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The Michigan–Ontario Ozone Source Experiment (MOOSE): An Overview

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Abstract: The Michigan–Ontario Ozone Source Experiment (MOOSE) is an international air quality field study that took place at the US–Canada Border region in the ozone seasons of 2021 and 2022. MOOSE addressed binational air quality issues stemming from lake breeze phenomena and transboundary transport, as well as local emissions in southeast Michigan and southern Ontario. State-of-the-art scientific techniques applied during MOOSE included the use of multiple advanced mobile laboratories equipped with real-time instrumentation; high-resolution meteorological and air quality models at regional, urban, and neighborhood scales; daily real-time meteorological and air quality forecasts; ground-based and airborne remote sensing; instrumented Unmanned Aerial Vehicles (UAVs); isotopic measurements of reactive nitrogen species; chemical fingerprinting; and fine-scale inverse modeling of emission sources. Major results include characterization of southeast Michigan as VOC-limited for local ozone formation; discovery of significant and unaccounted formaldehyde emissions from industrial sources; quantification of methane emissions from landfills and leaking natural gas pipelines; evaluation of solvent emission impacts on local and regional ozone; characterization of the sources of reactive nitrogen and $PM_{2.5}$; and improvements to modeling practices for meteorological, receptor, and chemical transport models.

Keywords: ozone; air quality; field studies; methane; formaldehyde; ozone; atmospheric chemistry; lake breezes; transboundary transport; emissions inventories; fine particulate matter ($PM_{2.5}$); volatile organic compounds (VOC); nitrogen oxides (NO_x); reactive nitrogen reservoirs

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1. Introduction

In 2015, the United States (US) Environmental Protection Agency (USEPA, Washington, DC, USA) set the US National Ambient Air Quality Standard (NAAQS) for ozone (O_3) at 70 parts per billion by volume (ppb) averaged over 8 h [1]. Attainment of the standard is based on an O_3 design value, defined as the three-year average of the annual fourth highest daily 8 h O_3 maximum concentration measured at a monitoring station. Areas with design values that exceed the NAAQS may be designated by the USEPA as in nonattainment of the O_3 standard within several categories of increasing seriousness ranging from marginal to severe, with corresponding increases in mandatory offsets of new emissions of ozone precursors and progressive reductions of existing emissions, as well as other regulatory

requirements. The federal government of Canada has an even more stringent Canadian Ambient Air Quality Standard (CAAQS) for O_3 of 62 ppb averaged over 8 h, which will be reduced to 60 ppb in 2025 [2]. The statistical form of the Canadian standard is equivalent to that of the US design value.

The Michigan–Ontario Ozone Source Experiment (MOOSE), which took place during 2021 and 2022, was the result of conversations initiated by the Michigan Department of Environment, Great Lakes, and Energy (EGLE) with Environment and Climate Change Canada (ECCC) and the Province of Ontario's Ministry of the Environment, Conservation, and Parks (MECP). In 2007, the Canadian agencies conducted the Border Air Quality and Meteorology Study (BAQS-Met) [3], which revealed the importance of lake breeze phenomena in modulating high O_3 episodes in the southern Great Lakes region (Figure 1). A key discovery of the BAQS-Met study was the enhancement of regional O_3 peaks over the Great Lakes by up to 30 ppb due to lake breeze effects that transported ozone precursors from urban areas and increased their ozone production over water. The pool of high ozone was then available to be transported back to land areas by the lake breeze cycle [3].

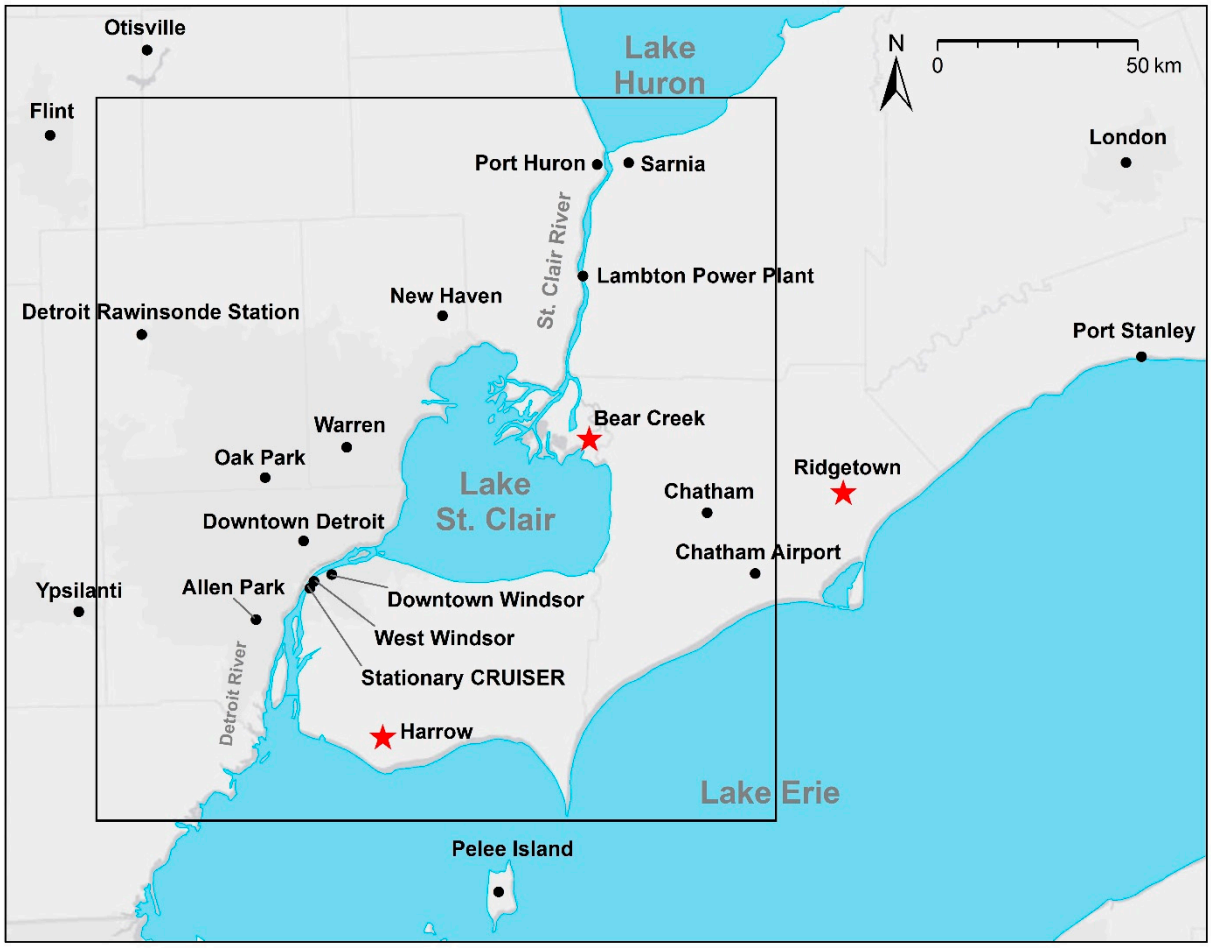

Figure 1. Map of the southern Great Lakes area showing the locations of the major cities in the region and all participating ozone monitoring sites (supersites indicated by red stars) during the BAQS-Met study [3] (modified reproduction licenced under Creative Commons CC BY 3.0). The approximate area that was the subject of the MOOSE field campaign is indicated by the black rectangle.

