Sensitivity of Particulate Matter Concentrations to Revised Estimates of Onroad Ammonia Emissions

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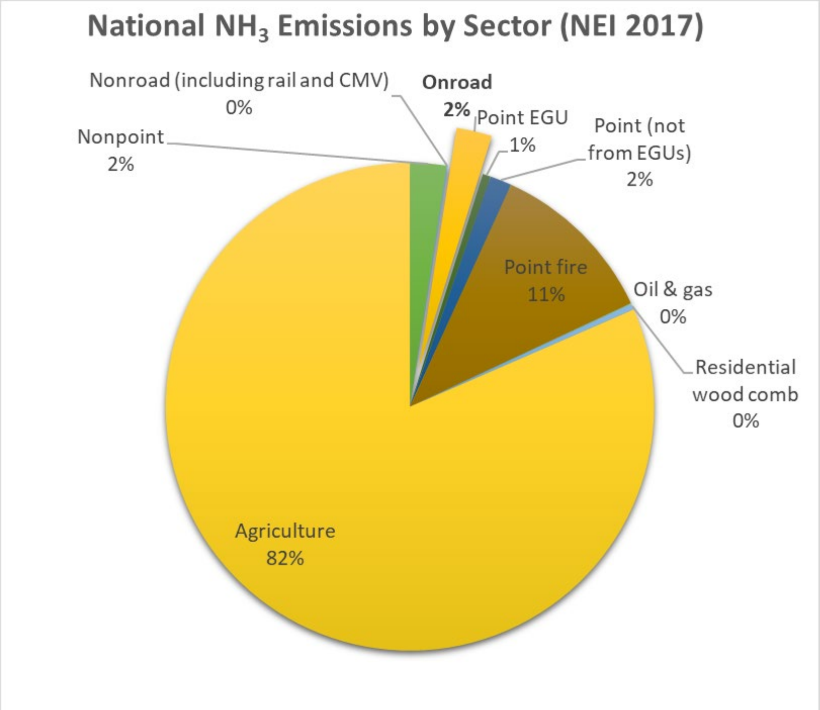
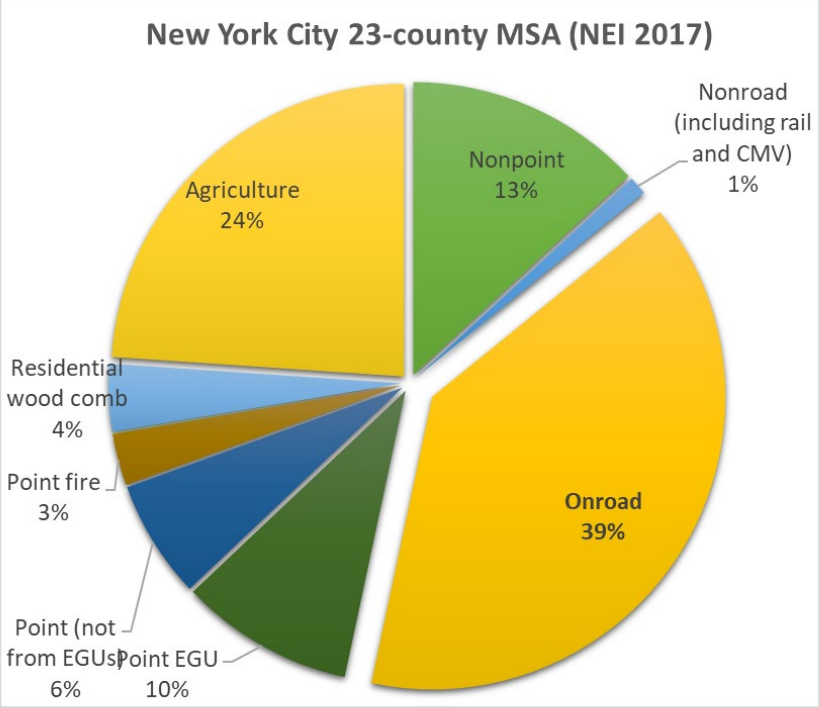
Background

- Nationwide, ammonia (NH_3) emissions dominated by agriculture and fires
- In urban areas, onroad vehicles are significant source of NH_3 emissions
  - **Light-duty gasoline vehicles:** Catalytic reduction of NO to form NH_3 in three-way catalytic converter under fuel rich conditions
  - **Heavy-duty diesel vehicles**: Overdosing of urea in selective catalytic reduction (SCR) systems used to control NOx leads to "ammonia slip"
- EPA's Motor Vehicle Emission Simulator (MOVES)
  - Estimates onroad and nonroad vehicle emissions for EPA's emissions modeling platform
  - Onroad NH_3 emissions based on studies conducted in 2001 and earlier on limited number of vehicles





Motivation

- Research suggests mobile-source NH_3 inventories in urban areas are underestimated by MOVES and the EPA emissions modeling platform
  - Sun et al. (2017) – On-road measurements of NH_3/CO_2 suggest that mobile-source NH_3 is more than 2 X higher than reported in the 2011 NEI
  - Moravek et al. (2019) and Emery et al. (2020) found better air quality model agreement to ammonia and ammonium-nitrate in Salt Lake City when mobile NH_3 increased by 2 X
- Arter et al. (2021) estimated that mobile-source ammonia emissions contribute to significant health burden
  - Estimated that NH_3 emissions have larger health impacts than NOx emissions from onroad vehicles in the northeastern United States

