MICHIGAN DEPARTMENT OF ENVIRONMENT, GREAT LAKES, AND ENERGY

# Key Findings and Policy Implications from the MOOSE Study

Jay Olaguer
Assistant Director
Air Quality Division

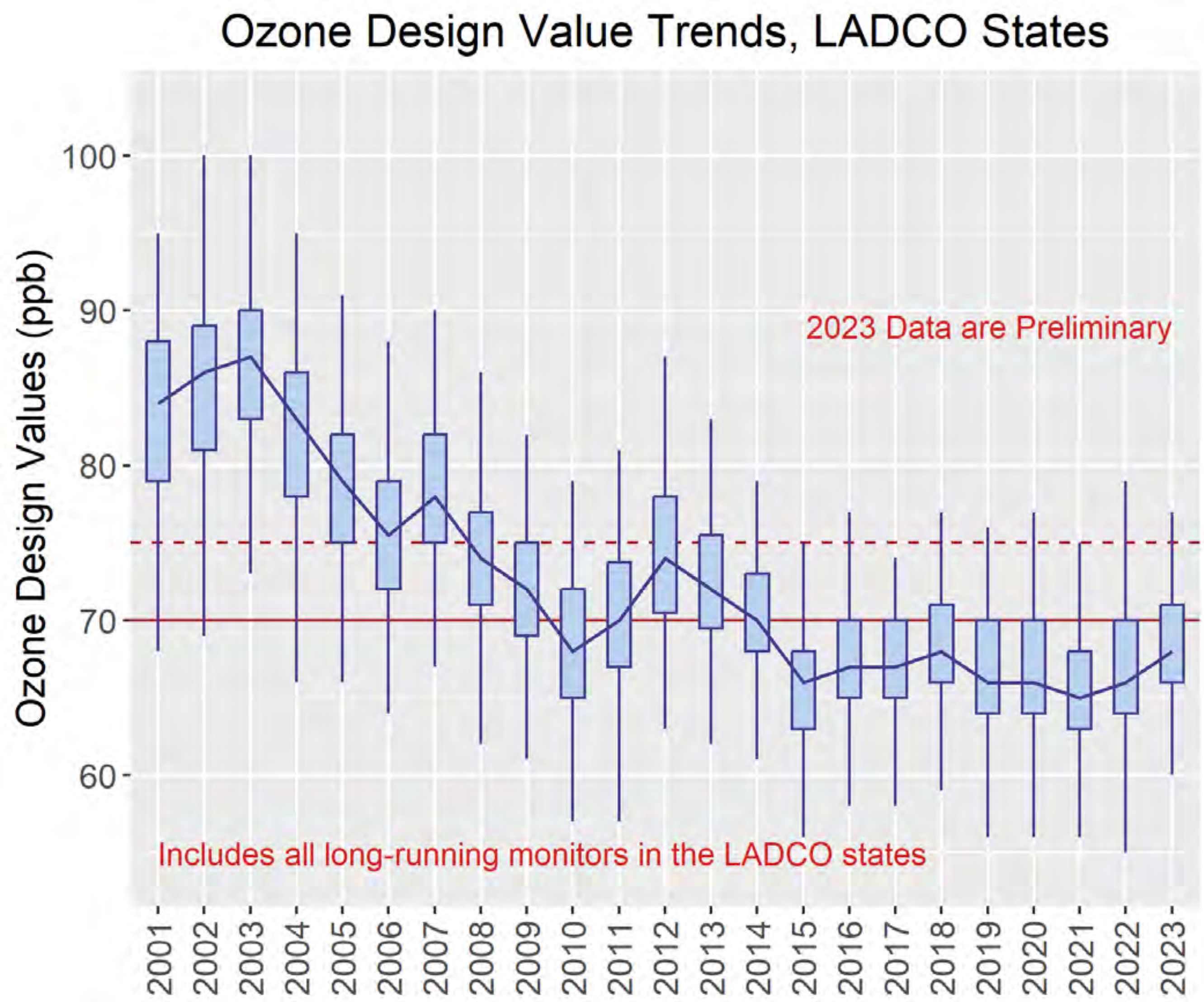
April 22, 2024

# Motivation

- ***EGLE faces challenges in maintaining the National Ambient Air Quality Standard (NAAQS = 70 ppb, 8 h) for ozone ($O_3$):***
  - Downward $O_3$ trends across Region 5 states flattening despite long-term reductions in precursor ($NO_x$, VOC) emissions
  - Transport of pollution due to lake breeze effects
  - Climate change and El Niño impacts on ozone
  - Inadequacy of Reasonably Available Control Technologies (RACT) and difficulty of finding effective alternative strategies
- ***Immediate Needs***
  - SE Michigan is now a maintenance area for which additional controls will be triggered once certain $O_3$ thresholds are passed
  - The 2023 ozone season reflects the likelihood that contingency controls will eventually be required
  - MOOSE helps identify appropriate controls for consideration



EGLE




***Courtesy of Lake Michigan Air Directors Consortium (LADCO)***

# Michigan-Ontario Ozone Source Experiment (MOOSE)

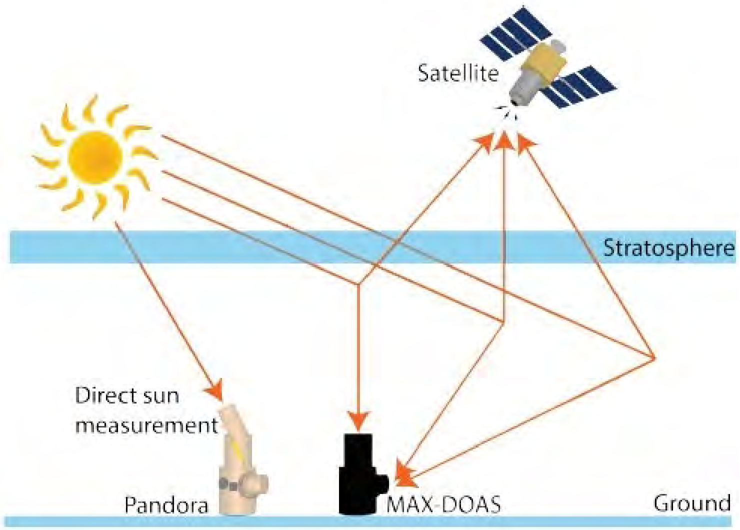
- ***International / Intergovernmental Collaboration***
  - **United States:** Michigan EGLE, USEPA, NASA, NSF, U.S. Forest Service, U.S. Department of Energy
  - **Canada:** Environment and Climate Change Canada (ECCC), Ontario Ministry of Environment, Conservation, and Parks (MECP)
- ***Governance***
  - ECCC served as official convenor
  - Executive Committee consisting of government reps developed an official Science Plan