MOOSE was designed to further address US–Canada Border air quality issues, such as the possibility that southeast Michigan (SEMI), which includes the city of Detroit, would be "bumped up" from its original designation by the USEPA as a marginal O_3 nonattainment area to a moderate nonattainment area with more stringent emissions control requirements. This bump-up occurred very briefly in 2023, until an exceptional event demonstration by

EGLE attributing two days of high O_3 values in June 2022 to a Canadian wildfire was accepted and approved by the USEPA. This enabled SEMI to be re-designated back to attainment of the O_3 NAAQS based on a clean data determination for the three-year period of 2020–2022, as opposed to O_3 design values for 2018–2020 that were the basis for the prior bump-up. However, SEMI remains an ozone maintenance area subject to contingency provisions that would require EGLE to implement new control strategies once a certain ozone concentration threshold has been passed. By providing information on which ozone precursors contribute most to exceedances of the NAAQS, the results of the MOOSE study will help inform the design of new control strategies that may be needed to maintain the current attainment designation for SEMI, as well as to decrease population exposure to other criteria pollutants and to air toxics.

Ozone, fine particulate matter ($PM_{2.5}$), and their precursors are generated both locally and regionally. They can travel hundreds of kilometers, affecting areas far from the emission sources. Long-range transport and transboundary flow of air pollutants play a significant role in Ontario's air quality. During the summer, elevated levels of air pollutants are often associated with distinct weather patterns (i.e., slow-moving high-pressure systems originating from south of the lower Great Lakes) that result in the long-range transport of these pollutants into Ontario from neighboring US industrial and urbanized states during south to southwesterly flow conditions [4].

The Ontario MECP routinely monitors conventional air pollutants, including O_3 and $PM_{2.5}$, at 38 Air Quality Health Index air monitoring stations across the province, including Windsor and Sarnia at the US–Canada Border. Southern Ontario continues to experience persistent elevated ozone levels. In 2020, 13 of the 26 designated CAAQS air monitoring stations exceeded the federal O_3 standard. The provincial Ambient Air Quality Criteria (AAQC) for O_3 (80 ppb averaged over 1 h) was also exceeded several times across southern Ontario in 2020. In southwestern Ontario, when ozone levels are elevated, over 95% of the ozone is attributable to transboundary sources, with the US contributing as much as 40% and the remainder due to the global background [5]. Windsor is the southernmost major city in Ontario and often experiences poor air quality owing to industrial and traffic emissions and transboundary transport of air pollution from neighboring states in the US. The city reported one of the highest $PM_{2.5}$ values in 2020 (i.e., 19 $\mu g/m^3$ for the 24 h CAAQS metric and 7.8 $\mu g/m^3$ for the annual metric) among the 26 designated CAAQS stations in Ontario [5]. Furthermore, Ontario's 24 h AAQC for $PM_{2.5}$ (27 $\mu g/m^3$) was exceeded in Windsor in 2020, although both the ambient levels of $PM_{2.5}$ and its primary emissions to air have decreased over the past 10 years [5].

MOOSE became a reality once EGLE secured funding from the USEPA for the operation of two advanced mobile laboratories to investigate both air toxics and ozone issues in SEMI in a focused study known as the Chemical Source Signatures Experiment (CHESS), which eventually became a sub-experiment of the larger MOOSE campaign (see below). After initial funding was secured by EGLE, both ECCC and MECP committed to adding significant resources to MOOSE, as did federal agencies in the US in addition to the USEPA, including the National Aeronautics and Space Administration (NASA), the US Forest Service, and the National Science Foundation (NSF). MOOSE thus became an international air quality field experiment conducted by federal, state, and provincial governments on both sides of the US–Canada Border.

An Executive Committee was set up with ECCC serving as the official convenor. All official exchanges of the Committee were at the government-to-government level, although contractors hired by a government agency could contribute to deliberations. An early action of the Executive Committee was to develop a Science Plan [6] to guide the deployment of technical resources and define the scientific objectives of MOOSE.

According to the Science Plan, MOOSE would consist of three sub-experiments with the following objectives:

Great Lakes Meteorology and Ozone Recirculation (GLAMOR)

- To understand and successfully simulate complex 3D flows associated with lake breeze circulations;
- To understand and successfully simulate the urban heat island (UHI) and its interaction with the lake breeze;
- To understand and successfully simulate the impact of lake breezes and the UHI on ozone and ozone precursor transport;
- To determine the conceptual picture (mesoscale meteorological patterns and photochemical production locations) for ozone exceedances in the Border region;
- To select representative ozone episodes for each identified mesoscale pattern, which can then be used as model base case periods for future ozone attainment demonstrations;
- To conduct modeling and data analyses in support of an ozone attainment demonstration for SEMI or, if warranted, a US Clean Air Act 179B(b) petition or ozone exceptional event demonstration.

Chemical Source Signatures (CHESS)

- To characterize the ozone precursor signatures at key monitoring stations in the Border region where design values are highest during ozone exceedances in a normal year;
- To characterize emission plumes from point sources in the Border region and their impacts on ozone design values on both sides of the U.S.–Canada Border;
- To develop emission source fingerprints for the most important industrial facilities and source sectors in the Border region;
- To characterize the horizontal variations (including upwind, interior, and downwind concentrations) and vertical gradients of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the Border region;
- To perform receptor modeling, source apportionment, and ozone culpability analyses to improve emission inventories and inform potential control strategies;
- To perform air quality model simulations of potential emission control strategies.

Methane Releases from Landfills and Gas Lines (MERLIN)

- To determine the natural gas leakage rate of pipeline or other infrastructure in SEMI;
- To quantify methane, formaldehyde, and other emissions from landfills in the Border region;
- To determine the contributions of large methane sources to ozone exceedances in the Border region, thereby informing potential control strategies.

