differences, thus quantifying N2O sources. These results are then compared to emission rates in the EDGAR inventory.
REFERENCES