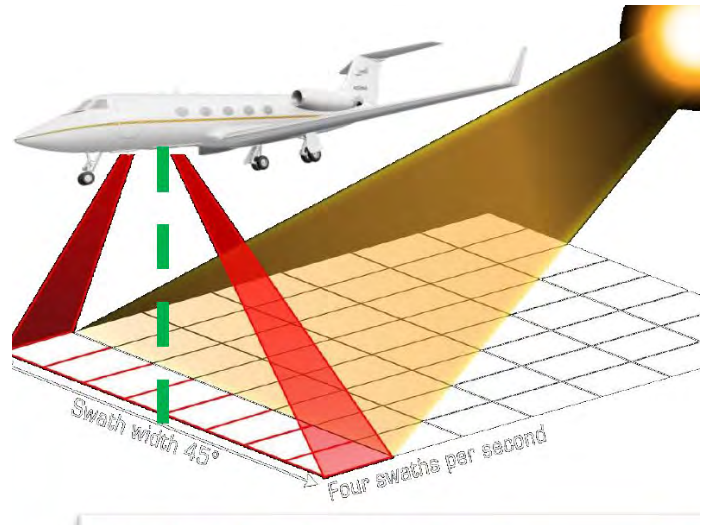


EGLE

# MOOSE Science Plan Sub-Experiments

- ***Great Lakes Meteorology & Ozone Recirculation (GLAMOR)***
  - Physics and chemistry of $O_3$ and precursor transport
- ***Chemical Source Signatures (CHESS)***
  - Fingerprints of industrial facilities and source regions (U.S. EPA-approved Quality Assurance Project Plan)
- ***Methane Releases from Landfills and Gas Lines (MERLIN)***
  - Quantification of fugitive emissions of methane and their impact on ozone chemistry