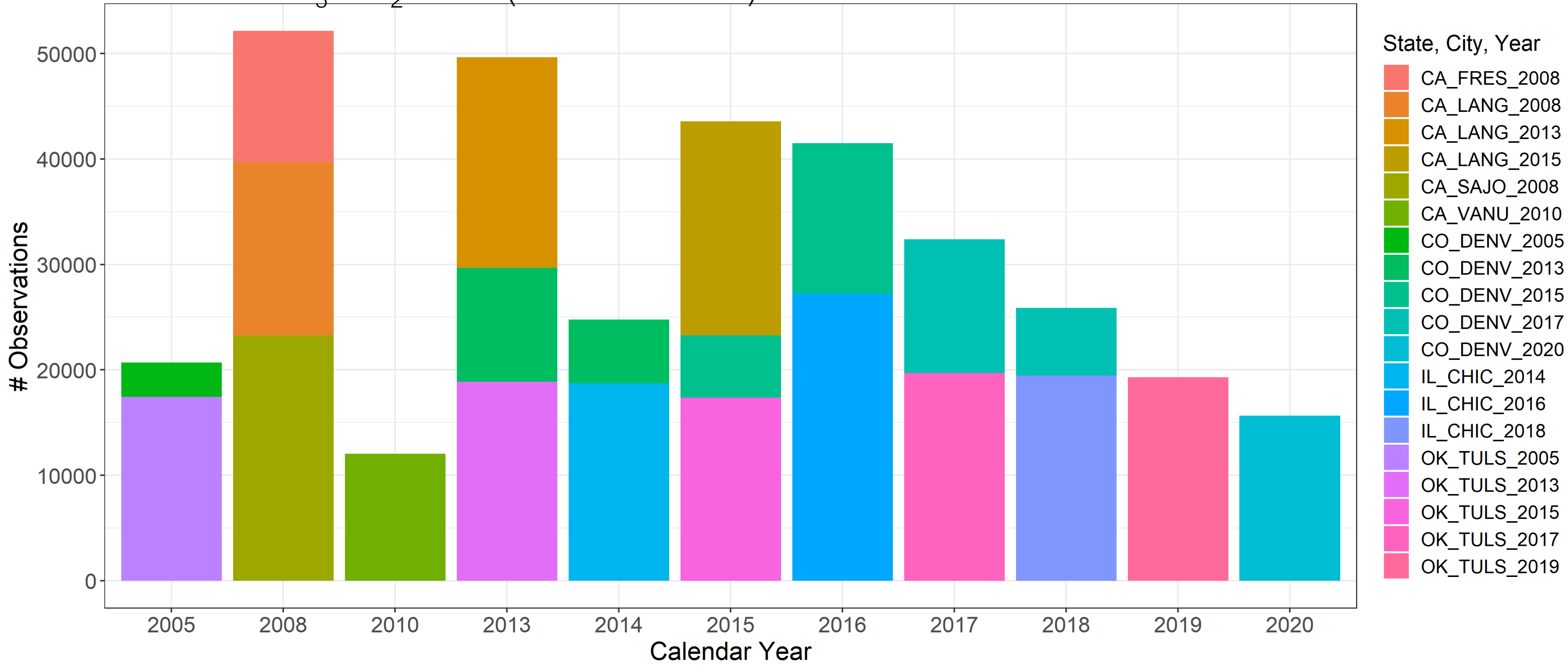
Objectives

- Compare NH_3 emission rates in MOVES to recent remote sensing and road-side studies
- Estimate sensitivity of air quality to changes in onroad NH_3 emissions

Light-Duty Remote Sensing Data (RSD)

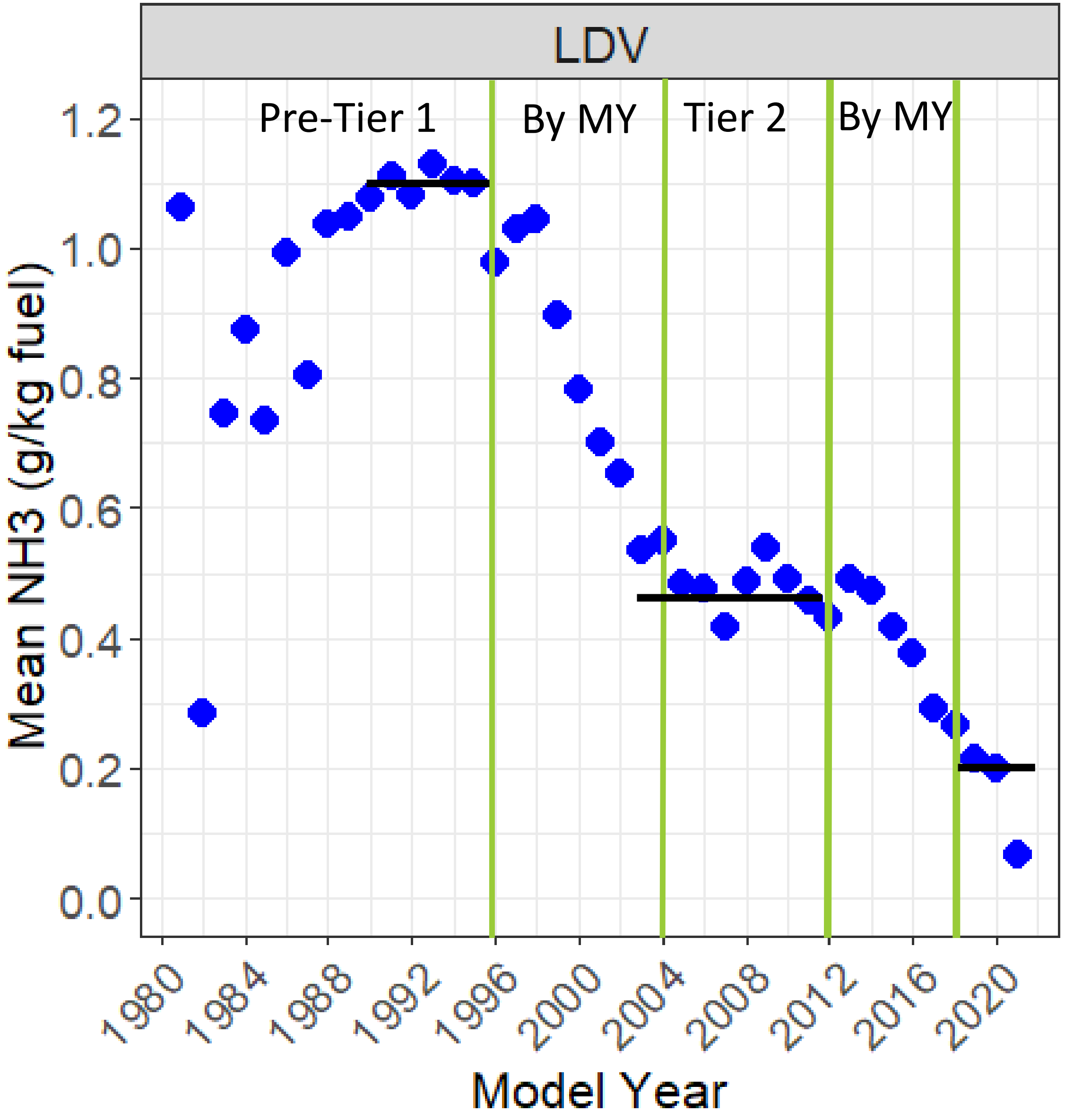
- RSD collected by University of Denver (see Bishop et al. 2015)
  - Over 335,000 light-duty gasoline vehicle-specific NH_3 observations made in campaigns conducted from 2005 to 2020 available at http://www.feat.biochem.du.edu/
  - Seven locations throughout the United States (four in California)
  - Each measurement includes vehicle model, model year/age, vehicle speed, and acceleration
- Fleet average measurements from University of Denver compare well to tunnel and onroad fleet NH_3/CO_2 ratios (Sun et al. 2017)

Photo from Bishop et al. 2015
(Used with permission)