2. Experimental Methods

With USEPA funding, EGLE hired Aerodyne Research, Inc. (Billerica, MA, USA) and the University of Michigan (Ann Arbor, MI, USA) to conduct mobile laboratory investigations in SEMI during MOOSE. Prior to the execution of the intensive portion of the field study in the late spring and summer of 2021, EGLE developed a Quality Assurance Project Plan (QAPP) for the CHESS experiment that was approved by the USEPA [7]. USEPA-approved QAPPs are acceptable benchmarks of quality for state regulatory agencies in the US.

The Aerodyne Mobile Laboratory (AML) [8–10] measured O_3 (2BTech), total NO_x (cavity-attenuated phase shift spectrometer coupled to an ozone reactor—Aerodyne CAPS-NO_x); nitric oxide (NO) and nitrogen dioxide (NO_2), methane, ethane, formaldehyde, and CO (4 separate tunable infrared laser direct absorption spectrometers—Aerodyne TILDAS [11–13]); CO_2 (non-dispersive infrared spectrometry); and VOCs (gas chromatography electron impact mass spectrometer—Aerodyne GC-EI-ToF [14] and Aerodyne Vocus proton transfer reaction—mass spectrometer [15,16]). The Michigan Pollution Assessment Laboratory (MPAL) [17,18] run by the University of Michigan deployed cavity ring-down spectrometers (Picarro G2401 and G2204) to measure methane, CO, and CO_2, as well as other instruments to measure O_3 (API 400A) and NO_x (EcoPhysics CLD 700 AL). Both the AML and MPAL were equipped with a global positioning system (GPS) and meteorological

sensors to measure wind speed and direction, surface pressure, temperature, and relative humidity.

MECP also deployed a mobile monitoring platform equipped with GPS and real-time instrumentation for the measurements of O_3, SO_2, NO_x, particulate matter, and VOCs [19]. Specifically, a proton transfer reaction-mass spectrometer (IONICON PTR-ToF-MS 8000) measured aromatics including benzene, toluene, and xylenes, in addition to other VOCs with proton affinities greater than that of water. The platform was used in summer 2021 to measure air pollutant concentrations and chemical fingerprints immediately downwind of industrial sources in Sarnia and Windsor, Ontario.

Mobile measurements were complemented by measurements at monitoring stations in SEMI and Windsor, Ontario during the 2021 intensive portion of the MOOSE study. In SEMI, these included continuous column measurements of formaldehyde and NO_2 by two Pandora UV-visible spectrometers [13,20] operated jointly by USEPA and NASA, specialized isotopic measurements of reactive nitrogen compounds by a university team funded by NSF (as described in Section 3.1), and continuous measurements of boundary layer wind profiles (mini sodar) and mixing height (ceilometer) by the US Forest Service. These were in addition to routine meteorological and chemical measurements by EGLE at regulatory monitoring stations.

In collaboration with ECCC, MECP enhanced air monitoring activities at its Windsor West air monitoring station prior to the MOOSE field campaign in 2021. Additional research-grade instruments deployed at this station included: (1) Xact 625 particulate metal analyzer (Pall Corporation, Port Washington, NY, USA); (2) online gas chromatograph (AMA Instruments GmbH, Ulm, Germany); (3) Vaisala CL51 Ceilometer (Vaisala, Helsinki, Finland); (4) Pandora spectrometer [20]; (5) Vaisala Weather Transmitter WXT520 (Vaisala, Helsinki, Finland). All enhanced air monitoring activities are currently ongoing at Windsor West station except that the ceilometer was decommissioned in March 2023.

During the MOOSE intensive field campaign in 2021 (Figure 2), ECCC conducted high-resolution, real-time meteorological and air quality forecasts to guide the deployment of mobile platforms on both sides of the Border by using ECCC's operational Global Environmental Multiscale-Modeling Air-Quality and Chemistry (GEM-MACH) model [21] with a nested domain covering SEMI and southern Ontario at 2.5 km grid spacing. The GEM-MACH model was further used to simulate episodic ozone events in the Great Lakes region, with the aim of characterizing the dynamics of lake breezes, accounting for the influence of the local urban heat island.

The activities of the three mobile labs on a given day were determined by a conference of the field study teams based on daily real-time meteorology and air quality forecasts provided by ECCC. These forecasts also guided flights of a NASA Gulfstream aircraft equipped with a UV-visible spectrometer capable of measuring column densities of formaldehyde and NO_2 at sub-kilometer resolution. The three mobile labs sometimes collaborated with each other to map ozone and ozone precursor concentrations in relation to lake breeze fronts, especially during high ozone episodes, or to perform coordinated transects on either side of the US–Canada Border. For example, lake breeze effects were observed during regional transects via elevation of the background levels of CO and ethane. The mobile laboratories also worked individually to characterize point source emissions in the Border region.

Among the emission sources investigated by the mobile labs in Michigan were landfills in SEMI, two of which were also subject to monitoring by EGLE with sensors mounted on Unmanned Aerial Vehicles (UAVs), obtained with funding from the US Department of Energy. The AML and MPAL also investigated methane leaks from underground pipelines in SEMI as part of the MERLIN sub-experiment.

Figure 2. Examples of ground mobile, airborne, and stationary measurement platforms deployed during the MOOSE field campaign.

Datasets for the campaign were uploaded to a centralized repository hosted by NASA (see data accessibility statement). The data have undergone extensive analyses using advanced techniques such as very high-resolution 3D modeling at regional, urban, and neighborhood scales, fine-scale inverse modeling of emissions based on measured ambient air concentrations, chemical fingerprinting based on real-time measurements, and source apportionment with positive matrix factorization (PMF). Major policy-relevant results of completed analyses are presented in the following section.

3. Major Field Study Findings and Implications

3.1. Findings Relevant to Ozone Maintenance or Attainment in Southeast Michigan

Conclusions drawn from recent analyses of MOOSE field study data that are relevant to maintenance or attainment of the US ozone NAAQS in SEMI are highlighted in bold.

Local ozone production in SEMI is likely most sensitive to VOC emission controls.

VOCs and NO_x are the two main types of O_3 precursors that are directly emitted into air by industrial, automotive, consumer and commercial, and natural sources. Knowing which type of precursor most controls the amount of O_3 formed in an airshed is vital to identifying the most effective control strategies for inclusion in a State Implementation Plan (SIP) to attain the US O_3 NAAQS.