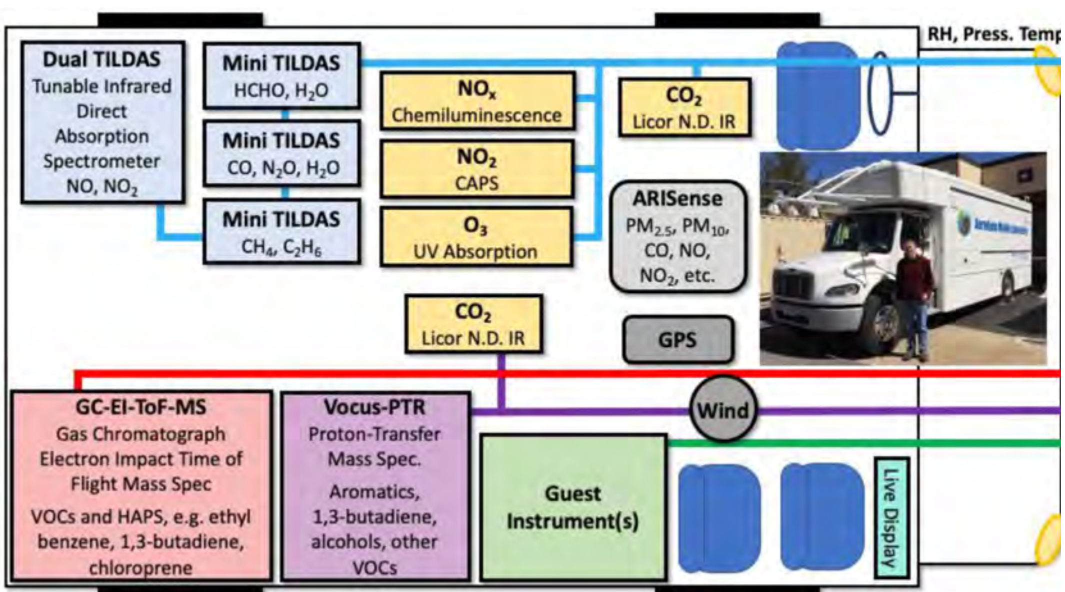




HCHO, $NO_2$

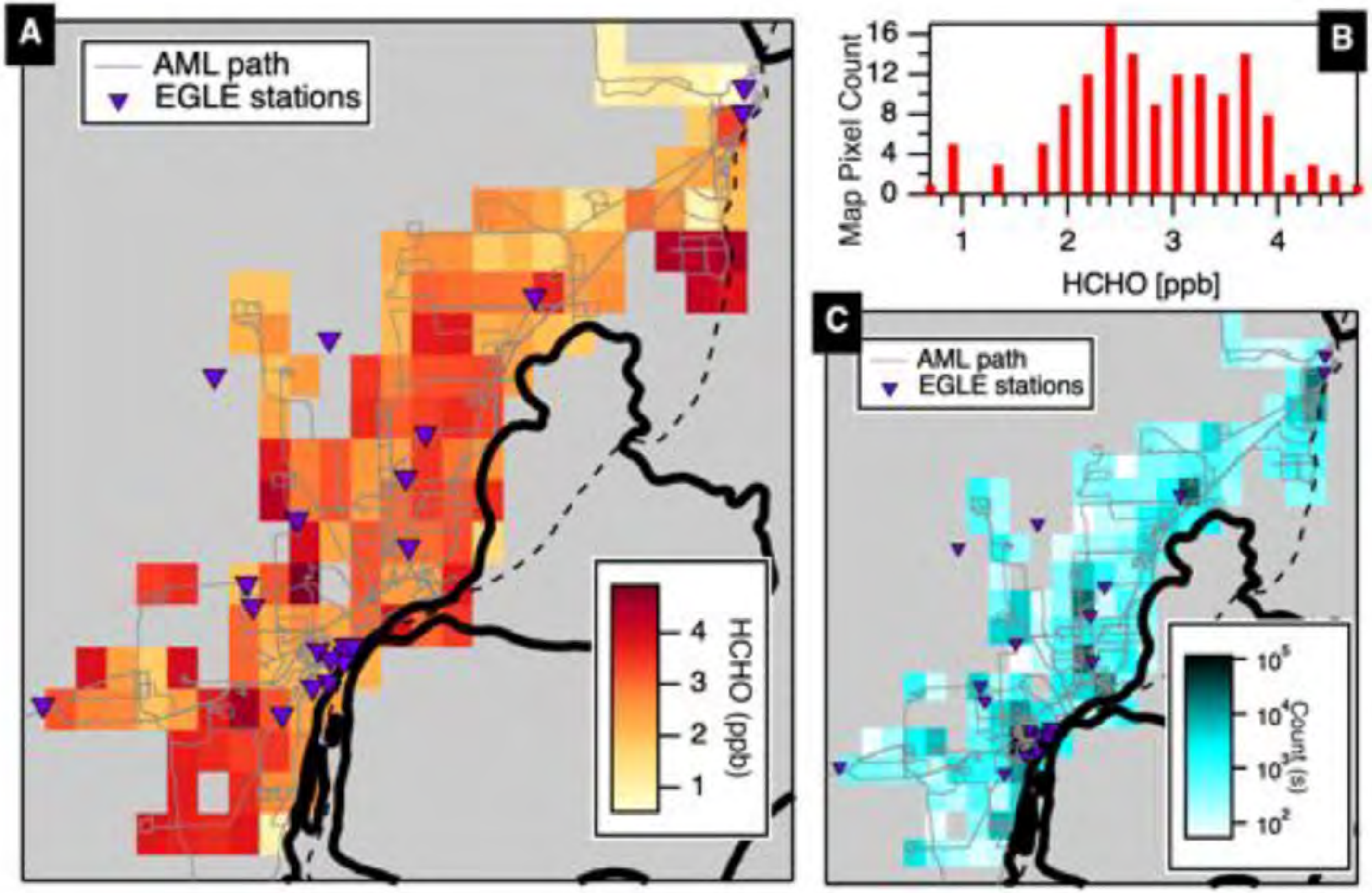
HCHO, $NO_2$

HCHO, $NO_x$, $CH_4$, CO, $O_3$

VOCs




# MOOSE Chemical Instrument Platforms

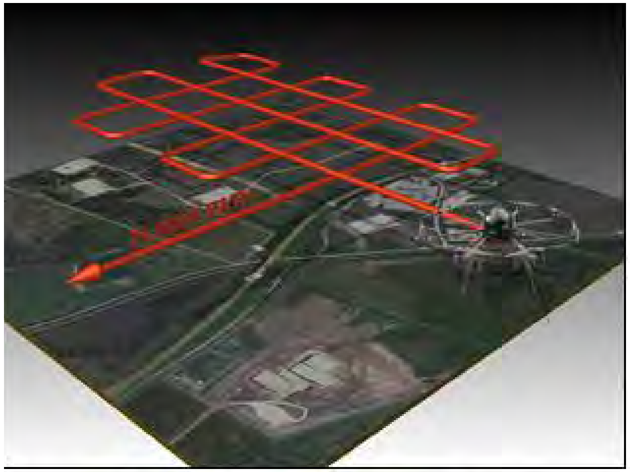


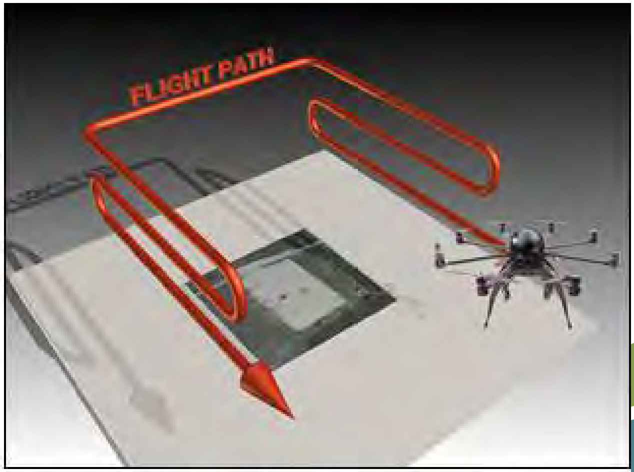

Formaldehyde (HCHO) average concentrations over Southeast Michigan. A histogram of average map pixel concentrations is shown, along with a map of data point counts. Traffic influences were filtered based on enhancements of CO and $NO_x$.

***(Courtesy of Aerodyne Research, Inc.)***

# Real-Time $CH_4$ Measurements at Landfills

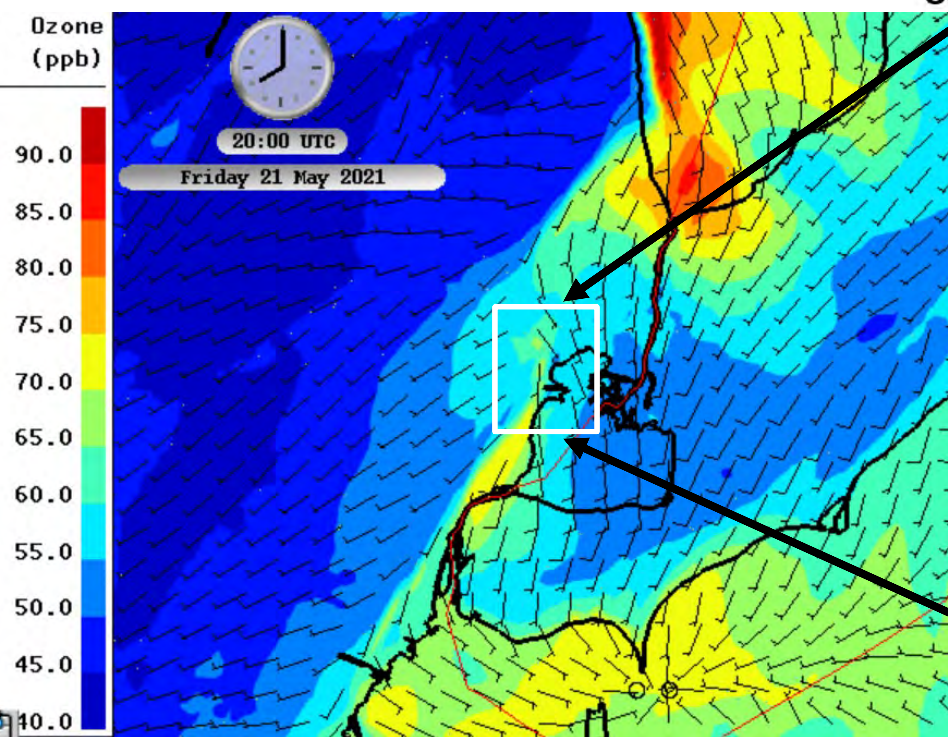
- Mobile IR Cavity Ringdown Spectrometry (Picarro) by University of Michigan Pollution Assessment Lab (MPAL)
- Two drone measurement platforms:
  - ❑ Aegis IEV2 drone with BlueHalo WP-V2 UAS Weather Payload
  - ❑ DJI M600 UAV with a Scentroid DR1000 flying lab encasing a Tunable Diode Laser Spectrometer (2 Hz, 0.4 ppm MDL)