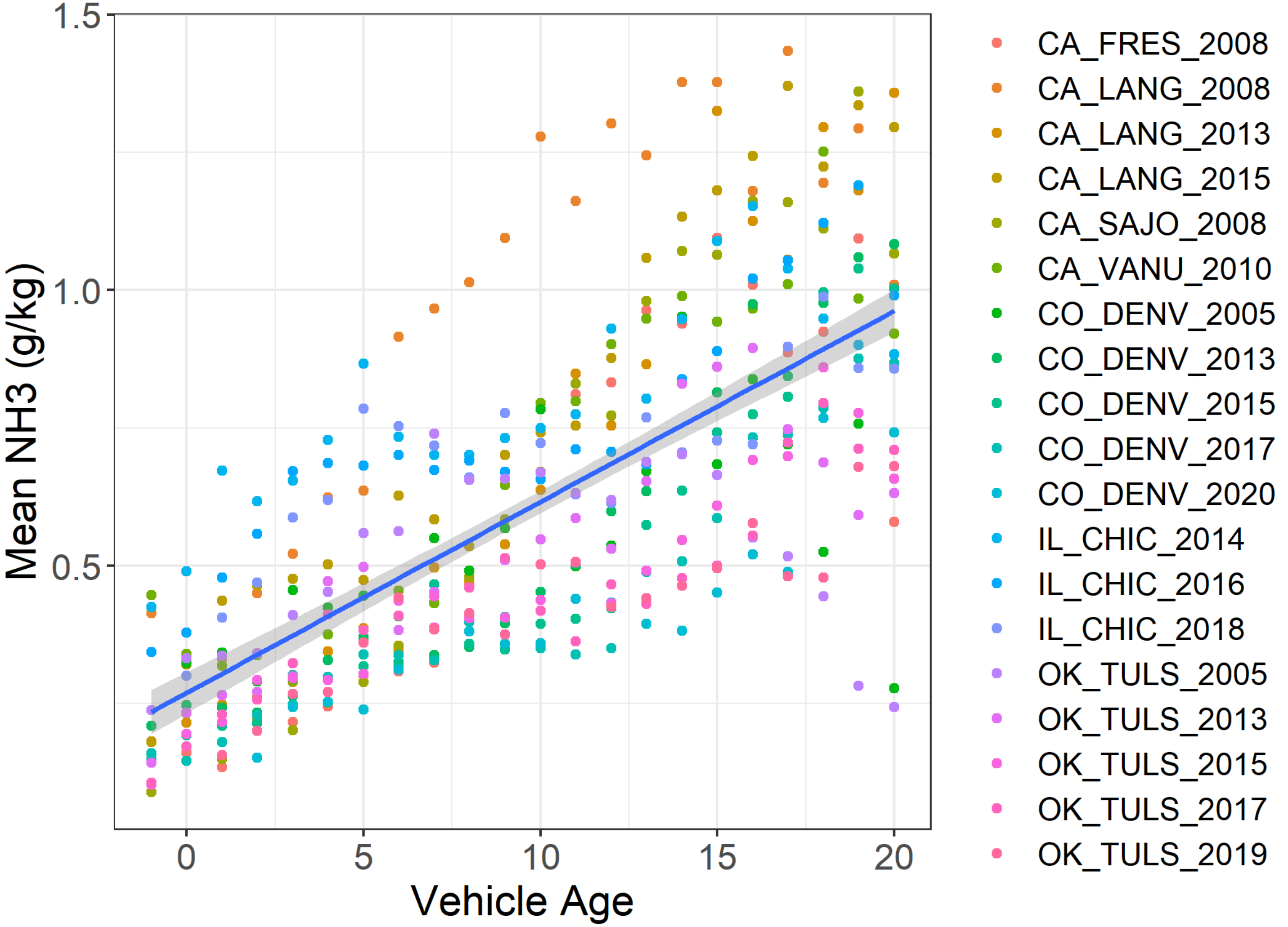
LD NH_3 Emission Rates for Sensitivity Analysis

- Model Year (MY) specific emission rates
  - Significant model year effect observed in RSD emission rates
  - Developed average rates for MY ranges where observed values are stable
  - Derived MY-specific rates for periods of rapid change
  - After MY 2018, rates projected to remain the same
  - Estimated separately for light-duty vehicles (LDV) and light-duty trucks (LDT)



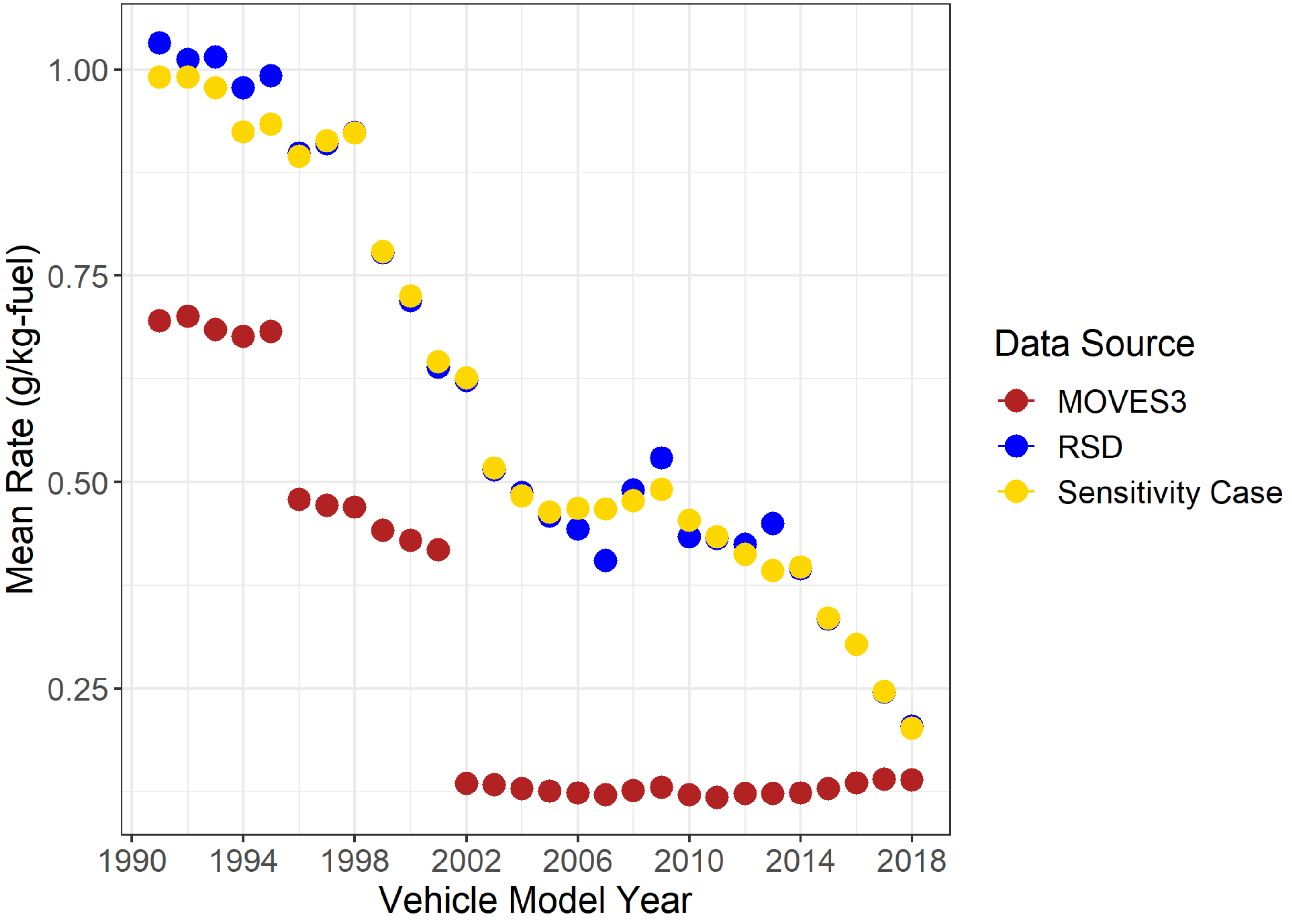
LD NH_3 Emission Rates for Sensitivity Analysis

- Age Effects
  - Significant age effect observed in light-duty remote sensing data
  - Estimated emission rates by model year and age group
  - For missing vehicle class, model year, and age combinations (e.g., age 2+ for MY 2018) applied the same age effects from earlier model years



