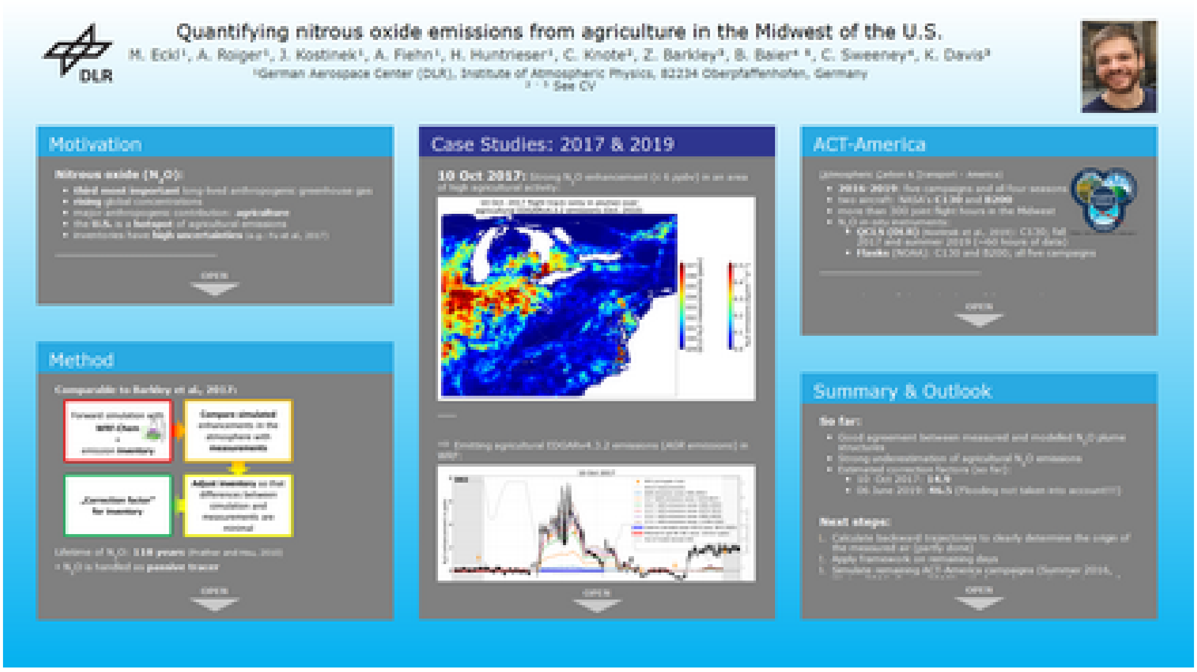
Quantifying nitrous oxide emissions from agriculture in the Midwest of the U.S.



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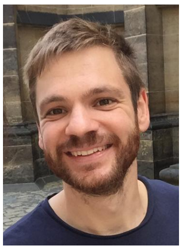


PRESENTED AT:

MOTIVATION

Nitrous oxide (N_2O):

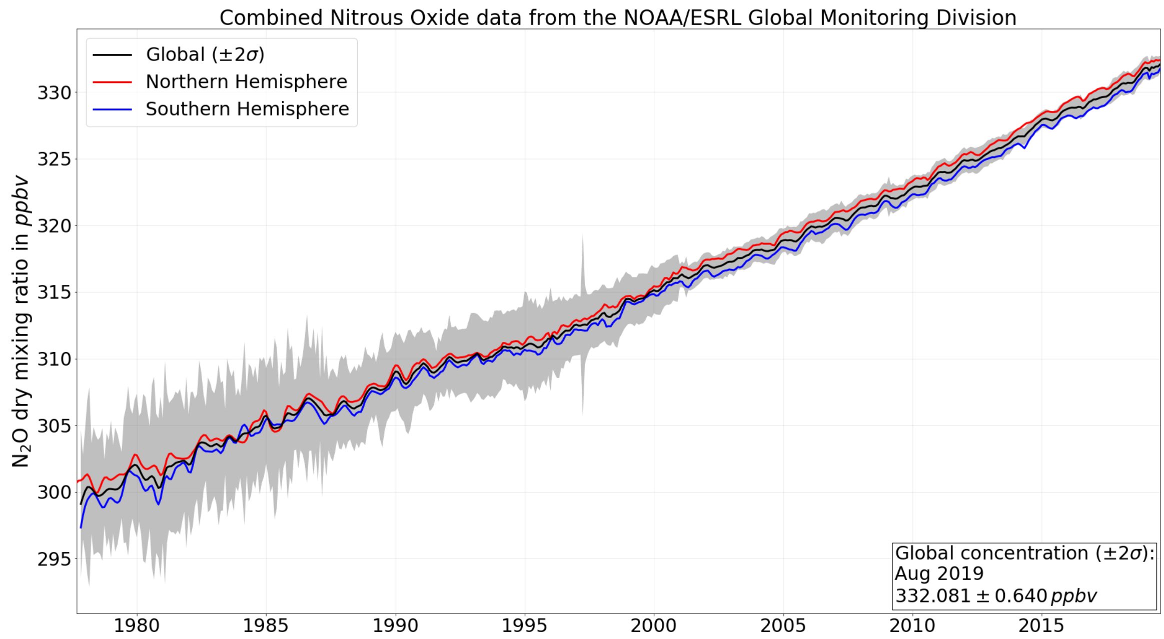
- **third most important** long-lived anthropogenic greenhouse gas
- **rising** global concentrations
- major anthropogenic contribution: **agriculture**
- the **U.S.** is a **hotspot** of agricultural emissions
- inventories have **high uncertainties** (e.g.: Fu et al., 2017)

N_2O 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 (GWP_{100}) is **265** (Myhre et al., 2013)
- nowadays the dominant ozone depleting species (Ravishankara et al., 2009)

Global Concentrations of N_2O:

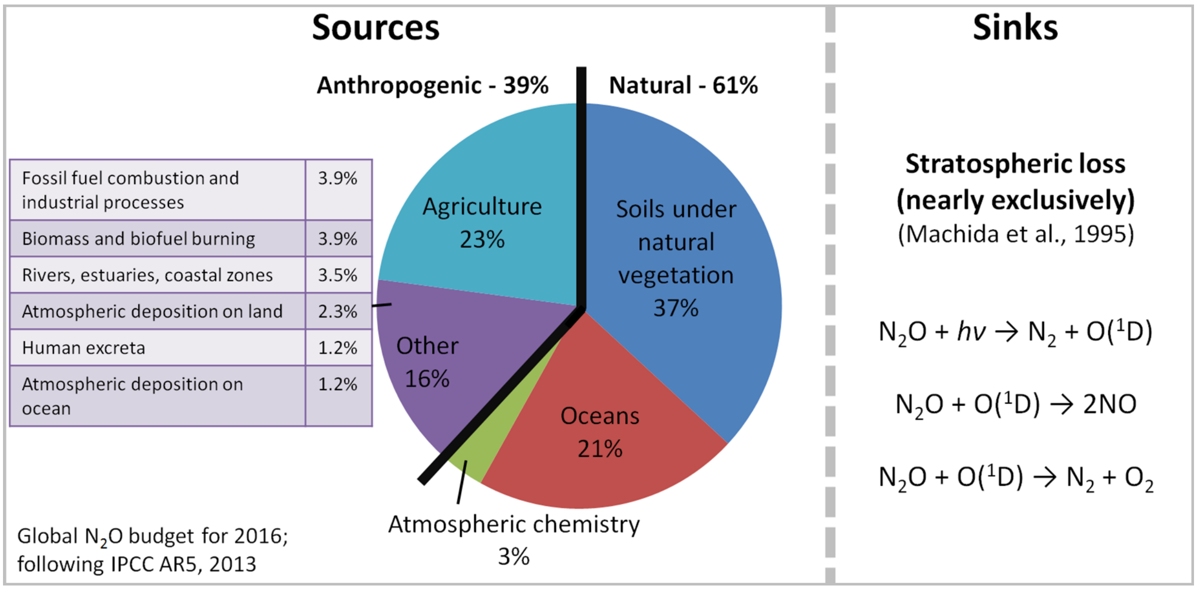
Global concentrations are rising: (https://www.n2olevels.org/) (https://www.n2olevels.org/)



- 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_2O:

Most important anthropogenic contribution: **Agriculture**.

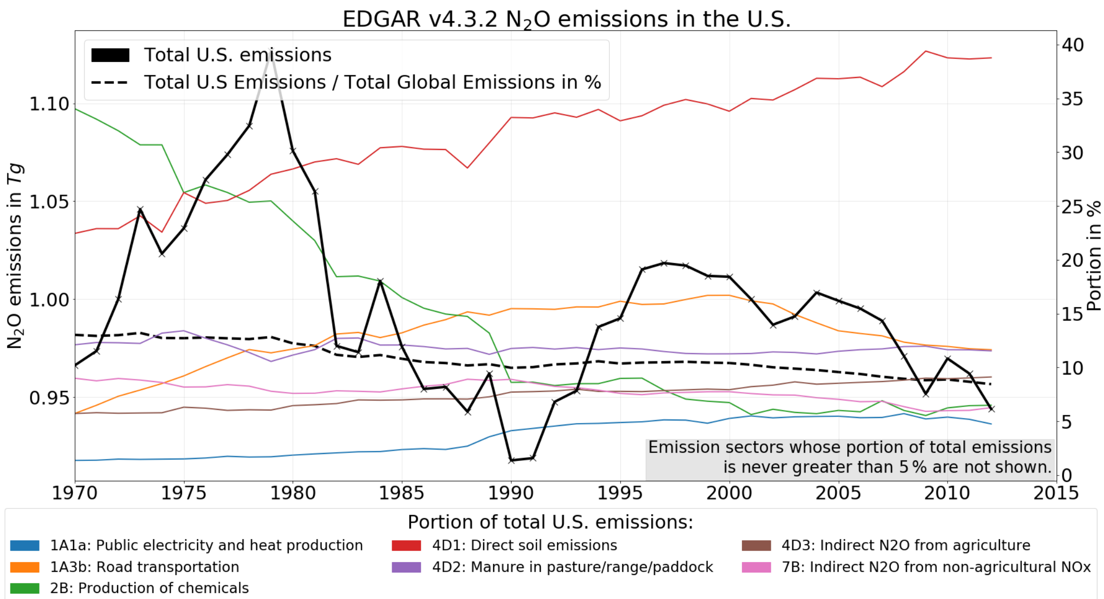


⇨ lifetime: **118 *years*** (Prather and Hsu, 2010)

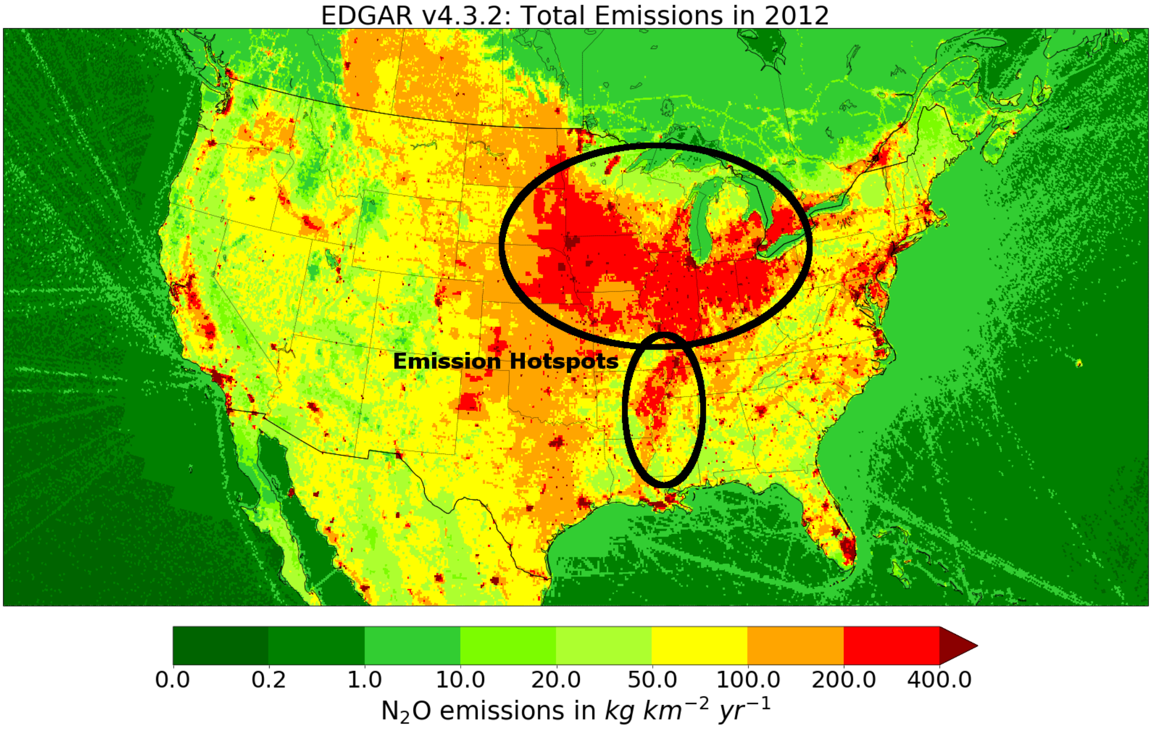
N_2O emissions in the U.S.:

(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 N_2O 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



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

Publication \ Inventory:	EDGAR32 FT2000	EDGAR4	EDGAR42	GEIA	DLEM
Miller et al., 2012 (2008, June)	5.4	10.1		4.5	2.1
Chen et al., 2016 (2010-2011)			1.9 - 4.6		
Griffis et al., 2013 (2010)			2.6	8.8	
Fu et al., 2017 (2010, June)			19.0 - 28.1		
Kort et al., 2008 (2003, May-June)	2.62			3.05	
Xiang et al., 2013 (2012, California)	1.14	1.62		1.62	

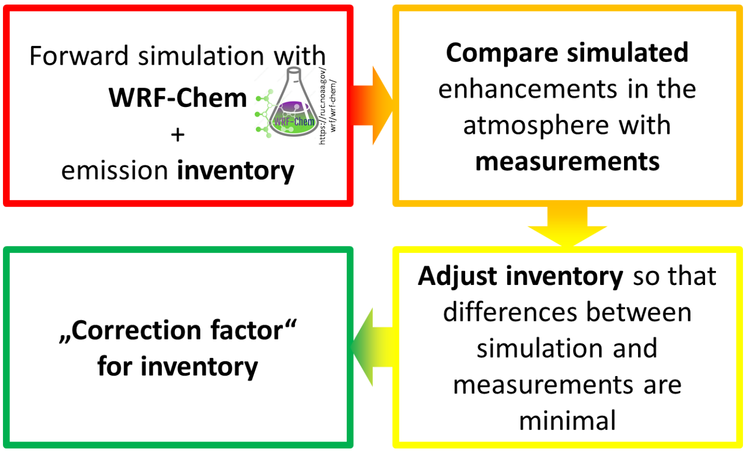
tall tower measurements + **Lagrangian** model (STILT+WRF): Miller et al., 2012; Chen et al., 2016; Griffis et al., 2013

tall tower measurements + **Eulerian** model (WRF-Chem): Fu et al., 2017

aircraft measurements + **Lagrangian** model (STILT+WRF): Kort et al., 2008; Xiang et al., 2013

METHOD

Comparable to Barkley et al., 2017:



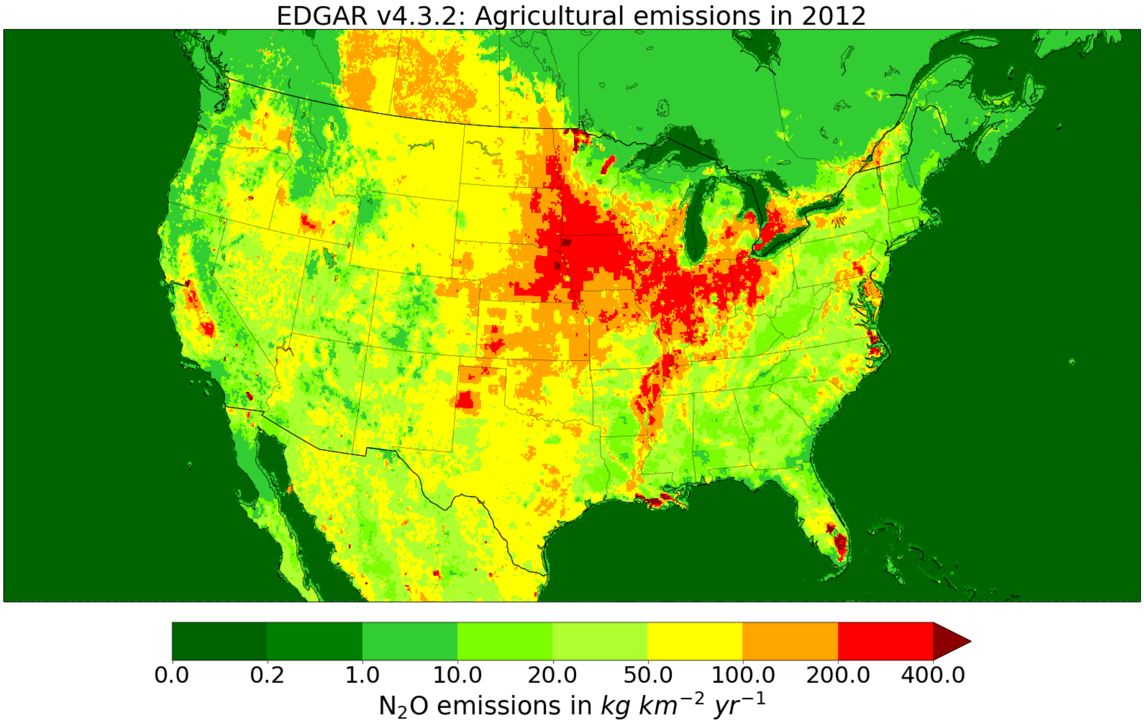
Lifetime of N_2O: **118 *years*** (Prather and Hsu, 2010)

⇨ N_2O is handled as **passive tracer**

Inventory:

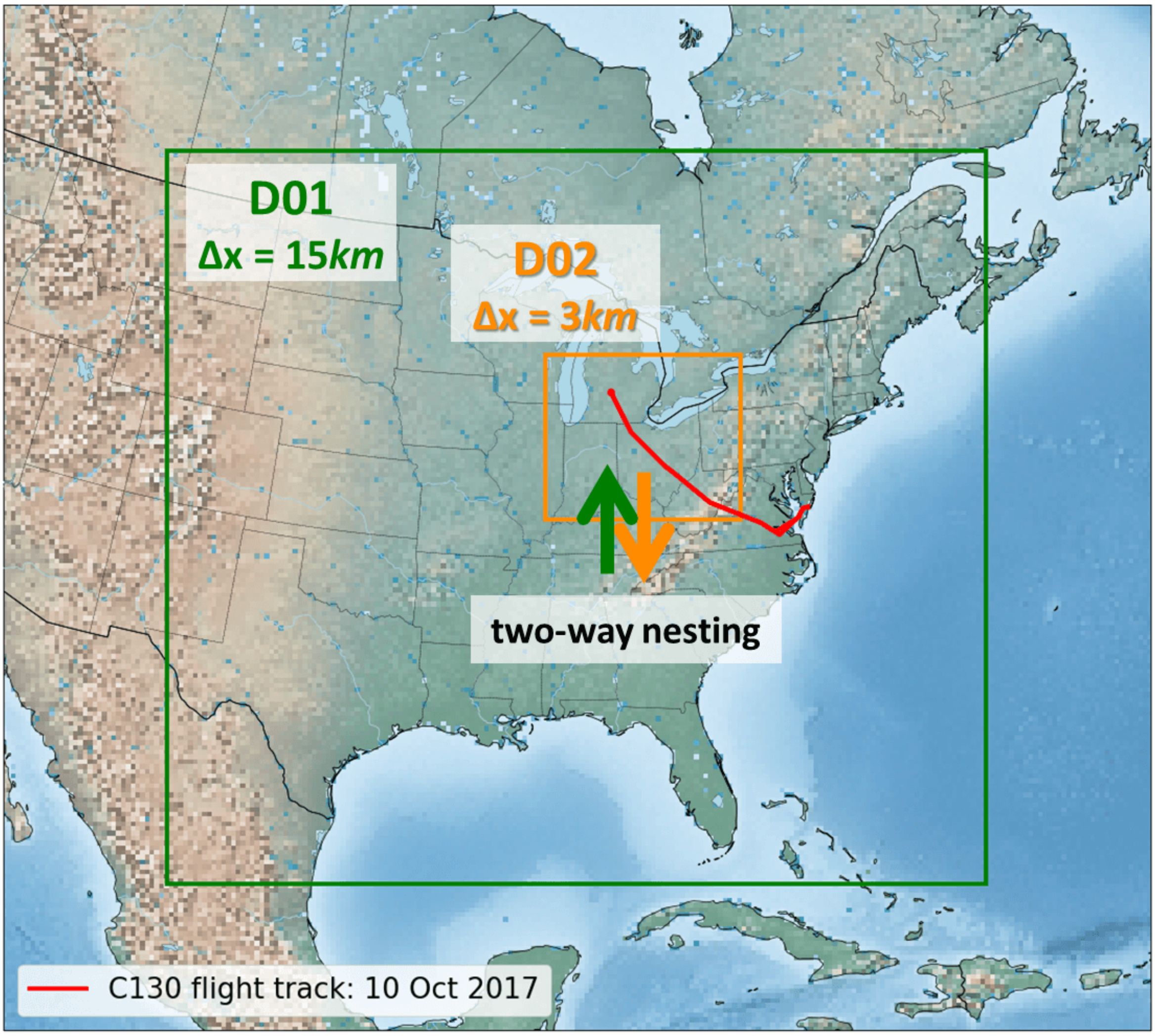
Only incorporated emission inventory (so far): **EDGAR v4.3.2** (http://edgar.jrc.ec.europa.eu/overview.php?v=432&SECURE=123) (Emissions Database for Global Atmospheric Research; https://data.europa.eu/doi/10.2904/JRC_DATASET_EDGAR (https://data.europa.eu/doi/10.2904/JRC_DATASET_EDGAR); Janssens-Maenhout et al., 2017)

- temporal resolution: yearly (1970-2012) and monthly (2010)
- spatial resolution: 0.1 ° x 0.1 °
- coverage: global



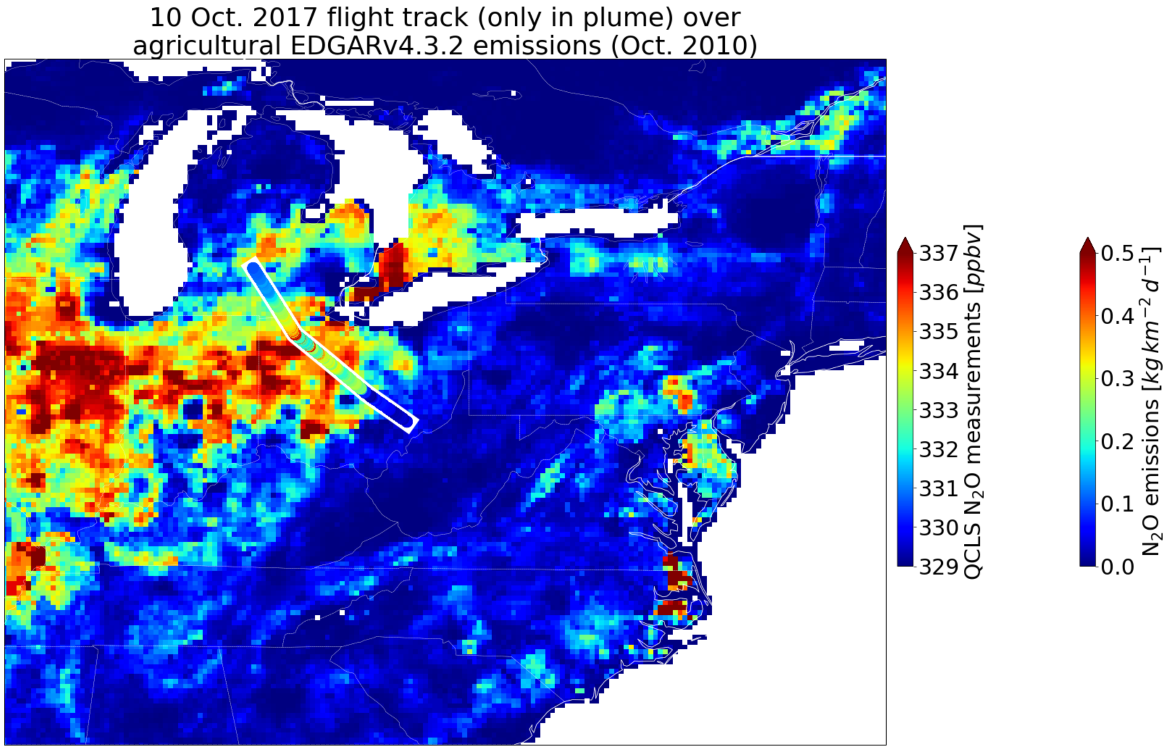
WRF-Chem setup:

- 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**)
  - emissions: **EDGAR** (http://edgar.jrc.ec.europa.eu/overview.php?v=432&SECURE=123) + **anthro_emiss** (https://www2.acom.ucar.edu/wrf-chem/wrf-chem-tools-community)
- Simulation performance: Comparison of in-flight measurements of meterological parameters (wind in the first place) with corresponding simulated values
- Example domain setup for 10 Oct 2017:


D01
Δx = 15km
D02
Δx = 3km
two-way nesting
C130 flight track: 10 Oct 2017

CASE STUDIES: 2017 & 2019

10 Oct 2017: Strong N_2O enhancement ($\lesssim$ 6 *ppbv*) in an area of high agricultural activity:



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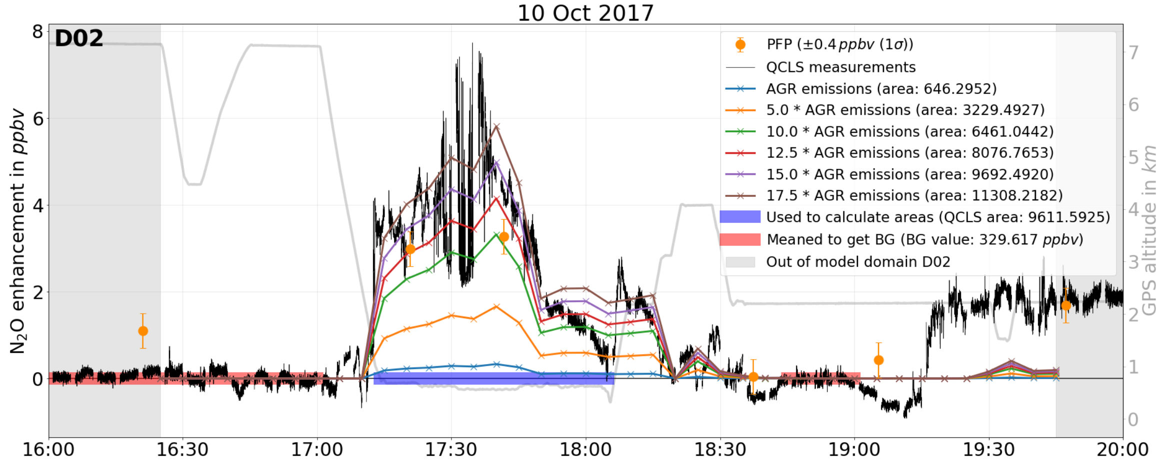
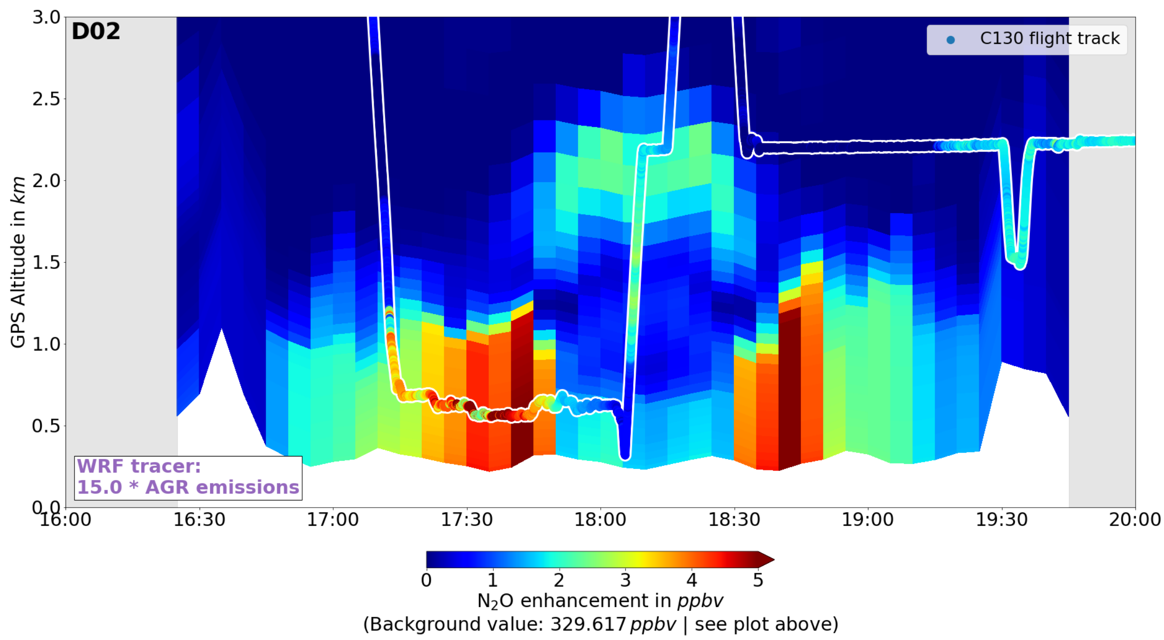
⇒ Emitting agricultural EDGARv4.3.2 emissions (*AGR emissions*) in WRF:





Qualitatively:

+ Simulated plumes spatially coincide with measured N_2O enhancements

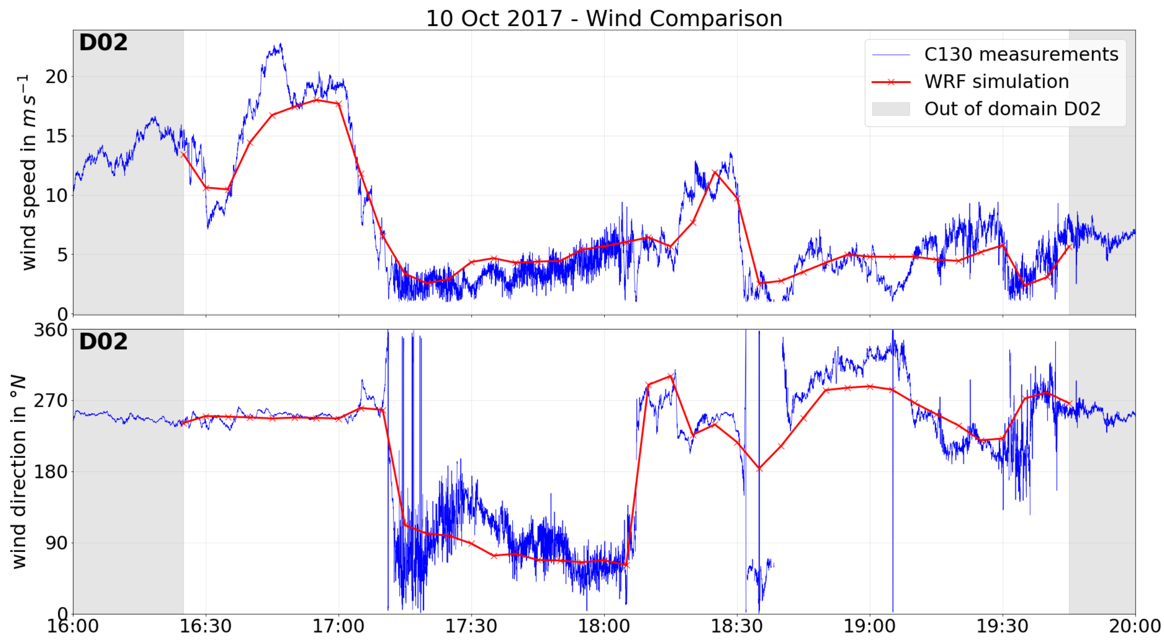
- Simulated enhancements are much too low

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 measurments and model simulations ⇒ The N_2O transport is assumed to be well represented in the model.