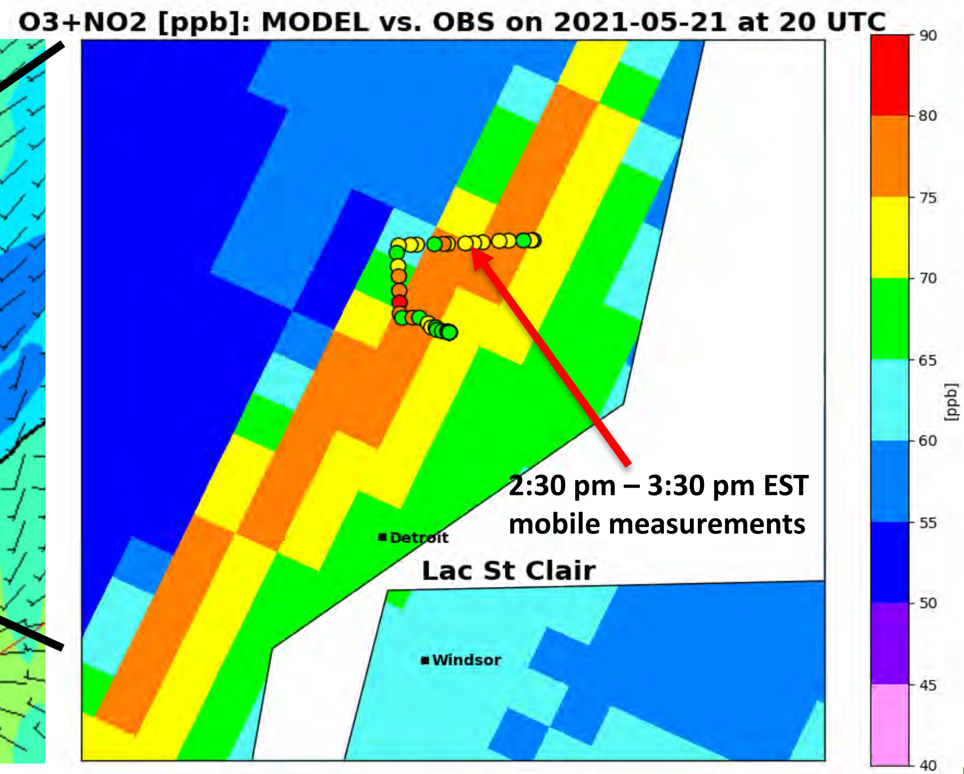




# High Resolution GEM-MACH Model Predictions vs. Observations

### *21 May 2021 Lake Breeze Front*



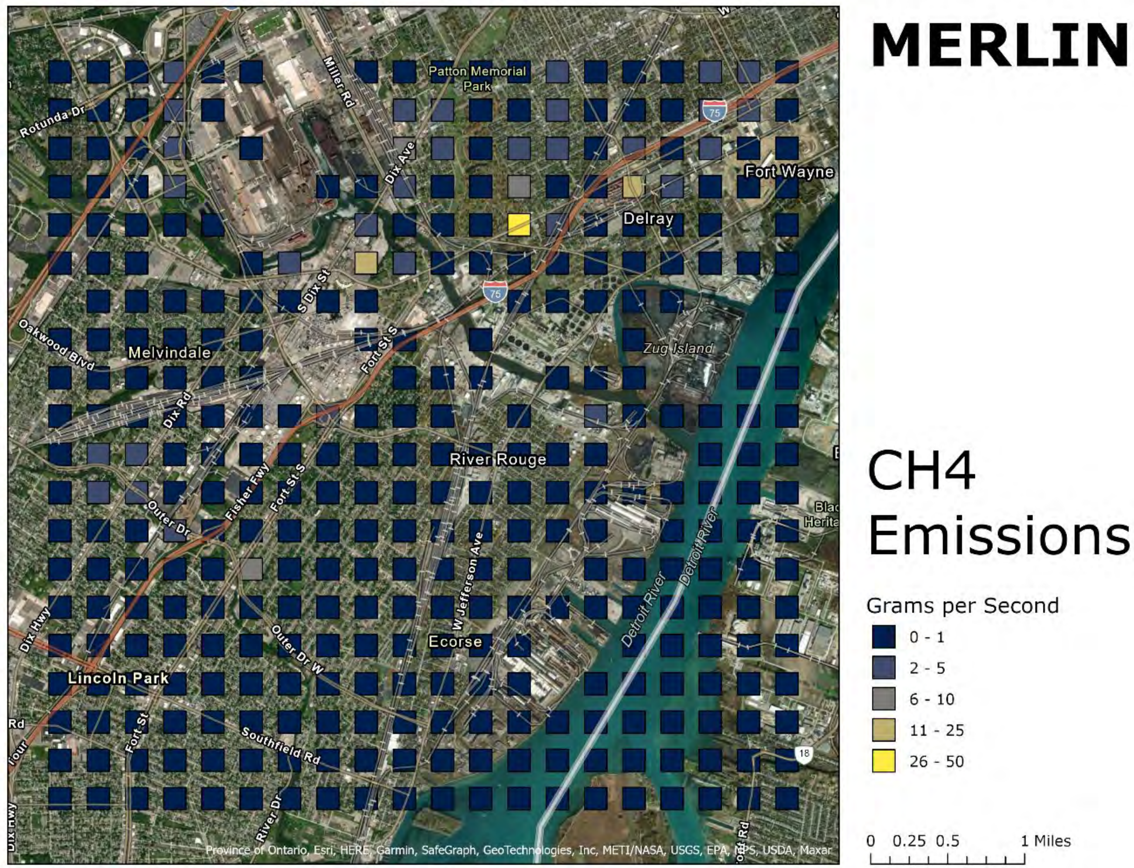




**Aerodyne lab measured high $O_x = O_3 + NO_2$ in narrow band consistent with GEM-MACH modeled $O_3 + NO_2$.**



EGLE

# Inverse Modeling of Pipeline Emissions




***Inverse modeling of methane leaks performed with MicroFACT model based on Aerodyne infrared laser measurements.***



EGLE

# Key Finding #1: SEMI is VOC-limited

- $O_3$ production in an area is classified as: a) $NO_x$-limited (most sensitive to $NO_x$ controls), b) VOC-limited (most sensitive to VOC controls), or c) transitional (between VOC and $NO_x$ sensitivity).