Based on prior analyses of satellite column and ground-based monitoring data, ozone modeling and trend assessments, and weekday–weekend differences, the Lake Michigan Air Directors Consortium (LADCO) had previously identified SEMI as in a transitional regime evolving away from VOC sensitivity towards NO_x sensitivity ([22] and references therein). Xiong et al. [23], however, used the observed ratio of formaldehyde (HCHO) to nitrogen dioxide (NO_2) in near-surface ambient air measured by the AML during MOOSE and a zero-dimensional (0-D) photochemical box model, F0AM (the Framework for 0-D Atmospheric Modeling), utilizing an extensive set of contemporaneous speciated VOC, NO_x, and O_3 measurements to analyze the sensitivity of local ozone in SEMI to VOC and NO_x controls. F0AM model sensitivity simulations by Xiong et al. [23] suggest that the $HCHO/NO_2$ ratio for the transition between the VOC- and NO_x-limited O_3 production regimes is 3.0 ± 0.3 in SEMI. The midday (12:00–16:00) averaged $HCHO/NO_2$ ratio during the MOOSE intensive campaign was 1.6 ± 1.0, implying that local O_3 production in SEMI is limited by VOC emissions.

Emissions of formaldehyde may be severely underestimated in official inventories. Formaldehyde controls are five times more effective by weight than NO_x controls in reducing ozone at monitors exceeding the NAAQS in SEMI.

Formaldehyde (HCHO) is a VOC that is especially important in ozone chemistry, given its role as an initial radical precursor [24]. It is also a carcinogen with additional non-cancer impacts that poses significant risks to human health [25]. A joint study by EGLE, Georgia Institute of Technology (Georgia Tech), and LADCO demonstrated that air quality models based on current emissions inventories drastically underpredict ambient HCHO compared to measurements at EGLE monitoring stations in SEMI, and that plausible order-of-magnitude corrections to the inventory for SEMI may result in over 1000 US tons per year total additional HCHO emissions [26]. Moreover, the same study showed that such plausible corrections to the HCHO inventory in a high-resolution (1.3 km horizontal grid) air quality model (CAMx) resulted in unexpectedly high ambient HCHO concentrations in the vicinity of New Haven, Michigan, which were confirmed by measurements during MOOSE.

Olaguer [27] used an advanced microscale (400 m grid) forward and inverse model (MicroFACT) constrained by AML ambient air measurements to rigorously estimate emissions in the Dearborn/southwest Detroit area, and to conclude that several large industrial facilities may emit over 1 US ton of HCHO per year, typically two orders of magnitude above values reported to or inferred from activity data using USEPA emission factors [28] by the State of Michigan. The microscale model also helped to explain the observed horizontal gradient in routine stationary measurements of HCHO performed by EGLE using the dinitrophenylhydrazine (DNPH) cartridge technique. This gradient was attributed to local primary (directly emitted) HCHO rather than secondary (atmospherically formed) HCHO.

A related modeling study by Georgia Tech [29] simulated the ozone impacts of various SEMI control strategy options being considered by EGLE, including Reasonably Available Control Technology (RACT) required by the US Clean Air Act for moderate ozone nonattainment areas. HCHO emission reductions were found to be five times more effective on a pound-for-pound basis than NO_x reductions in mitigating ozone at monitors in SEMI that previously exceeded the NAAQS. This study indicates that HCHO may be the most important VOC to control for ozone mitigation in SEMI.

Industrial solvent VOC emissions are substantial and possibly under-reported. Controlling these emissions may have significant ozone mitigation benefits.

Solvents are part of a subset of VOCs referred to as Volatile Chemical Products (VCPs). Estimates of VOC emissions from solvent use have recently increased. The USEPA has updated solvent emissions in the 2020 US National Emissions Inventory (NEI) based on the Volatile Chemical Product framework (VCPy) [30]. However, 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. During MOOSE, the AML found significant enhancements of VOCs such as acetone, aromatics, and chlorinated solvents 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 [31,32]. Trans-border emissions from certain large Canadian point sources were also observed on the US side of the Border [32].

Stroud et al. [33] used the Canadian GEM-MACH model to assess the importance of solvent emissions (see Section 3.2). GEM-MACH at 2.5 km grid spacing was evaluated for 8 h daily ozone with 2018 summer data at Windsor and showed good skill (model vs. measurements, slope = 1.04, correlation, R = 0.79). A GEM-MACH simulation of a 2021 ozone episode with U.S. VCPy emissions yielded similar maximum ozone reductions in Detroit (~0.6%) for 10% emission reductions in either solvents or mobile source NO_x. The contribution of solvent use to mono-aromatics in Detroit for the ozone episode was estimated at 60%.

Fugitive releases of methane from landfills and leaking natural gas pipelines may be large enough to significantly enhance ozone formation by VOC and NO_x sources.

Methane is not only a powerful greenhouse gas but can also enhance global concentrations of tropospheric ozone, another important greenhouse gas [34]. Although methane is an organic compound, it is not classified by the USEPA as a VOC due to its low reactivity compared to other organics, which limits the amount of local ozone it can form under normal conditions. However, large volumes of methane released from landfills and leaking natural gas pipelines may compensate for methane's low reactivity in enhancing O_3 formation by local sources of VOC and NO_x. Olaguer [35] applied the MicroFACT fine-scale air quality model to estimate O_3 impacts up to ~30 km downwind of a single hypothetical landfill in SEMI and concluded that a large methane leak of 3000 kg/h could enhance local ozone formation by landfill emission sources of O_3 precursors by at least a tenth of a ppb, and possibly several times more if sources of VOC and NO_x other than the landfill itself were accounted for.

During MOOSE, several advanced techniques were deployed by a team of Michigan scientists and engineers to measure methane emissions from two landfills in SEMI [36]. These techniques included mobile infrared cavity ringdown spectrometry, UAV-mounted meteorological sensors and tunable diode laser spectrometry, estimation of total landfill emissions of methane based on flux plane measurements, and Gaussian plume inverse modeling of distributed methane emissions in the presence of complex landfill terrain. The total methane emissions measured at the two landfills were of the order of 500 kg/h, with an uncertainty of around 50%. The results indicated that both landfill active faces and leaking gas collection systems are important sources of methane emissions.

Xia et al. [37] used the MPAL to measure ambient levels of methane at eight large operating landfills in SEMI. Elevated methane levels were typically found along the downwind side or corner of the landfills, reaching up to 38 parts per million by volume (ppm) in the morning and dropping to near-baseline levels during midday. Both mechanistically based dilution-type models and multivariate models identified wind speed, boundary layer height, barometric pressure changes, and landfill temperature as key determinants of methane levels, explaining most (r^2 = 0.89) of the variation in the maximum methane levels at the most-visited landfill.

The University of Michigan and Aerodyne mobile laboratories also measured ambient methane concentrations in the vicinity of natural gas pipelines and distribution networks. The MPAL collected 20 days' worth of 1 s methane measurements over 1100 km of surface streets in Detroit and detected 534 distinct methane peaks, equivalent to roughly one peak per 2 km traveled [38]. The AML, on the other hand, made repeated traverses of a heavily industrialized area in Dearborn and southwest Detroit where natural gas leaks were frequently encountered [32]. Methane emissions from a persistent leak observed at the intersection of Dearborn Street and Fort Street were quantified using an inverse model and found to be around 200 kg/h [39].