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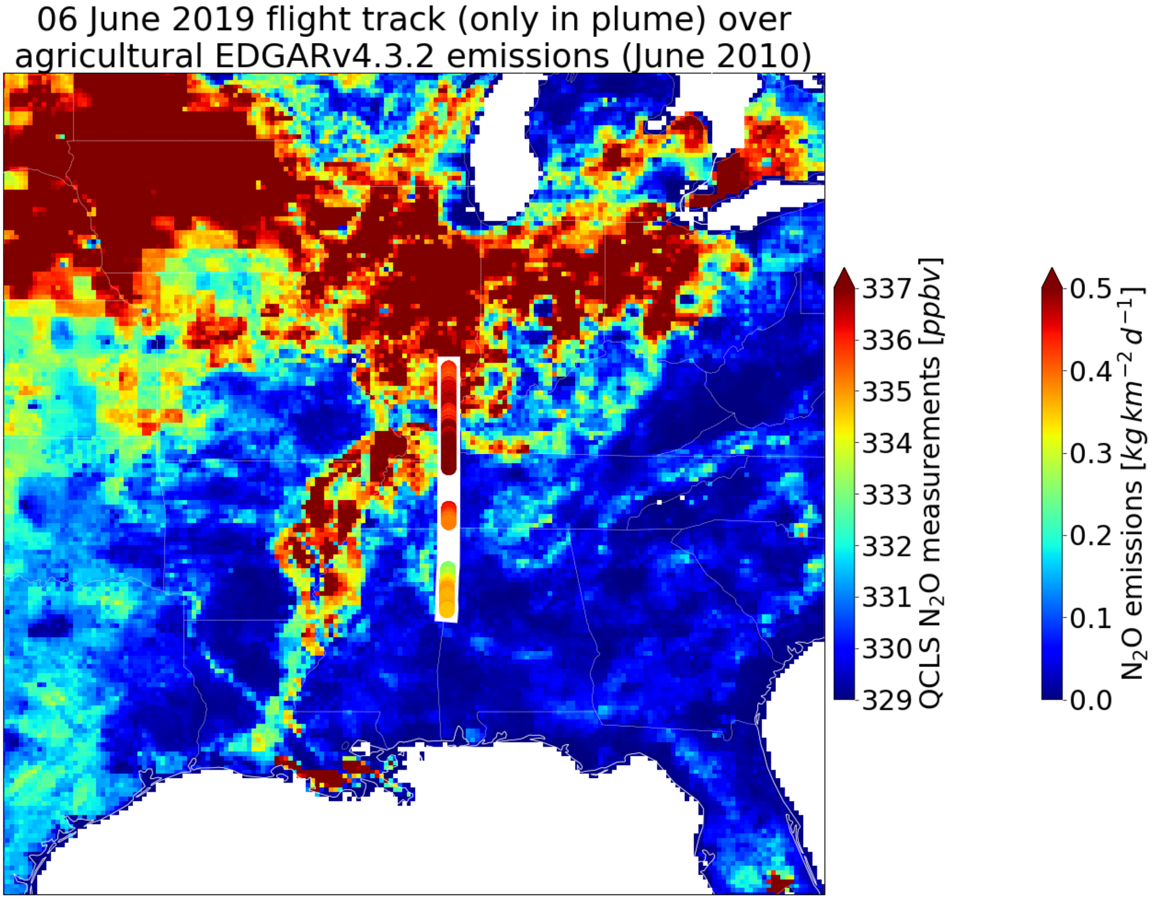
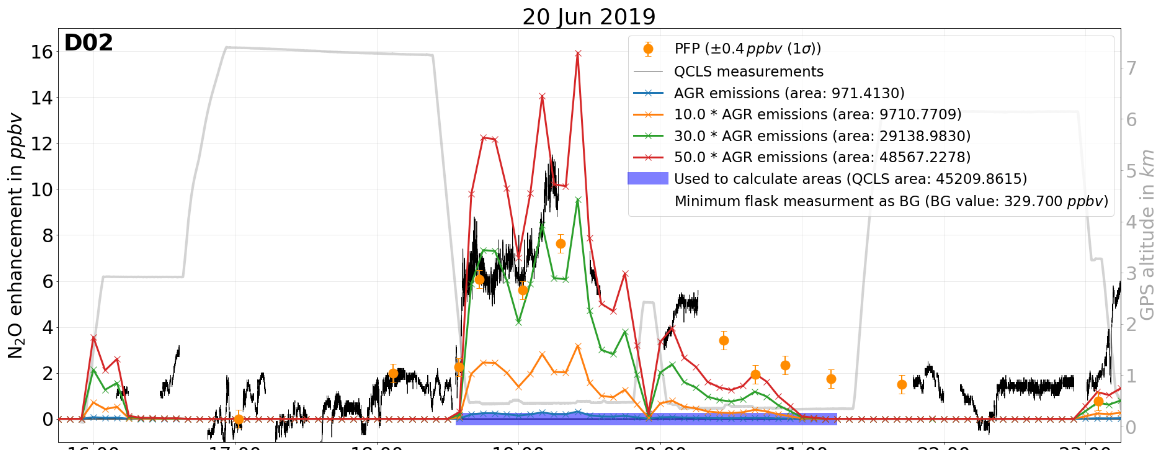
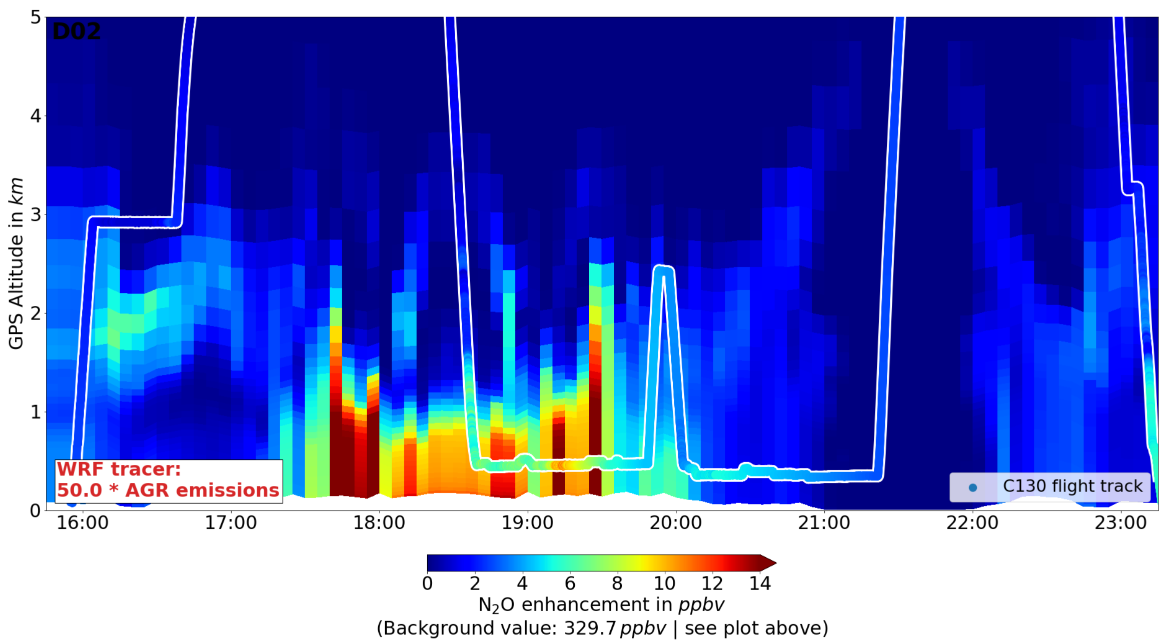
20 June 2019: Strong N_2O enhancement ($\lesssim$ 10 *ppbv*) downwind of the Mississippi area:



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





Qualitatively (again, like 10 Oct 2017):

+ Simulated plumes spatially coincide with measured N_2O enhancements

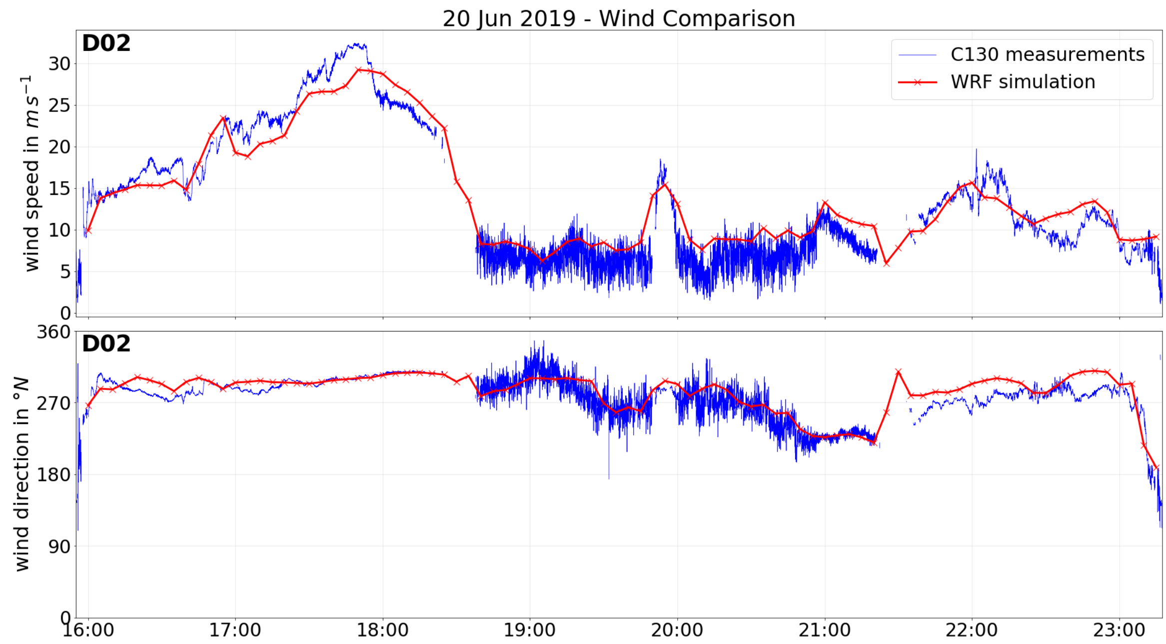
- Simulated enhancements are much too low

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 N_2O emissions ⇒ **Further analysis necessary!**

Simulation performance:



Good agreement between onboard wind measurments and model simulations ⇒ The N_2O transport is assumed to be well represented in the model.

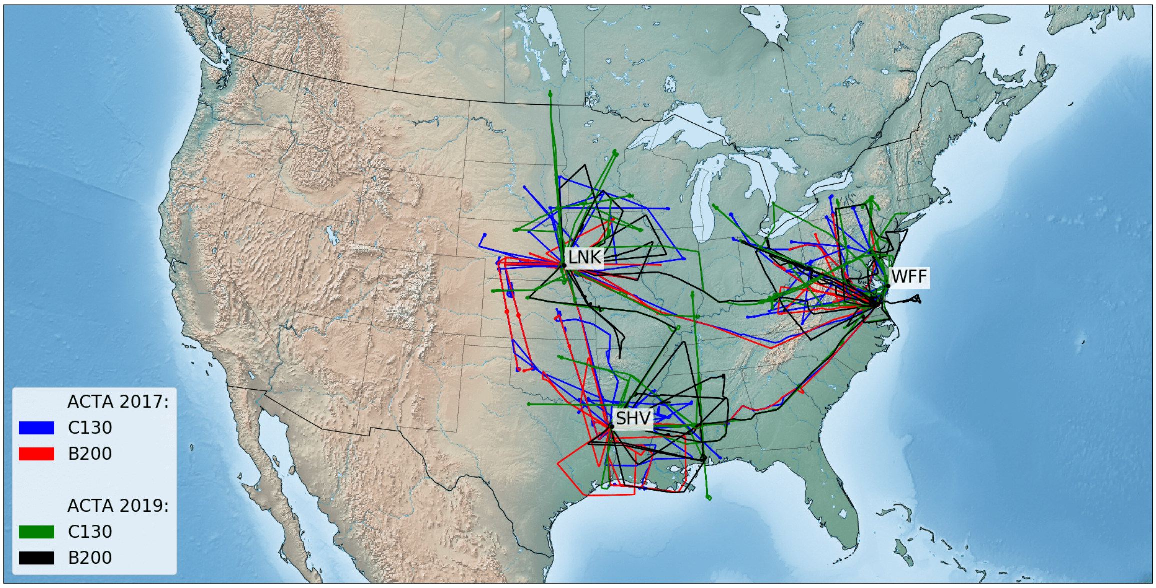
ACT-AMERICA

(Atmospheric Carbon & Transport - America)

https://act-america.larc.nasa.gov/

- **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_2O *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 (continous N_2O data available (QCLS)):