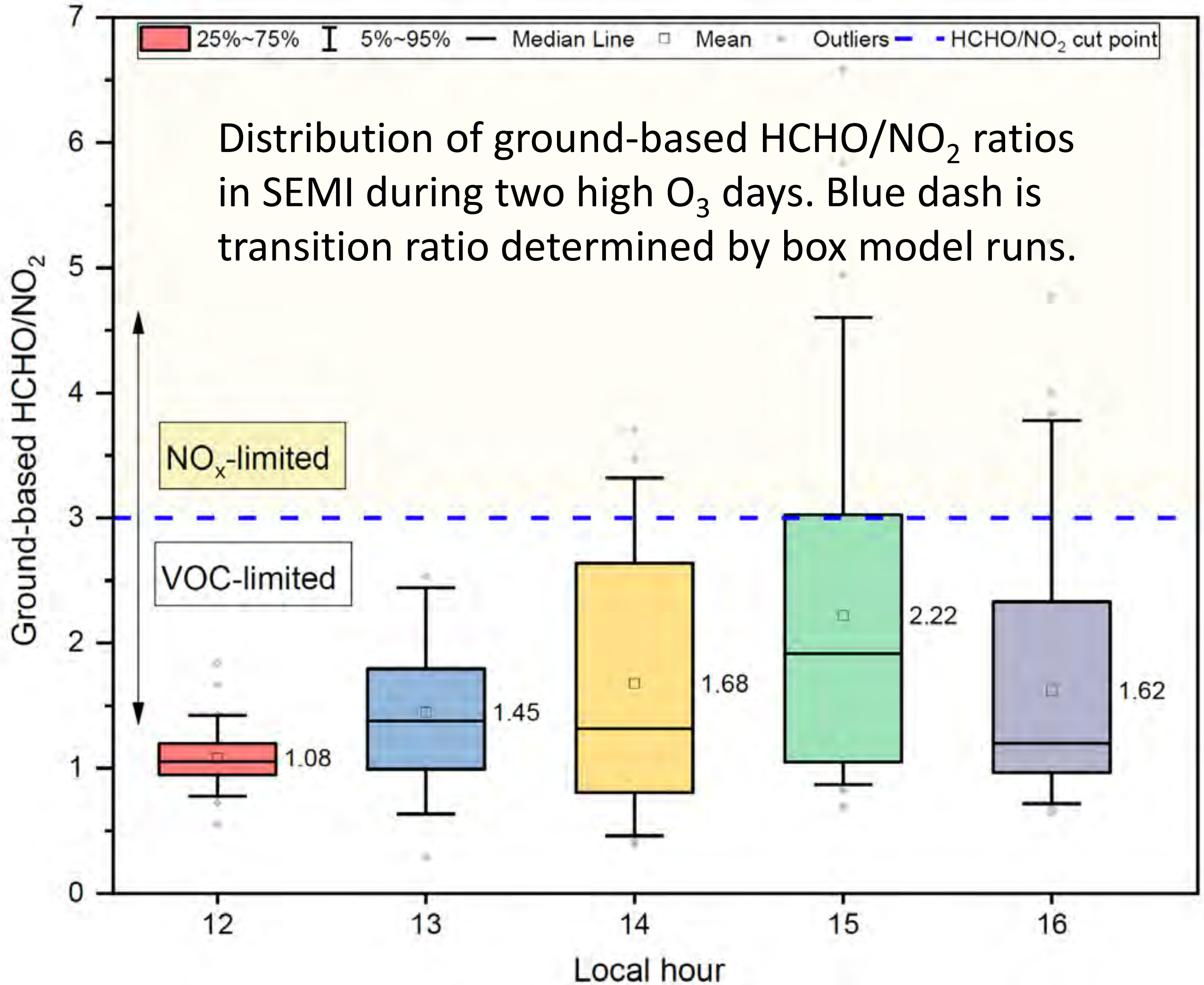
- Based on a compilation of previous studies, LADCO identified SEMI as in a transitional regime. The studies on which LADCO based this conclusion suffered from technical shortcomings related to coarse resolution in either models or (satellite, ground-based) observations.

- MOOSE generated a large set of data near the surface at high temporal and spatial resolution. A team of university scientists used the observed ratio of formaldehyde (HCHO) to $NO_2$ in ambient air and chemical box model simulations utilizing an extensive set of speciated VOC, $NO_x$, and ozone measurements to conclude that, contrary to LADCO's conclusion, SEMI is VOC-limited.



EGLE

# VOC vs $NO_x$ Sensitivity




***Courtesy of Yaoxian Huang, Wayne State University***



EGLE

# Key Finding #2: Formaldehyde is underestimated in models

- HCHO may be the most important VOC to control, because it significantly affects the initial pool of atmospheric radicals available to other VOCs. This helps determine an area's ozone productivity.

- Ratios of HCHO to CO emissions in combustion sources are grossly underestimated in air quality models relative to stack and field tests. Correction of this deficit may result in higher ambient HCHO concentrations in models in better agreement with observations.

- Inverse modeling of HCHO based on Aerodyne mobile lab measurements during MOOSE indicates emissions greater than 1 US ton/yr from several industrial facilities in Dearborn/SW Detroit (over 2 orders of magnitude greater than reported emissions).