Temporary storage of NO_x in reactive nitrogen reservoir compounds such as HONO and HNO_3 may be inadequately simulated in current air quality models. This may affect estimates of ozone formed both locally and from precursors transported farther downwind of urban emission sources.

In the summers of 2021 (June 7–29) and 2022 (June 6–28), Chai et al. [40] performed speciated sample collections (3–12 h integration time) to quantify the isotopic composition ($^{15}N/^{14}N$, $^{17}O/^{16}O$, $^{18}O/^{16}O$) of the O_3 precursors, NO_x, NO_2, and nitrous acid (HONO), as well as their oxidation products, nitric acid (HNO_3) and particulate nitrate. These offline measurements were combined with real-time measurements of NO, NO_2, and O_3 to characterize the sources, chemical pathways, transport, and sinks of reactive nitrogen species under the influence of urban emissions and changing meteorological conditions, especially the land–lake breeze.

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 [24]. The radicals generated by HCHO and HONO, both relatively short-lived compounds, determine how much local O_3 is formed in an airshed from emissions of VOC and NO_x. 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.

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. Measured HONO concentrations ranged from 0.6 to 4.0 ppb and 0.4 to 3.0 ppb at Trinity and New Haven, respectively, in 2021, whereas HONO concentrations significantly decreased in 2022 with corresponding ranges of 0.4–2.1 ppbv and 0.3–1.9 ppbv. Over both summers, the average ratio of offline HONO to real-time NO_x in New Haven was 2–3 times that at Trinity.

Preliminary stable isotopic composition measurement and analysis performed by Chai et al. [41] demonstrated interesting differences in source contributions and secondary chemistry influence between urban and suburban sites. The isotopic data provided direct evidence of secondary HONO production via O_3 in reactive nitrogen cycling during a lake breeze in addition to impacts on O_3 over land. It also appears from the offline measurements that local concentrations of HONO may 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. The cycling of reactive nitrogen reservoirs may thus require improvements in future models. This may affect both the simulated efficacy of local control strategies and the influence of an urban airshed on modeled ozone in areas far downwind.

Contributions of secondary sulfate and nitrate to $PM_{2.5}$ in Detroit are lower than in previous assessments, and mobile sources now represent the dominant $PM_{2.5}$ contributor.

Yang et al. [42] applied the PMF source apportionment technique with spatially and temporally diverse datasets to assess source contributions and temporal trends of $PM_{2.5}$ pollution in Detroit, Michigan, including pandemic-related effects. The approach consolidated measurements from 2016 to 2021 collected at three sites where long-term $PM_{2.5}$ levels averaged from 8.63 to 10.83 $\mu g\ m^{-3}$. Most $PM_{2.5}$ was due to mobile sources (characterized

by elemental and organic carbon with some K^+) representing 33–44% of $PM_{2.5}$, depending on site and apportionment approach, followed by secondary sulfate at 24–29% of $PM_{2.5}$, and then secondary nitrate at 17–22% of $PM_{2.5}$. Smaller contributions arose from soil/dust, ferrous and non-ferrous metals, and road salt sources. Several sources varied significantly by season and site. Pandemic-related changes were generally modest. Compared to earlier apportionments, contributions of secondary sulfate and nitrate were lower, and mobile sources now represent the dominant $PM_{2.5}$ contributor.

3.2. Findings and Implications of Canadian Studies during MOOSE

Conclusions drawn from analyses of MOOSE field study data relevant to air quality improvement in southern Ontario are summarized below.

Entirely different target abatement actions may be required to reduce VOC emissions.

Healy et al. [19] collected VOC data over five days in a heavily industrialized region of southwestern Ontario containing several refineries, petrochemical production facilities, and a chemical waste disposal facility by using a mobile monitoring platform. PMF analysis was used to apportion industrial VOCs with high time-resolution measurements collected while stationary and while moving. Concentrations of VOCs (including toluene, benzene, methyl ethyl ketone, and butene) were generally elevated close to industrial facilities. Factors associated with petroleum, chemical waste, and rubber production were identified and ambient mixing ratios of selected aromatic, unsaturated, and oxygenated VOCs were apportioned to local and background sources. Fugitive emissions of benzene, highly localized and predominantly associated with storage facilities, were found to be the dominant local contributor to measured ambient benzene mixing ratios. Toluene and substituted aromatics were predominantly associated with refining and traffic, while methyl ethyl ketone was linked to chemical waste handling. The findings indicated that entirely different target abatement actions would be required to reduce local emissions of each of these VOCs. The approach to source apportionment of VOCs in this study, using fine spatial and temporal resolution, can be used to identify problematic source locations and to inform VOC emission abatement strategies.

A combined precursor reduction strategy is required to address both regional and local contributions to ozone production.

Stroud et al. [33] studied the impact of solvent emissions on reactive aromatics and ozone in the Great Lakes region by using both ECCC's operational air quality GEM-MACH model and a PMF model. VOC emissions from solvent use have increased as urban areas expand while transportation emissions have declined over the past decades. This study found that when the 2015 Canadian emissions and 2017 US emissions were used, model estimates of total mono-substituted aromatics from solvent emissions were smaller in Windsor than estimates from PMF analysis based on the 2018 measurements. The use of updated US solvent emissions for summer 2021 simulations increased the solvent use contributions and provided a more unform spatial distribution across the US–Canada Border. Long-chain alkanes were the dominant species in the model's air pollutant emission inventory and in the observation-derived solvent use factor.

The modeling further showed that summertime 8 h daytime O_3 levels decreased by 0.4% over Windsor when solvent use emissions were reduced by 10%. A 10% NO_x reduction from transportation emissions resulted in a 0.6% ozone decrease over Windsor and more widespread changes over the Great Lakes region. A combined regional-scale NO_x reduction from transportation sources and more localized industrial VOC source reductions in the Border region is likely the optimal strategy for reducing ozone episodes in southern Ontario. VOC reductions also have the co-benefit of decreasing public exposure to some toxic VOCs and secondary particulate matter.

A recent Canadian study by Zhang et al. [43] analyzed weekday/weekend data at Canadian and US urban sites in the Border region. The study showed that ozone in this region had a greater weekend positive increment due to lesser NO titration. Overall, the O_3

formation regime gradually shifted from VOC toward NO_x sensitivity during 1996–2007 then became more sensitive to VOCs during 2008–2015.

Local traffic and regional/transboundary industrial sources contribute about equally to particulate matter-bound elemental pollution in Windsor, Ontario.