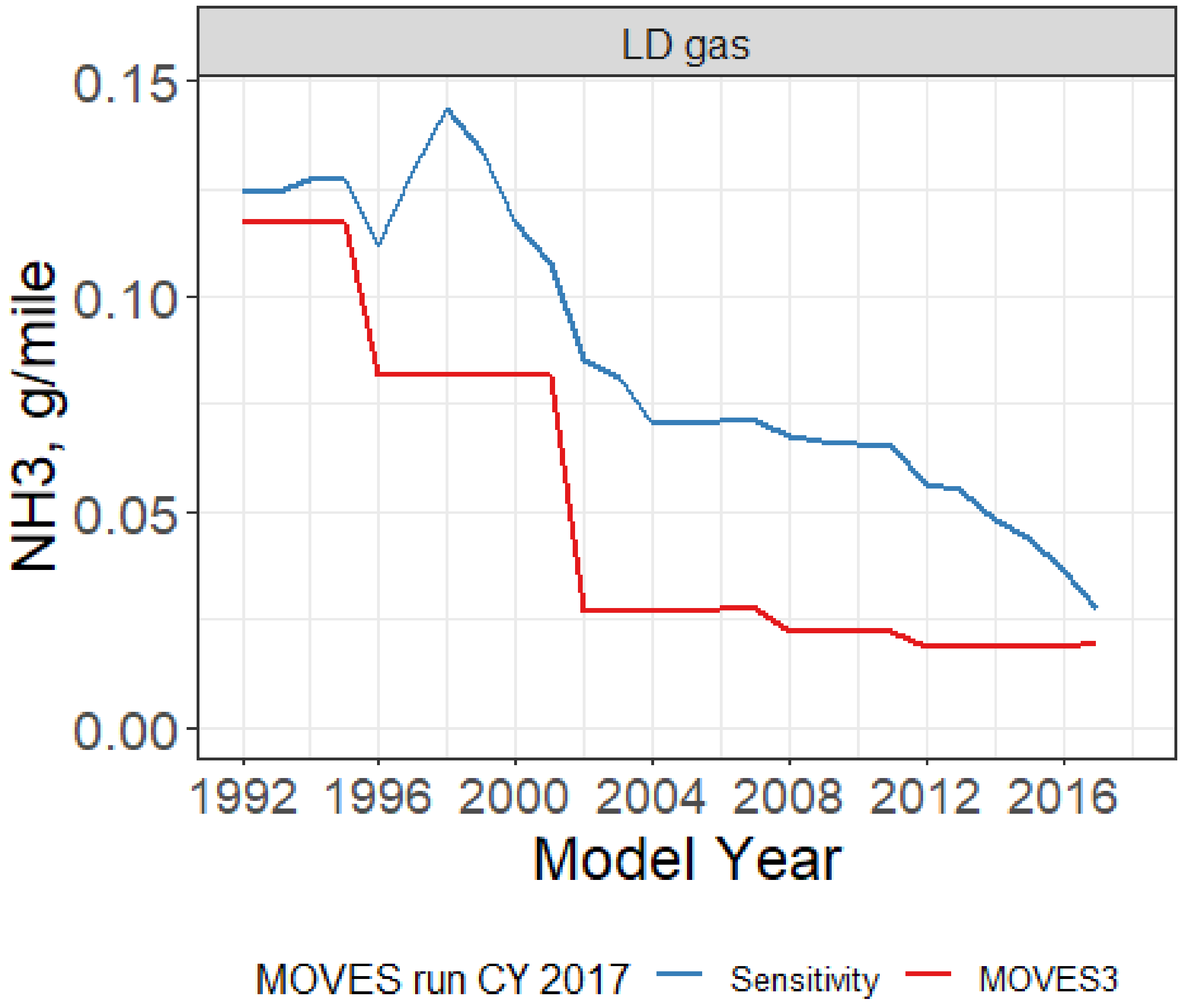
LD NH_3 Emission Rates for Sensitivity Analysis

- We assigned MOVES3 rates to each RSD observation based on
  - Vehicle class (LDV or LDT)
  - Model year (MY)
  - Vehicle age groups (e.g. 0-3, 4-5, 6-7)
  - Operating conditions (speed, acceleration)
- In the same way, we assigned the sensitivity rates newly developed to each RSD observation
- Finally, we averaged by MY to create the plot shown here
- **RSD fuel-based emission rates are significantly higher than MOVES3 across all model years**
- **LHD Sensitivity emission rates capture the magnitude and trend of the RSD data**
  - Small differences between RSD and Sensitivity rates are due to averaging across model year and vehicle age groups



LD NH_3 Emission Rates for Sensitivity Analysis

- Time-based emission rates (g/hour) = fuel-based (gNH_3/kg-fuel) rates from RSD x fuel consumption rates in MOVES (kg-fuel/hour)
  - Use MOVES fuel consumption rates by model year, vehicle class and running operating modes
  - Applied in MOVES run to estimate distance-based rate (g/mile) for individual calendar year and representative operating modes
- Similar trend observed in distance-based and fuel-based emission rates
  - Largest differences between sensitivity case and MOVES occurs for vehicles from ages 5 to 20 (Model years 1997-2012)

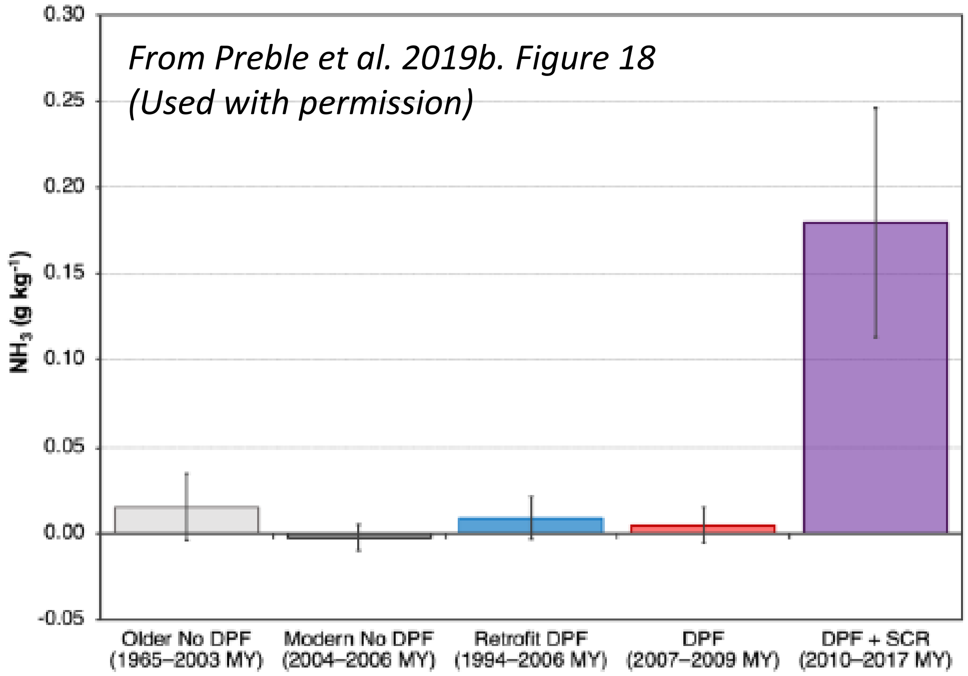


Heavy-duty (HD) Vehicle NH_3 Emission Data

- Caldecott Tunnel outside Oakland, California (Preble, et al. 2019a)
  - Over 900 diesel truck NH_3 measurements identified by model year
  - Observed large increase in NH_3 emissions with trucks equipped with selective catalytic reduction (SCR) aftertreatment systems (MY 2010+)
  - Measurements of pre-2010 MY heavy-duty diesel vehicles are low and uncertain
    - Comparable to previous measurements made in the Caldecott Tunnel in 2006
- Peralta Weigh Station near Anaheim, California (Haugen et al. 2018)
  - 1,844 diesel truck measurements
  - Large increase in NH_3 in the 2017 campaign compared to previous campaigns, due to presence of MY 2010+ trucks


Caldecott Tunnel, Preble et al. 2019a


Peralta Weight Station,Haugen et al. 2018



Model Year*	Caldecott Tunnel (g/kg)[1]	Peralta Weigh Station (g/kg)[2]
2010-2018	**0.18 ± 0.07; N = 547**	0.14
2007-2009	**0.00 ± 0.01; N = 181**	~0
2004-2006 (no DPF)	**0.00 ± 0.01; N =24**	~0
1960-2003	**0.02 ± 0.02; N = 62**	~0
2018 HDD fleet average	0.1; N = 1167	0.09; N = 1844