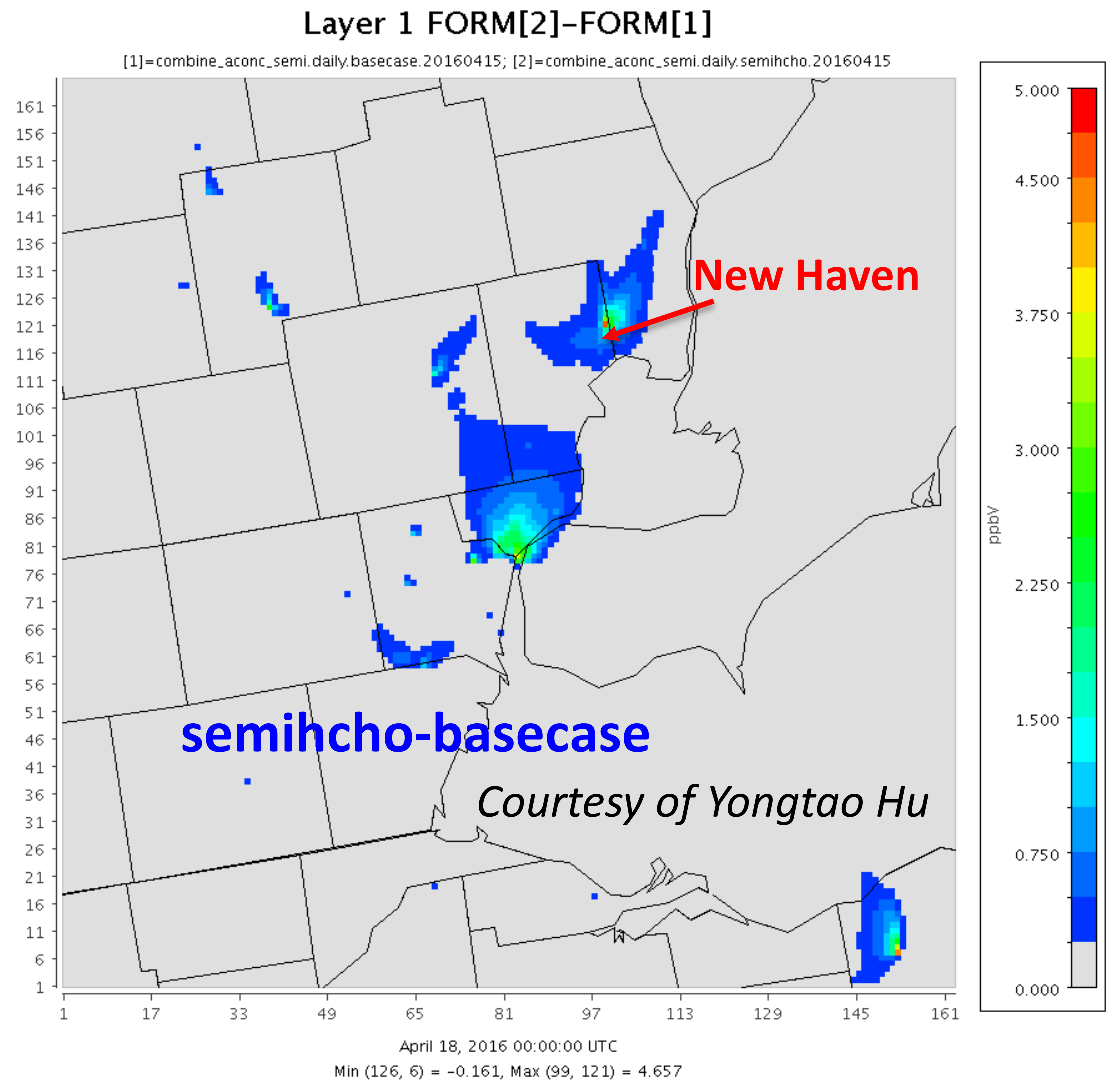
- HCHO controls are five times more effective by weight than $NO_x$ controls in reducing ozone at monitors exceeding the NAAQS in SEMI.

# 1.3-Δkm SEMI CAMx Run 4/18/2016

# ΔHCHO due to Added HCHO Emissions

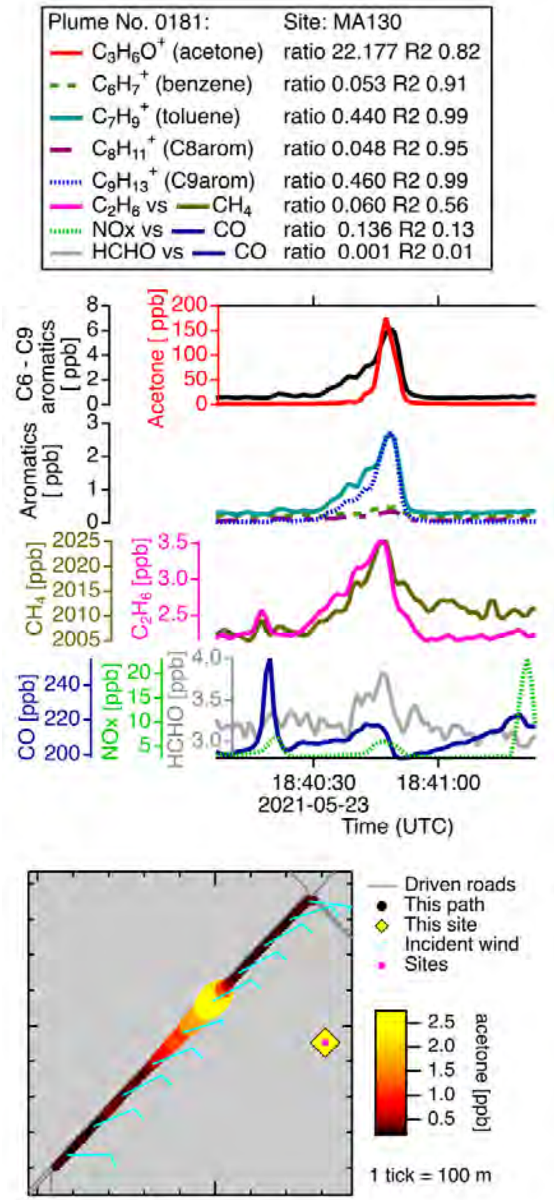
**MOOSE Measurements**

| New Haven Sample Date | 24-h HCHO (ppb) |
|---|---|
| 6/24/2021 | 5.46 |
| 6/25/2021 | 5.22 |
| 6/26/2021 | 5.05 |
| 6/27/2021 | 5.65 |
| 6/28/2021 | 5.62 |



# Key Finding #3: Industrial solvent VOCs are worth controlling

- Solvents are part of a subset of VOCs referred to as Volatile Chemical Products (VCPs). Estimates of VCP emissions from solvent use applications have recently increased. The U.S. EPA has updated U.S. solvent emissions in the 2020 National Emissions Inventory.
- During MOOSE, significant enhancements of VOCs such as acetone, aromatics, and chlorinated solvents were found downwind of industrial facilities, including auto makers, chemical waste sites, and coatings or cleaning product manufacturers, possibly indicating emissions from paint, coatings, and solvent use.
- Fugitive emissions of solvents from industrial manufacturing operations are very difficult to quantify and may still be considerably underestimated due to the lack of reliable emission factors.
- A 2021 GEM-MACH simulation with updated U.S. VCP emissions yielded similar maximum ozone reductions in Detroit (~0.6%) for 10% emission reductions in either solvents or mobile source $NO_x$.