Zhang et al. [44] assessed hourly measurements of $PM_{2.5}$-bound elements and black carbon collected from April–October 2021 at Windsor West station. A clear diurnal pattern was observed for most of the elements, likely related to the evolution of atmospheric mixing heights and local anthropogenic activities. Conversely, sulfur showed elevated levels in the afternoon, suggesting secondary formation of particulate sulfate from sulfur dioxide when ambient temperatures are high. Five source factors were identified by using the USEPA PMF model: three traffic-related sources (i.e., vehicular exhaust, crustal dust, and vehicle tire and brake wear factors), and two industrial sources (i.e., coal/heavy oil burning and metal processing factors). The three traffic-related sources were mostly local and contributed 47% to the total elemental concentrations, while the two industrial sources originated from regional/transboundary sources and contributed 53%.

Zhang et al. [45] conducted five additional scenarios of the PMF modeling to assess impacts of input data on source identification, source contribution, and model performance by using concentrations of $PM_{2.5}$-bound elements in Windsor, Ontario. The model outcomes and performance were found to be insensitive to data below method detection limits (MDLs) being replaced with $\frac{1}{2}$ MDLs, and to whether brown carbon data were excluded. Unique factors of fireworks and mineral dust were identified by analyzing two episodic events individually. Moreover, PMF model performance was improved greatly for event markers of the episodes and elements with less variability in concentration when compared with the base case scenario. Overall, the PMF model outcomes and performance were sensitive to the fraction of concentration measurements below MDLs and element concentrations with large variability due to high concentrations observed in episodes. The findings are useful for dealing with data below MDLs and episodic events in conducting future PMF source apportionment.

More accurate initialization of meteorological models may improve regional air quality model predictions of ozone, especially at levels exceeding the CAAQS.

Mashayekhi et al. [46] investigated the influence of meteorology initialization on prediction of surface O_3 in summer 2021 in the Great Lakes region by using ECCC's operational GEM-MACH model at a horizontal resolution of 2.5 km. In comparison to the conventional initialization (i.e., single-time full-field digital filter), the advanced meteorology initialization technique of the four-dimensional incremental analysis updating (IAU) method improved the model performance of surface O_3 for both exceedances and non-exceedances of the 2025 CAAQS of 60 ppb. The study highlights the importance of meteorological parameters in predicting surface O_3 levels. It was also found that the simulation initialized at 18Z demonstrated superior performance of the surface O_3 prediction, especially during days when O_3 levels exceeded the threshold of 60 ppb. In-depth analysis further indicated that better prediction of surface O_3 in the Great Lakes regions is linked to a more accurate representation of wind speed in the afternoon when O_3 levels are high.

4. Discussion and Conclusions

The results to date of the MOOSE field campaign have yielded important policy-relevant conclusions that can lead to improved air quality strategies in the US–Canada Border region. In SEMI, the results point to VOC controls as the necessary primary focus of local ozone mitigation efforts, especially the identification, quantification, and reduction of emissions of formaldehyde and, secondarily, industrial solvents. The control of NO_x emissions, however, may still be important in reducing regional ozone pollution. The State of Michigan's efforts at greenhouse gas reduction should also consider the local and

regional ozone co-benefits of identifying, quantifying, and reducing emissions of methane from landfills and natural gas pipelines.

In southern Ontario, local air quality will benefit from O_3 and $PM_{2.5}$ precursor reductions in Michigan due to the importance of transboundary pollution from the US. The extensive presence of the Canadian petrochemical industry in southwestern Ontario may require authorities to re-examine the accuracy and completeness of local emissions inventories related to products of incomplete combustion, including formaldehyde, as well as inventories of solvents and other VOCs. This is because GEM-MACH simulations suggest that VOC reductions in the Border region may yield comparable ozone mitigation benefits to mobile source NO_x reductions during ozone episodes.

Among the control strategies that could be evaluated by regulators on both sides of the US–Canada Border are: (1) flare elimination, minimization, and/or efficiency improvement; and (2) oxidation catalysts, which can decrease HCHO emissions from stationary engines by 96% according to State of New Jersey data [47]. The first strategy has been successfully implemented by the US petrochemical industry and may be imitated by the steel and coking industries in Michigan. Oxidation catalysts, on the other hand, could be considered as possible controls for large stationary engines at natural gas pipeline stations, gas-fired units at power plants, and landfill gas-to-energy conversion facilities.

In the case of fugitive emissions of organic solvents and methane, a major priority should be the development and implementation of contemporary methods for leak detection and repair (LDAR). The use of fast chemical instrumentation and inverse modeling methods applied during the MOOSE campaign, unlike more commonly used LDAR techniques, is scalable to large facilities and could be considered for wider adoption.

Lastly, the results of MOOSE emphasize the importance of improving modeling practice to better assess the impacts of control strategies in the Border region, including the use of finer-resolution air quality models and updating of initialization techniques in meteorological models. The USEPA should work with the states to promote the continued update of national formaldehyde and industrial solvent emissions inventories, and to re-examine the treatment of reactive nitrogen reservoirs in regional air quality models. Both these efforts will enhance the ability of regulatory air quality models used in ozone attainment demonstrations to accurately evaluate ozone control strategies. The updating of reactive nitrogen cycling in models will also help improve assessments of long-range transport of ozone and its precursors.