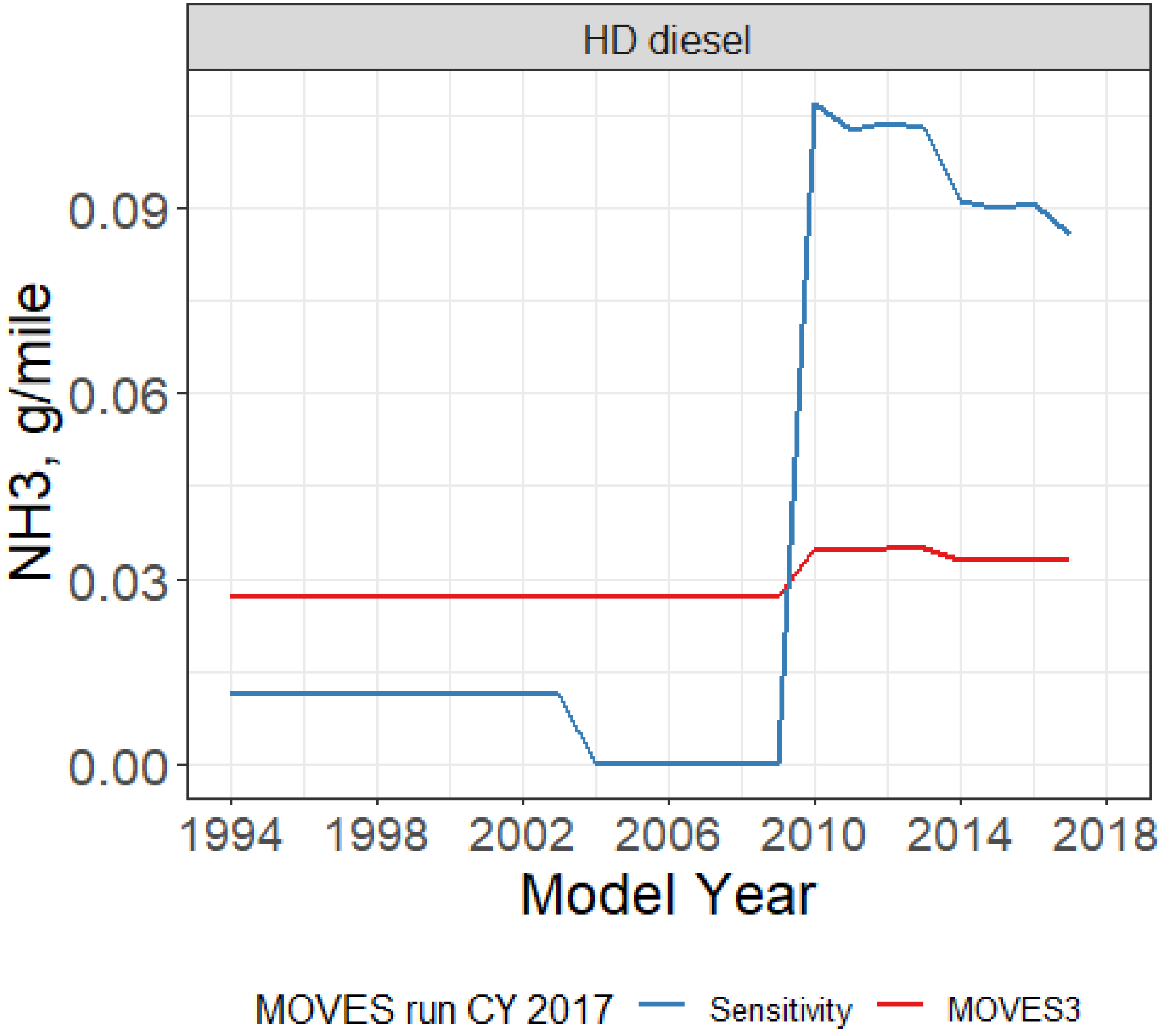
*With the 2010 NOx standards, HD diesel engines often lagged the chassis model year by 1 yr

[1] Engine model year

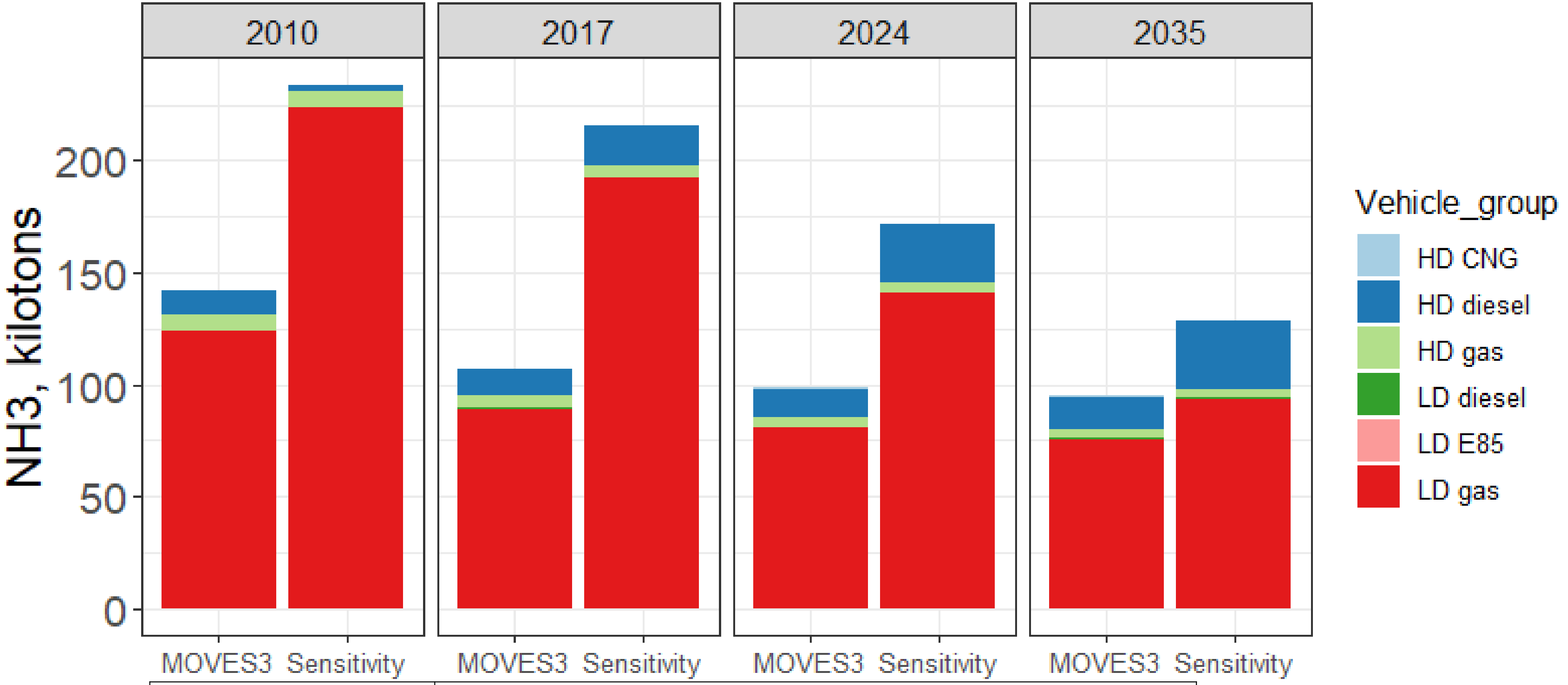
[2] Chassis model year

Heavy-duty diesel NH_3 Rates for Sensitivity Analysis

- Converted fuel-based rates from Caldecott Tunnel study (Preble et al. 2018) to time-based rates
  - Used model year groups from the Caldecott study
  - Used MOVES heavy-duty fuel consumption rates to convert to time-based emission rates
  - No aging effect applied
  - Applied in MOVES run to estimate distance-based rate (g/mile)
- Sensitivity rates based on Caldecott tunnel
  - Lower than MOVES3 for pre-MY 2010 rates
  - Significantly larger than MOVES3 for MY 2010+
    - Variation in MY 2010+ due to improved fuel economy, and sales of non-SCR equipped diesel trucks
    - MY 2010-MY 2018 NH_3 rates applied to MY 2019 and later heavy-duty diesel vehicles