Chemical fingerprint (species ratios to the sum of aromatics) for an industrial coatings (automotive, pipeline, electrical insulation) manufacturing facility (top). Time traces for various tracers are shown (middle). Map (bottom) shows concentration over the driven path.

*After Yacovitch et al. (2023)*



EGLE

# Key Finding #4: Large $CH_4$ leaks may significantly enhance $O_3$ formation

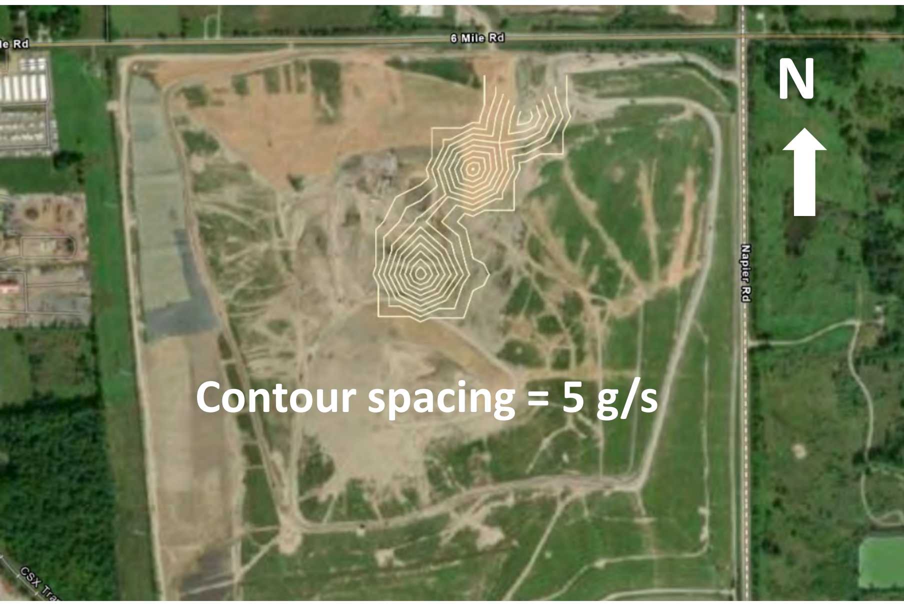
- Large volumes of methane ($CH_4$) released from landfills and leaking natural gas pipelines may compensate for methane's low reactivity in enhancing ozone formation by local sources of VOC and $NO_x$.

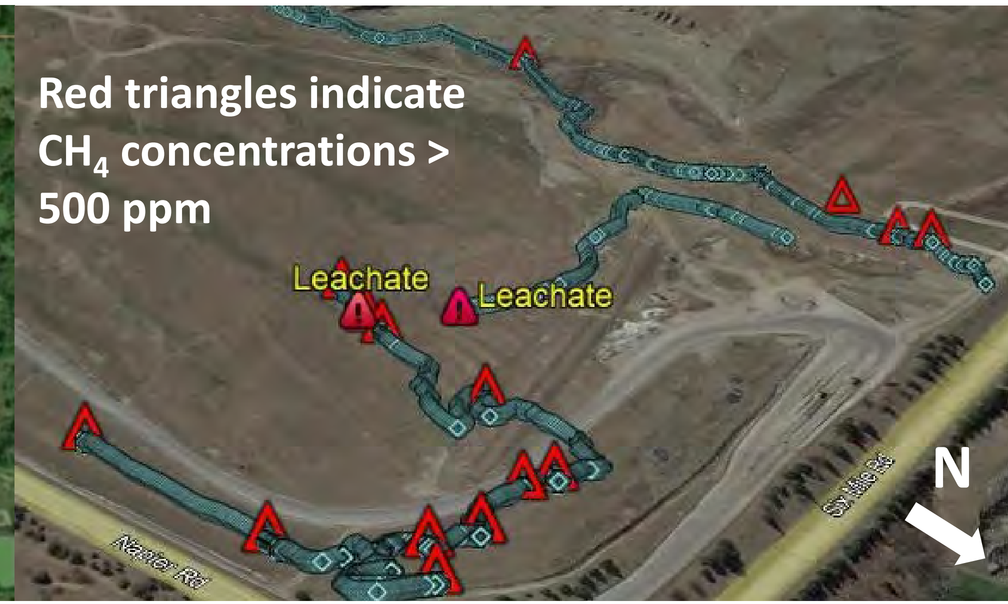
- Drone and mobile lab measurements of methane at two landfills in SE Michigan resulted in estimates of total methane emissions of ~500 kg/h (roughly the median of landfill measurements in the California Methane Survey). Significant emissions from both the active face and leaking gas collection systems were inferred.

- MPAL made 1-s near-ground $CH_4$ measurements collected on 20 days while driving 1100 km on surface streets in Detroit. Detected 534 distinct methane peaks, equivalent to ~1 peak per 2 km traveled. Inverse modeling of a persistent natural gas pipeline leak in SW Detroit based on Aerodyne laser measurements yielded estimated emissions of ~200 kg/h.