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References

1. U.S. Code of Federal Regulations, Title 40, Parts 50, 51, 52, 53, and 58. National Ambient Air Quality Standards for Ozone. *Fed. Regist.* **2015**, *80*, 206.
2. Canadian Council of Ministers of the Environment. *Guidance Document on Achievement Determination for Canadian Ambient Air Quality Standards for Ozone*; Canadian Council of Ministers of the Environment: Winnipeg, MB, Canada, 2021; 26p, ISBN 978-1-77202-067-0.
3. Brook, J.R.; Makar, P.A.; Sills, D.M.L.; Hayden, K.L.; McLaren, R. Exploring the nature of air quality over southwestern Ontario: Main findings from the Border Air Quality and Meteorology Study. *Atmos. Chem. Phys.* **2013**, *13*, 10461–10482. [CrossRef]
4. Yap, D.; Reid, N.; De Brou, G.; Bloxam, R. *Transboundary Air Pollution in Ontario*; Ontario Ministry of the Environment: Toronto, ON, Canada, 2005.
5. Ontario Ministry of the Environment, Conservation, and Parks (MECP). *Air Quality in Ontario 2020 Report*; MECP: Toronto, ON, Canada, 2020. Available online: https://www.ontario.ca/document/air-quality-ontario-2020-report (accessed on 21 August 2023).
6. Michigan-Ontario Ozone Source Experiment (MOOSE). Available online: https://www-air.larc.nasa.gov/missions/moose/docs/MOOSE%20Science%20Plan%20052521.pdf (accessed on 25 August 2023).
7. Michigan Department of Environment, Great Lakes, and Energy (EGLE). *Quality Assurance Project Plan for the Southeast Michigan Chemical Source Signature (CHESS) Experiment*; EGLE: Lansing, MI, USA, 2021.
8. Herndon, S.C.; Jayne, J.T.; Zahniser, M.S.; Worsnop, D.R.; Knighton, B.; Alwine, E.; Lamb, B.K.; Zavala, M.; Nelson, D.D.; McManus, J.B.; et al. Characterization of urban pollutant emission fluxes and ambient concentration distributions using a mobile laboratory with rapid response instrumentation. *Faraday Discuss.* **2005**, *130*, 327–339. [CrossRef]
9. Kolb, C.E.; Herndon, S.C.; McManus, J.B.; Shorter, J.H.; Zahniser, M.S.; Nelson, D.D.; Jayne, J.T.; Canagaratna, M.R.; Worsnop, D.R. Mobile laboratory with rapid response instruments for real-time measurements of urban and regional trace gas and particulate distributions and emission source characteristics. *Environ. Sci. Technol.* **2004**, *38*, 5694–5703. [CrossRef]
10. Yacovitch, T.I.; Herndon, S.C.; Roscioli, J.R.; Floerchinger, C.; Knighton, W.B.; Kolb, C.E. Air pollutant mapping with a mobile laboratory during the BEE-TEX field study. *Environ. Health Insights* **2015**, *9* (Suppl. S4), 7–13. [CrossRef] [PubMed]
11. McManus, J.B.; Zahniser, M.S.; Nelson, D.D.; Shorter, J.H.; Herndon, S.C.; Jervis, D.; Agnese, M.; McGovern, R.; Yacovitch, T.I.; Roscioli, J.R. Recent progress in laser-based trace gas instruments: Performance and noise analysis. *Appl. Phys. B* **2015**, *119*, 203–218. [CrossRef]
12. Yacovitch, T.I.; Herndon, S.C.; Roscioli, J.R.; Floerchinger, C.; McGovern, R.M.; Agnese, M.; Pétron, G.; Kofler, J.; Sweeney, C.; Karion, A.; et al. Demonstration of an ethane spectrometer for methane source identification. *Environ. Sci. Technol.* **2014**, *48*, 8028–8034. [CrossRef]
13. Spinei, E.; Whitehill, A.; Fried, A.; Tiefengraber, M.; Knepp, T.N.; Herndon, S.; Herman, J.R.; Müller, M.; Abuhassan, N.; Cede, A.; et al. The first evaluation of formaldehyde column observations by improved Pandora spectrometers during the KORUS-AQ field study. *Atmos. Meas. Tech.* **2018**, *11*, 4943–4961. [CrossRef]
14. Claflin, M.S.; Pagonis, D.; Finewax, Z.; Handschy, A.V.; Day, D.A.; Brown, W.L.; Jayne, J.T.; Worsnop, D.R.; Jimenez, J.L.; Ziemann, P.J.; et al. An in situ gas chromatograph with automatic detector switching between PTR- and EI-TOF-MS: Isomer-resolved measurements of indoor air. *Atmos. Meas. Tech.* **2021**, *14*, 133–152. [CrossRef]
15. Krechmer, J.; Lopez-Hilfiker, F.; Koss, A.; Hutterli, M.; Stoermer, C.; Deming, B.; Kimmel, J.; Warneke, C.; Holzinger, R.; Jayne, J.; et al. Evaluation of a new reagent-ion source and focusing ion–molecule reactor for use in proton-transfer-reaction mass spectrometry. *Anal. Chem.* **2018**, *90*, 12011–12018. [CrossRef]
16. Riva, M.; Rantala, P.; Krechmer, J.E.; Peräkylä, O.; Zhang, Y.; Heikkinen, L.; Garmash, O.; Yan, C.; Kulmala, M.; Worsnop, D.; et al. Evaluating the performance of five different chemical ionization techniques for detecting gaseous oxygenated organic species. *Atmos. Meas. Tech.* **2019**, *12*, 2403–2421. [CrossRef]
17. Xia, T.; Catalan, J.; Hu, C.; Batterman, S. Development of a mobile platform for monitoring gaseous, particulate and greenhouse gas (GHG) pollutants. *Environ. Monit. Assess.* **2021**, *193*, 7. [CrossRef] [PubMed]
18. University of Michigan School of Public Health. *The Southeast Michigan Chemical Source Signature (CHESS) Experiment, Final Report*; Michigan Department of Environment, Great Lakes, and Energy: Lansing, MI, USA, 2023.
19. Healy, R.M.; Sofowote, U.M.; Wang, J.M.; Chen, Q.; Todd, A. Spatially resolved source apportionment of industrial VOCs using a mobile monitoring platform. *Atmosphere* **2022**, *13*, 1722. [CrossRef]
20. Zhao, X.; Fioletov, V.; Alwarda, R.; Su, Y.; Griffin, D.; Weaver, D.; Strong, K.; Cede, A.; Hanisco, T.; Tiefengraber, M.; et al. Tropospheric and surface nitrogen dioxide changes in the greater Toronto area during the first two years of the COVID-19 pandemic. *Remote Sens.* **2022**, *14*, 1625. [CrossRef]