Onroad national emissions inventory impact



	Ratio (Sensitivity Case/MOVES3)			
	2010	2017	2024	2035
HD diesel	0.3	1.5	2.0	2.1
LD gas	1.8	2.2	1.7	1.2
All non-diesel	1.8	2.1	1.7	1.2
Total	1.7	2.1	1.7	1.3

Replaced the MOVES3 emission rates with the sensitivity rates and ran MOVES for the entire U.S

All other inputs left as MOVES3 defaults

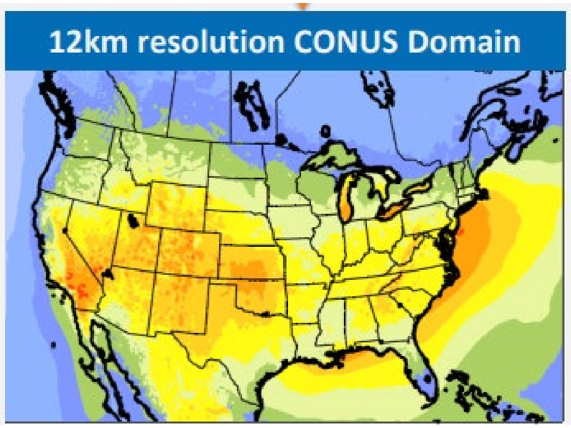
AQ Model Run Methods/Description

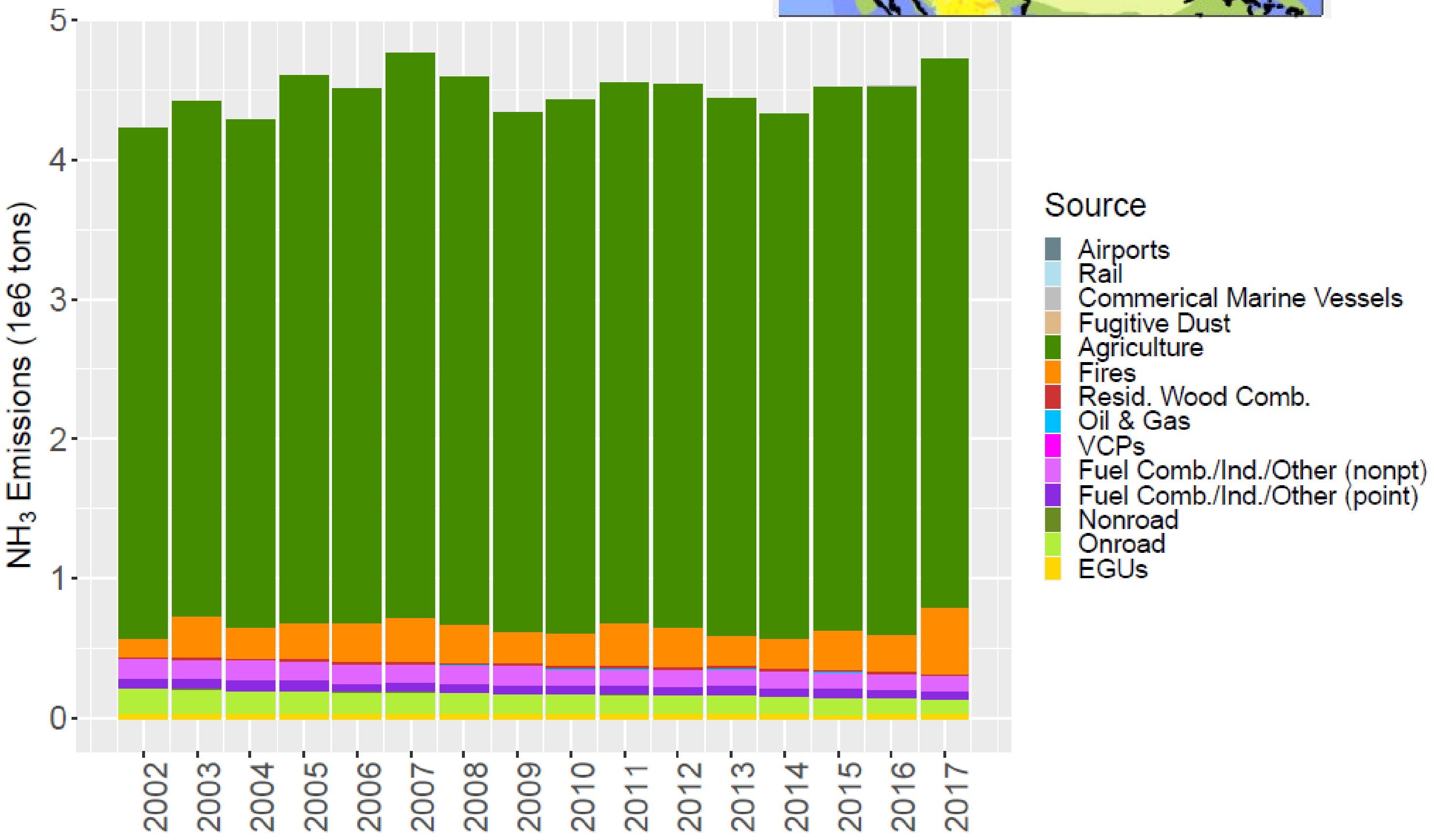
Base-Case: Annual 2017 Conterminous US simulation from the **E**PA's Air **QUA**lity **T**im**E** **S**eries (EQUATES) project

- www.epa.gov/EQUATES
- WRFv4.1.1 and CMAQv5.3.2
- 12 km horizontal resolution
- Chemistry: Carbon Bond 6, Aero7
- Deposition: Surface Tiled Aerosol-Gas Exchange (STAGE) Module with Bidirectional Ammonia (NH_3 BiDi) transfer.
- Emissions: 2017 NEI primary base year.
  - Onroad and nonroad inventories based on MOVES3 except for CA (EMFAC2017) and TX (TexN2 model)

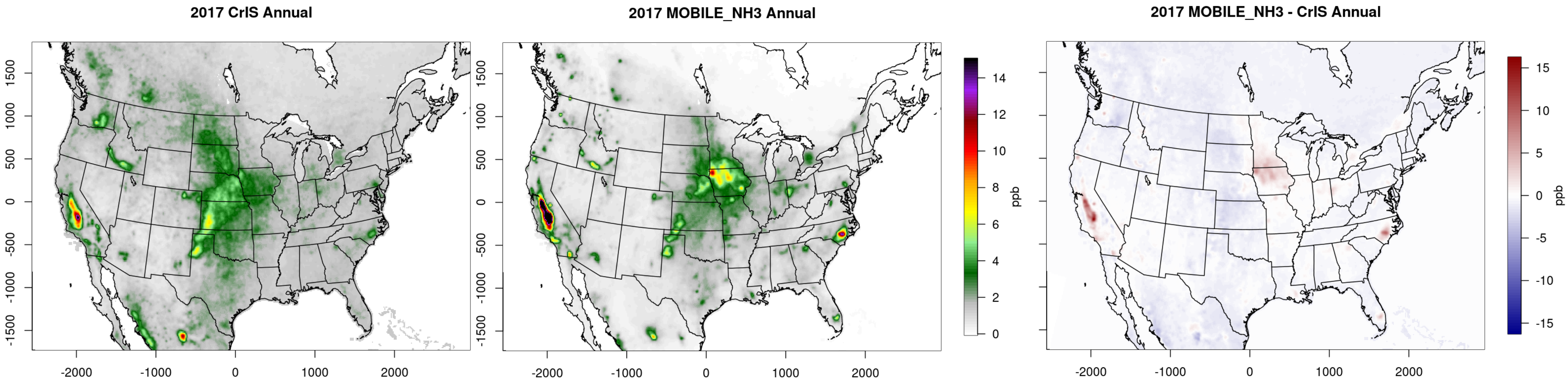
2017 Mobile NH_3 Sensitivity Case:

- Scaled onroad diesel NH_3 emissions by factor of **1.54**
- Scaled onroad non-diesel NH_3 emissions by factor **2.08**
- All other input data and parameters are held constant.