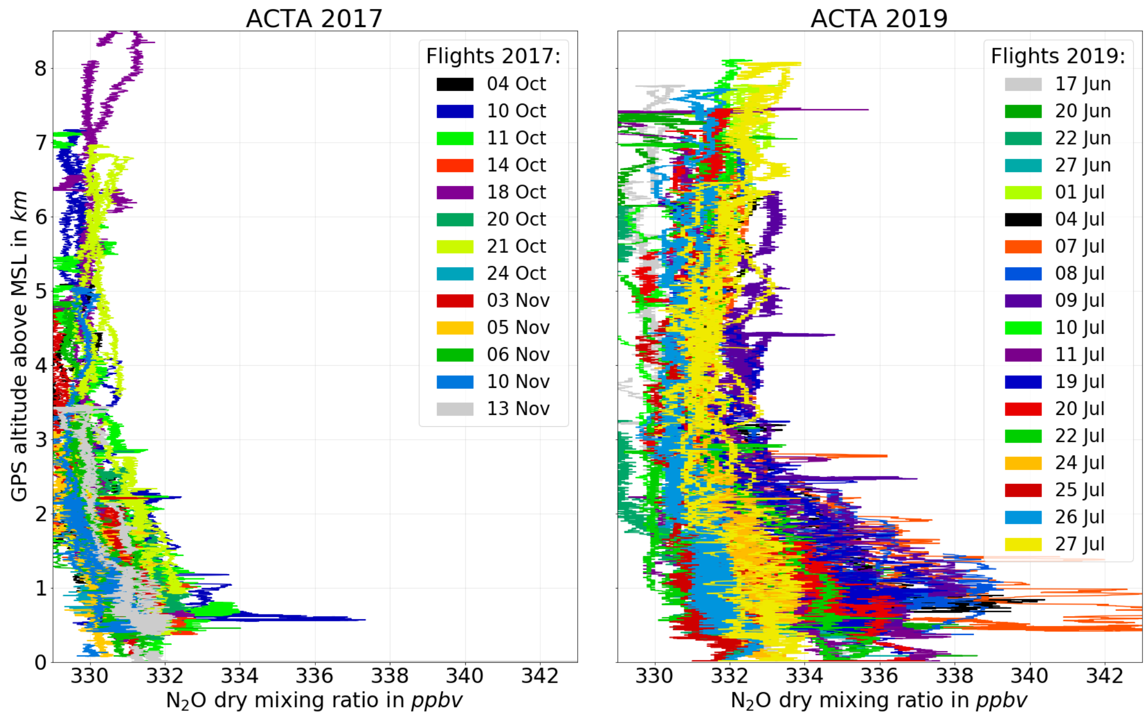


During each campaign the team was stationed for two weeks in:

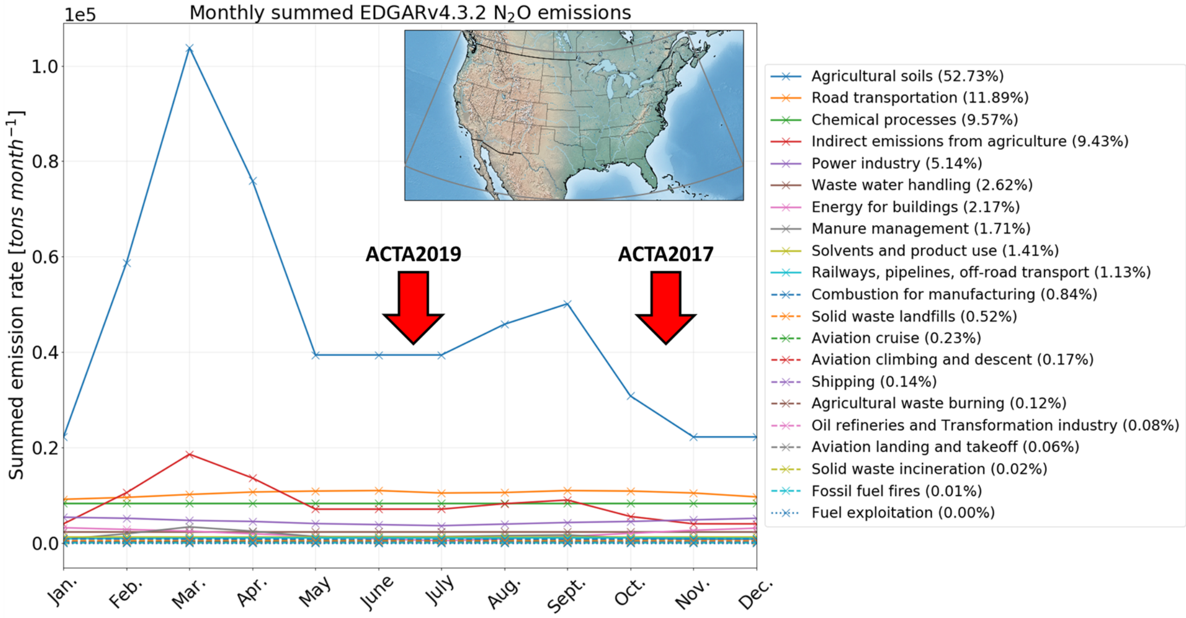
- WFF: Wallops Flight Facility, Virginia
- LNK: Lincoln, Nebraska
- SHV: Shreveport, Louisiana

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:



Monthly summed EDGARv4.3.2 N_2O emissions
1e5
Summed emission rate [tons month^{-1}]
1.0
0.8
0.6
0.4
0.2
0.0
ACTA2019
ACTA2017
Jan.
Feb.
Mar.
Apr.
May
June
July
Aug.
Sept.
Oct.
Nov.
Dec.
Agricultural soils (52.73%)
Road transportation (11.89%)
Chemical processes (9.57%)
Indirect emissions from agriculture (9.43%)
Power industry (5.14%)
Waste water handling (2.62%)
Energy for buildings (2.17%)
Manure management (1.71%)
Solvents and product use (1.41%)
Railways, pipelines, off-road transport (1.13%)
Combustion for manufacturing (0.84%)
Solid waste landfills (0.52%)
Aviation cruise (0.23%)
Aviation climbing and descent (0.17%)
Shipping (0.14%)
Agricultural waste burning (0.12%)
Oil refineries and Transformation industry (0.08%)
Aviation landing and takeoff (0.06%)
Solid waste incineration (0.02%)
Fossil fuel fires (0.01%)
Fuel exploitation (0.00%)

SUMMARY & OUTLOOK

So far:

- Good agreement between measured and modelled N_2O plume structures
- Strong underestimation of agricultural N_2O 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.

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