21. Stroud, C.A.; Ren, S.; Zhang, J.; Moran, M.; Akingunola, A.; Makar, P.; Munoz, R.A.; Leroyer, S.; Bélair, S.; Sills, D.; et al. Chemical analysis of surface-level ozone exceedances during the 2015 Pan American Games. *Atmosphere* **2020**, *11*, 572. [CrossRef]
22. Dickens, A.F. *Ozone Formation Sensitivity to NO_x and VOC Emissions in the LADCO Region*; Lake Michigan Air Directors Consortium: Hillside, IL, USA, 2022; Available online: https://www.ladco.org/wp-content/uploads/Projects/Ozone/Ozone-Formation-Sensitivity-Report-FINAL-9-8-22.pdf (accessed on 25 August 2023).
23. Xiong, Y.; Chai, J.; Mao, H.; Mariscal, N.; Yacovitch, T.; Lerner, B.; Majluf, F.; Canagaratna, M.; Olaguer, E.P.; Huang, Y. Examining the summertime ozone formation regime in southeast Michigan using MOOSE ground-based HCHO/NO_2 measurements and F0AM box model. *J. Geophys. Res. Atmos.* **2023**, *128*, e2023JD038943. [CrossRef]
24. Olaguer, E.P.; Rappenglück, B.; Lefer, B.; Stutz, J.; Dibb, J.; Griffin, R.; Brune, W.H.; Shauck, M.; Buhr, M.; Jeffries, H.; et al. Deciphering the role of radical precursors during the Second Texas Air Quality Study. *J. Air Waste Manag. Assoc.* **2009**, *59*, 1258–1277. [CrossRef]
25. U.S. Environmental Protection Agency. *Assessment Overview for the Toxicological Review of Formaldehyde—Inhalation [CASRN 50-00-0]*; EPA/635/R-22/039c; Office of Research and Development: Washington, DC, USA, 2022.
26. Olaguer, E.P.; Hu, Y.; Kilmer, S.; Adelman, Z.E.; Vasilakos, P.; Odman, M.T.; Vaerten, M.; McDonald, T.; Gregory, D.; Lomerson, B.; et al. Is there a formaldehyde deficit in emissions inventories for southeast Michigan? *Atmosphere* **2023**, *14*, 461. [CrossRef]
27. Olaguer, E.P. Inverse modeling of formaldehyde emissions and assessment of associated cumulative ambient air exposures at fine scale. *Atmosphere* **2023**, *14*, 931. [CrossRef]
28. U.S. Environmental Protection Agency. AP-42: Compilation of Air Emissions Factors. Available online: https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emissions-factors (accessed on 25 August 2023).
29. Georgia Institute of Technology. *Southeast Michigan Air Quality Modeling Study*; Lake Michigan Air Directors Consortium: Hillside, IL, USA, 2022; Available online: http://semap.ce.gatech.edu/LADCO/SEMI_Modeling_Final_Report.pdf (accessed on 16 October 2023).
30. Seltzer, K.M.; Pennington, E.; Rao, V.; Murphy, B.N.; Strum, M.; Isaacs, K.K.; Pye, H.O.T. Reactive organic carbon emissions from volatile chemical products. *Atmos. Chem. Phys.* **2021**, *21*, 5079–5100. [CrossRef]
31. Yacovitch, T.I.; Daube, C.; Lerner, B.; Claflin, M.; Fortner, E.; Canagaratna, M.; Herndon, S.C. *Aerodyne Mobile Laboratory Measurements during the Chemical Source Signatures Experiment (CHESS): 2021 Michigan-Ontario Ozone Source Experiment (MOOSE), Final Report*; Michigan Department of Environment, Great Lakes, and Energy: Lansing, MI, USA, 2022.
32. Yacovitch, T.I.; Lerner, B.; Canagaratna, M.; Daube, C.; Healy, R.M.; Wang, J.M.; Fortner, E.; Majluf, F.; Claflin, M.; Roscioli, J.R.; et al. Mobile laboratory investigations of industrial point source emissions during the MOOSE field campaign. *Atmosphere* **2023**, *14*, accepted.
33. Stroud, C.A.; Zhang, J.; Boutzis, E.I.; Zhang, T.; Mashayekhi, R.; Nikiema, O.; Majdzadeh, M.; Wren, S.N.; Xu, X.; Su, Y. Impact of solvent emissions on reactive aromatics and ozone in the Great Lakes region. *Atmosphere* **2023**, *14*, 1094. [CrossRef]
34. Stocker, T.F.; Qin, D.; Plattner, G.-K.; Tignor, M.; Allen, S.K.; Boschung, J.; Nauels, A.; Xia, Y.; Bex, V.; Midgley, P.M. Intergovernmental panel on climate change (IPCC). In *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press: Cambridge, UK; Cambridge University Press: New York, NY, USA, 2013.
35. Olaguer, E.P. The potential ozone impacts of landfills. *Atmosphere* **2021**, *12*, 877. [CrossRef]
36. Olaguer, E.P.; Jeltema, S.; Gauthier, T.; Jermalowicz, D.; Ostaszewski, A.; Batterman, S.; Xia, T.; Raneses, J.; Kovalchick, M.; Miller, S.; et al. Landfill emissions of methane inferred from unmanned aerial vehicle and mobile ground measurements. *Atmosphere* **2022**, *13*, 983. [CrossRef]
37. Xia, T.; Borjigin, S.G.; Raneses, J.; Stroud, C.A.; Batterman, S.A. Mobile measurements of atmospheric methane at eight large landfills: An assessment of temporal and spatial variability. *Atmosphere* **2023**, *14*, 906. [CrossRef]
38. Xia, T.; Raneses, J.; Batterman, S.A. Improving the performance of pipeline leak detection algorithms for the mobile monitoring of methane leaks. *Atmosphere* **2022**, *13*, 1043. [CrossRef]
39. Olaguer, E.P. *Combining Real-Time Ambient Air Measurements with Inverse Modeling to Estimate Oil and Gas Industry Emissions*; EM Magazine, Air and Waste Management Association: Pittsburgh, PA, USA, September 2023.
40. Chai, J.; Donnachie, E.; Lee, T.; Kim, H.; Xiong, Y.; Huang, Y.; Mao, H.; Hastings, M. Characterization of summertime HONO to NO_x ratio at an urban and suburban site in southeast Michigan during MOOSE campaign. *Manuscript in preparation*.
41. Chai, J.; Donnachie, E.; Lee, T.; Kim, H.; Xiong, Y.; Huang, Y.; Mao, H.; Hastings, M.G. Tracking the Fates and Transport of NO_x and HONO and their Interplay with O_3 in Summertime Southeast Michigan. In Proceedings of the American Meteorological Society, 103rd Annual Meeting, Denver, CO, USA, 9 January 2023.
42. Yang, Z.; Islam, M.K.; Xia, T.; Batterman, S. Apportionment of $PM_{2.5}$ Sources across Sites and Time Periods: An Application and Update for Detroit, Michigan. *Atmosphere* **2023**, *14*, 592. [CrossRef]
43. Zhang, T.; Xu, X.; Su, Y. Long-term measurements of ground-level ozone in Windsor, Canada and surrounding areas. *Chemosphere* **2022**, *294*, 133636. [CrossRef]
44. Zhang, T.; Su, Y.; Debosz, J.; Noble, M.; Munoz, A.; Xu, X. Continuous measurements and source apportionment of ambient $PM_{2.5}$-bound elements in Windsor, Canada. *Atmosphere* **2023**, *14*, 374. [CrossRef]
45. Zhang, T.; Su, Y.; Debosz, J.; Noble, M.; Munoz, A.; Xu, X. Sensitivity of source apportionment of ambient $PM_{2.5}$-bound elements to input concentration data. *Atmosphere* **2023**, *14*, 1269. [CrossRef]

46. Mashayekhi, R.; Stroud, C.A.; Zhang, J.; Nikiema, O.; Trotechaud, S. The influence of meteorology initialization on ozone forecasting in the Great Lakes region during MOOSE study. *Atmosphere* **2023**, *14*, 1383. [CrossRef]
47. Ratzman, K. Formaldehyde Emissions from Landfill Gas and Natural Gas Engines. In Proceedings of the Mid-Atlantic Regional Air Management Association (MARAMA) Air Toxics Workshop, Towson, MD, USA, 21–23 August 2018.