2017 Mobile NH_3 Compared to **Cr**oss-trac **I**nfrared **S**ounder (CrIS) Observations

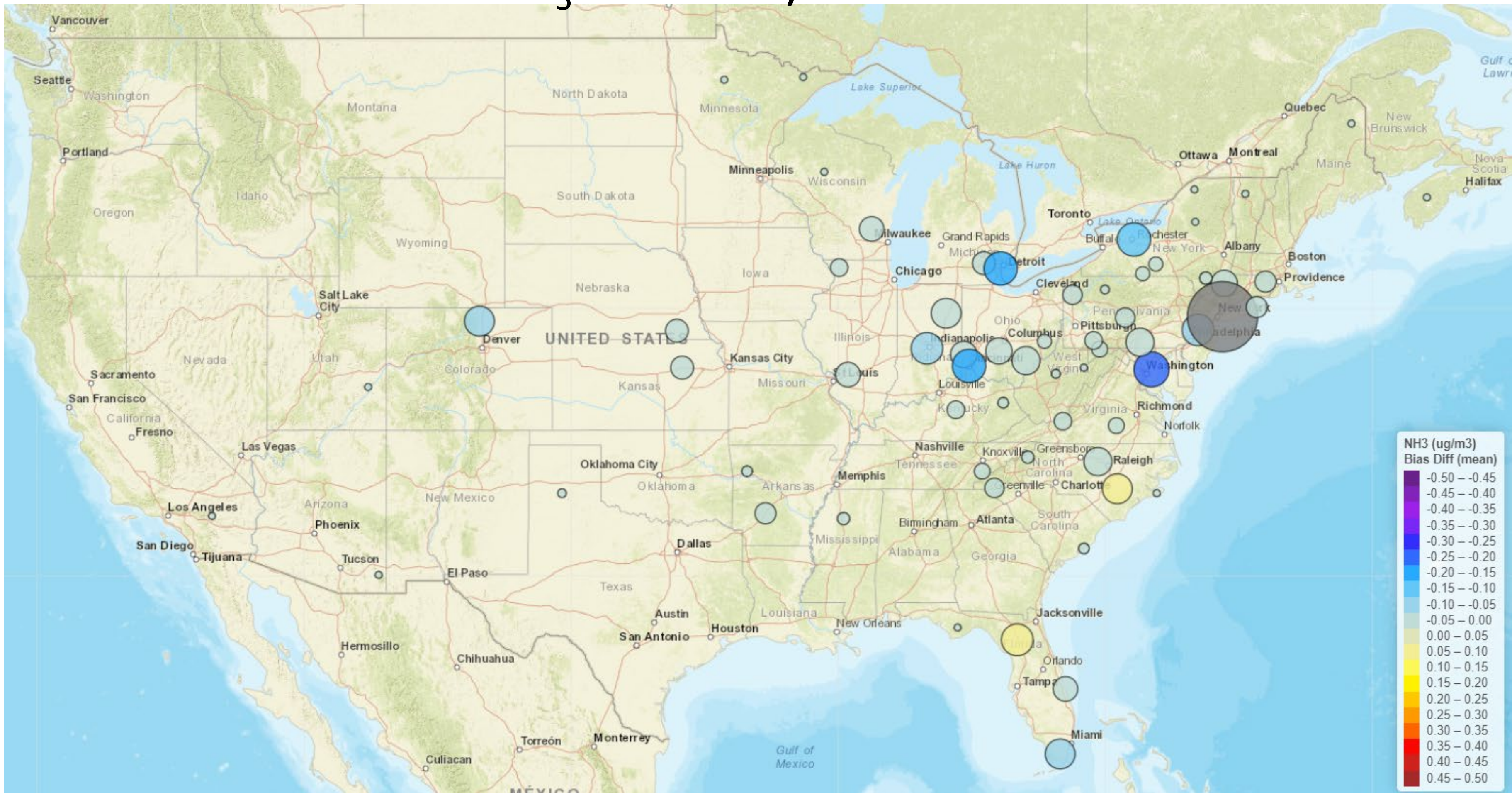


- 2017 Annual CMAQ and CrIS observations were matched in space and time plotted above
- Spatial patterns are similar
  - CMAQ overestimates concentrations in agricultural areas (typically several ppb) and underestimated concentrations elsewhere (typically less than 1 ppm)
  - CrIS overpass at 13:00 local time misses the mobile NH_3 emission peaks during morning and evening commutes

National NH_3 Impacts (2017 Annual Mean)

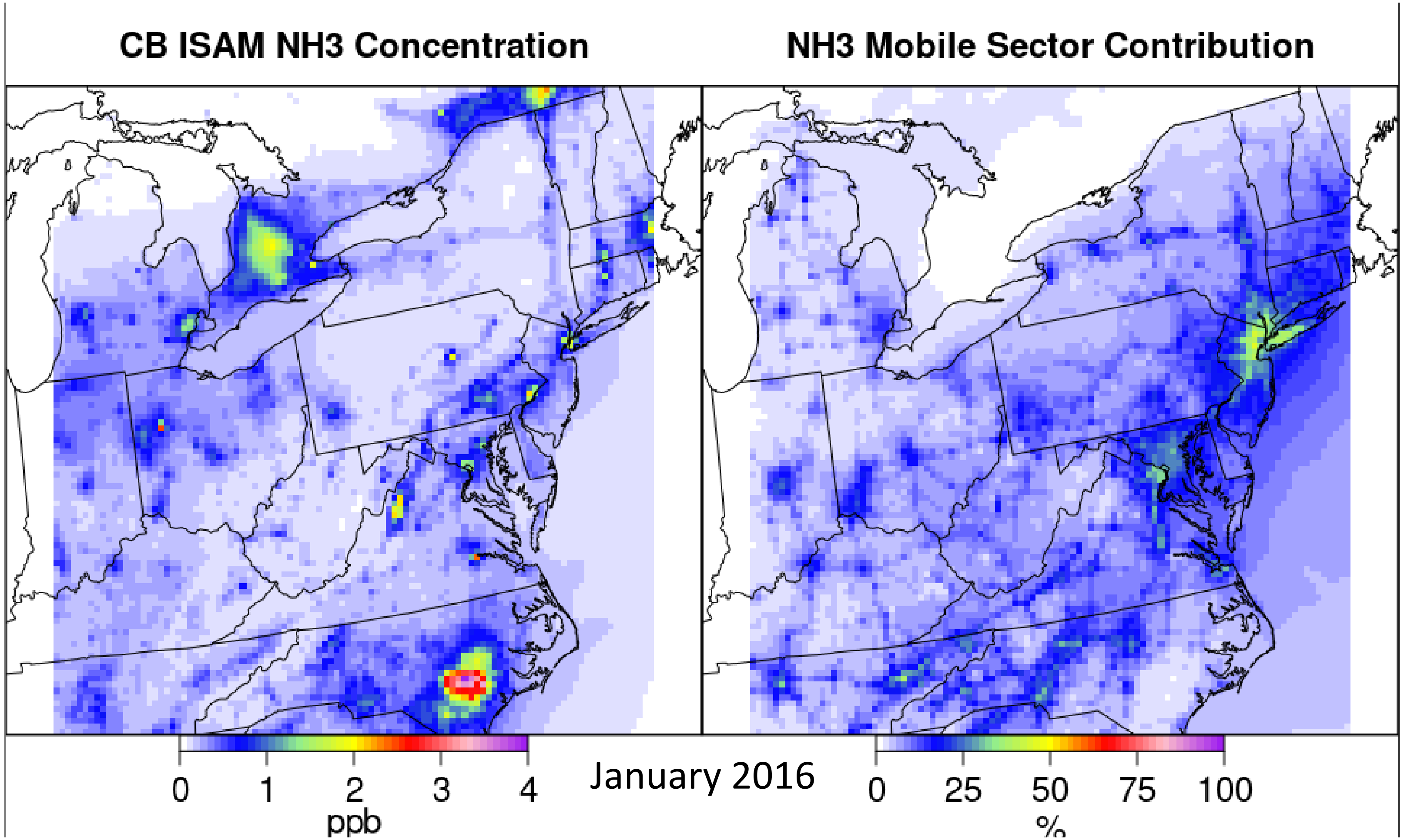
- Modeled NH_3 concentrations compared to the **Ambient Ammonia Monitoring Network (AMoN)**
- Use of sensitivity-case onroad NH_3 emission factors reduce model bias at AMoN sites
    - Annual bias and error are reduced, by up to 1 µg m^{-3}, at 96.8% and 93.7% of AMoN sites, respectively
- Cool colors and grey indicate reductions in biases and warm colors indicate increases in biases
- The size of the circle corresponds to the magnitude of the change in bias