# Inverse Modeling of Landfill Emissions

**Total Methane Emissions = 555 kg/h**




**Drone-Inferred Emissions**

**Handheld Surface Emissions Monitoring**

***Inverse modeling performed with a Gaussian plume model with complex terrain features.***



EGLE

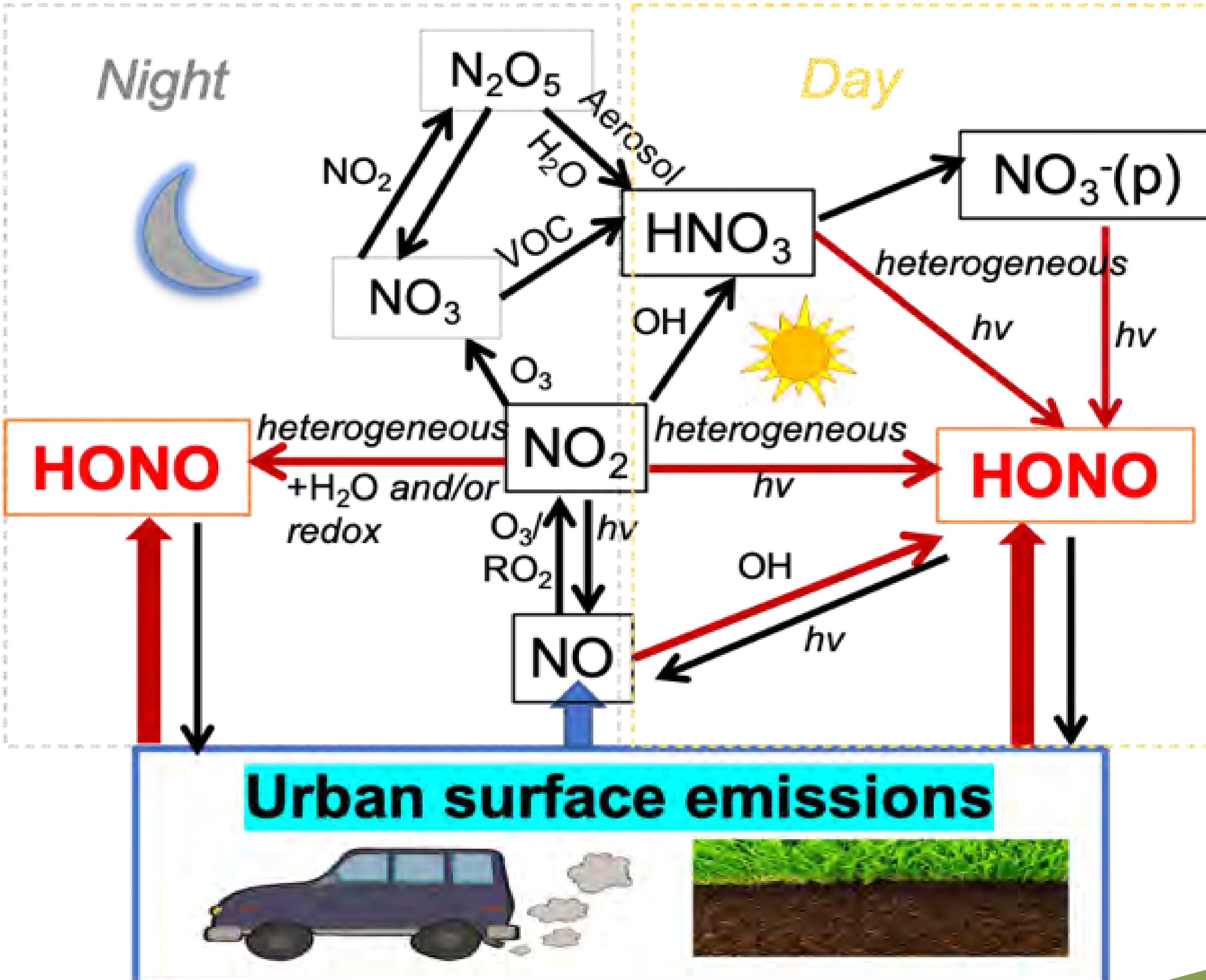
# Key Finding #5: Reactive nitrogen is poorly understood

- HONO and $HNO_3$ are important temporary reservoirs of $NO_x$. In addition to being a $NO_x$ reservoir, HONO is also a radical precursor on par with HCHO so it can influence local ozone productivity. $HNO_3$, on the other hand, is much longer-lived and can re-release sequestered $NO_x$ back to the atmosphere after traveling long distances downwind of the original $NO_x$ sources.

- Isotopic reactive nitrogen measurements were conducted at two sites during MOOSE: an urban Detroit site (Trinity) which is greatly influenced by emissions from motor vehicles, urban soils, and industrial sources, and a suburban site (New Haven) ~40 miles north of Detroit and typically downwind of major urban emission sources during ozone episodes.

- Local concentrations of HONO were found to be much larger than concentrations of nitric acid, with [HONO]/[$HNO_3$] in the range of 1–21 (mean = 3), contrary to the predictions of standard air quality models.

# Reactive Nitrogen Measurements



# Key Finding #6: Mobile sources now dominate PM2.5 unlike before

- University of Michigan applied Positive Matrix Factorization to measurements from 2016 to 2021 at three sites with long-term PM2.5 levels from 8.63 to 10.83 $\mu g/m^3$.
- Most PM2.5 was due to mobile sources (marked by elemental and organic carbon with some K+) representing 33–44% of PM2.5, followed by secondary sulfate at 24–29%, and secondary nitrate at 17–22%.



EGLE

# $O_3$ Control Strategy Implications

- Although $NO_x$ emissions are important in regional ozone formation, **local ozone in SEMI is VOC-limited**, except in heavily vegetated areas to the west.
- Controls of organic species such as **HCHO, industrial solvents, and $CH_4$** from landfills and large pipeline leaks are new strategies that should be investigated and possibly implemented. **Their emissions are underestimated in official inventories**.
- Pound-for-pound, **HCHO controls are 5 times more effective than $NO_x$ controls** in reducing ozone design values at the key monitoring sites with historical NAAQS exceedances.
- Possible control strategies to consider:
  - Oxidation catalysts for stationary engines
  - Flare minimization or efficiency improvements
  - Advanced leak detection and repair to minimize fugitive emissions.



EGLE

# Other Lessons

- Michigan should work with other states and the U.S. EPA to promote the continued update of national formaldehyde, industrial solvent, and methane emissions inventories.
- Need to improve modeling practice to better assess the impacts of control strategies, including the use of finer resolution and the updating of reactive nitrogen chemistry in air quality models. This will help improve assessments of both local ozone formation and long-range transport of ozone and its precursors.
- The insights derived from MOOSE were made possible by advanced science and technology, which is needed to address more difficult air quality issues than in the past.

# MOOSE Special Issue

- The peer-reviewed journal, *Atmosphere*, has a special issue devoted to MOOSE at: https://www.mdpi.com/journal/atmosphere/special_issues/Michigan_Ontario_Ozone.

- There is a total of 13 papers available as part of the special issue, including an Overview article summarizing the field study.



EGLE