Mobile NH_3 Sensitivity bias – Base bias



Mid-Atlantic Case Study

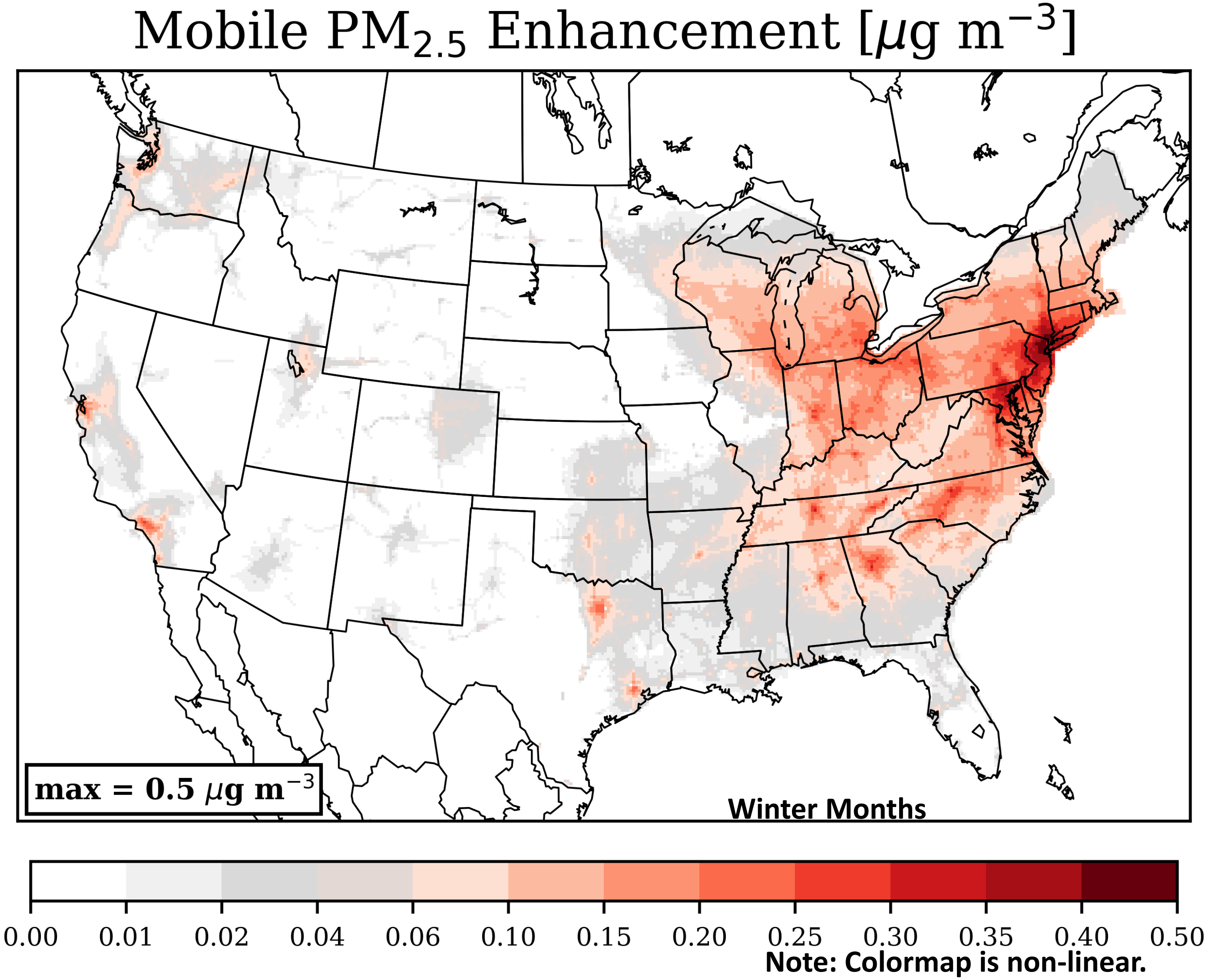
- CMAQ with Integrated Source Apportionment Method (ISAM) was run for 2016 for a Mid-Atlantic Subdomain
  - EQUATES inputs used
  - Multiple EGU, Mobile, Marine, and Agriculture sectors were considered
- Mobile NH_3 was a substantial fraction of the ambient NH_3 (up to 50% in January and 35% in July) along the I-95 Corridor
- NH_3 emission factors based on RSD and tunnel measurements increase this contribution from approximately 5% to 10% of the total ambient concentration
  - Reduced model bias and error by 10% and 4% in January and July respectively



Impacts on Ambient Air Quality

$PM_{2.5}$ Enhancement: Difference between sensitivity simulation and base-case.

- Increases were dominated by NH_4NO_3 during cooler months.
- Largest enhancements were in NYC-region, followed by mid-Atlantic/upper Midwest and other urban cores.
- Population-weighted state-wide increases in NJ/CT/NY region during cooler months: 0.3 – 0.4 μg m^{-3}. Increases < 0.1 μg m^{-3} during warm months.



AQ Sensitivity Conclusions

- Sensitivity-case NH_3 emission factors for onroad gasoline and diesel sectors roughly doubled overall mobile NH_3 emissions in CY2017
  - Note: Differences between MOVES3 and sensitivity-case mobile emissions vary across calendar years and fuel types
- Increases predicted urban NH_3 ambient concentrations by up to 2.3 ppbv in winter and 3.0 ppbv in summer. For winter, this could be up to 50% increase in urban NH_3.
- Resulting $PM_{2.5}$ enhancements in Winter are up to 0.5 μg m^{-3}.

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