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LETTER

Mortality-based damages per ton due to the on-road mobile sector in the Northeastern and Mid-Atlantic U.S. by region, vehicle class and precursor

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Abstract

On-road vehicular emissions contribute to the formation of fine particulate matter and ozone which can lead to increased adverse health outcomes near the emission source and downwind. In this study, we present a transportation-specific modeling platform utilizing the community multiscale air quality model (CMAQ) with the decoupled direct method (DDM) to estimate the air quality and health impacts of on-road vehicular emissions from five vehicles classes; light-duty autos, light-duty trucks (LDT), medium-duty trucks, heavy-duty trucks (HDT), and buses (BUS), on $PM_{2.5}$ and O_3 concentrations at a 12 $\times$ 12 kilometer scale for 12 states and Washington D.C. as well as four large metropolitan statistical areas in the Northeast and Mid-Atlantic U.S. in 2016. CMAQ-DDM allows for the quantification of sensitivities from individual precursor emissions (NO_X, SO_2, NH_3, volatile organic compounds, and $PM_{2.5}$) in each state to pollution levels and health effects in downwind states. In the region we considered, LDT are responsible for the most $PM_{2.5}$-attributable premature mortalities at 1234 with 46% and 26% of those mortalities from directly emitted primary particulate matter and NH_3, respectively; and O_3-attributable premature mortalities at 1129 with 80% of those mortalities from NO_X emissions. Based on a detailed source-receptor matrix of sensitivities with subsequent monetization of damages that we computed, we find that the largest damages-per-ton estimate is approximately $4 million per ton of directly emitted primary particulate matter from BUS in the New York-Newark-Jersey City metropolitan statistical area. We find that on-road vehicular NH_3 emissions are the second largest contributor to $PM_{2.5}$ concentrations and health impacts in the study region, and that reducing 1 ton of NH_3 emissions from LDT is ~75 times and from HDT is ~90 times greater in terms of damages reductions than a 1 ton reduction of NO_X. By quantifying the impacts by each combination of source region, vehicle class, and emissions precursor this study allows for a comprehensive understanding of the largest vehicular sources of air quality-related premature mortalities in a heavily populated part of the U.S. and can inform future policies aimed at reducing those impacts.

1. Introduction

Emissions from fossil fuel combustion by mobile sources contribute to poor air quality through the formation of air pollutants such as fine particulate matter ($PM_{2.5}$) and ozone (O_3). Exposure to these air pollutants have been associated with increased premature mortalities. The mobile source sector remains one of the largest contributors to $PM_{2.5}$ and O_3 globally and in the U.S. (Anenberg *et al* 2017, Zawacki *et al* 2018, Yang *et al* 2019) with one study estimating ~385 000 $PM_{2.5}$ and O_3-attributable premature mortalities globally in 2015 from tailpipe emissions (Anenberg *et al* 2019). In the U.S., road

transportation mobile sources have been estimated to be the largest source of air pollution related premature mortalities (Caiazzo *et al* 2013), responsible for ~53 000 $PM_{2.5}$ and 5000 O_3-attributable premature mortalities.

Prior studies have quantified the health impacts from the on-road vehicle sector in the U.S.. Caiazzo *et al* (2013), Dedoussi and Barrett (2014), Davidson *et al* (2020) and Dedoussi *et al* (2020) quantified premature mortalities in the U.S. from road transportation in 2005 and 2011. Caiazzo *et al* (2013) broke down $PM_{2.5}$ and O_3 mortalities occurring in each state due to on-road emissions through zero-out approaches within a chemical transport model (CTM), but did not break down the contribution due to $PM_{2.5}$ and O_3 precursor emission species, source regions, or vehicle types. Dedoussi and Barrett (2014) broke down the $PM_{2.5}$ mortalities from on-road emissions by source states and emission precursors through an adjoint sensitivity analysis within a CTM but did not include O_3-attributable mortalities, and secondary organic aerosols were not included due to the limitations of the model. Both Caiazzo *et al* (2013) and Dedoussi and Barrett (2014) performed their studies for inventory years of 2005 that did not account for recent mobile source emission regulations such as the Tier 3 Motor Vehicle Emission and Fuel standards. Wolfe *et al* (2019) monetized $PM_{2.5}$ damages per ton of directly emitted $PM_{2.5}$, sulfur oxides (SO_2)/directly emitted sulfate (pSO_4), and nitrogen oxides (NO_X) from a subset of vehicle classes as defined in Davidson *et al* (2020), but did not calculate O_3 damages per ton and did not calculate $PM_{2.5}$ damages per ton of on-road emissions of ammonia (NH_3) and volatile organic compounds (VOCs). Goodkind *et al* (2019) utilized a reduced-complexity model (RCM) to calculate the $PM_{2.5}$ damages per ton of directly emitted $PM_{2.5}$, SO_2, VOC, NO_X, and NH_3 broken down by the on-road emission sector in 2011, but did not calculate O_3 damages per ton or account for differing effects of $PM_{2.5}$ by season due to the nature of the RCM. Dedoussi *et al* (2020) quantified $PM_{2.5}$ and O_3 mortalities in 2011 by source region, sector, and precursor through an adjoint sensitivity analysis but did not break down the on-road sector into vehicle types, secondary organic aerosols were excluded, and the coarse model resolution may not capture localized impacts in densely populated urban areas. Davidson *et al* (2020) broke down the $PM_{2.5}$ and O_3 mortalities by vehicles' fuel types in 2011 using source apportionment methodologies within a CTM, but did not break down impacts by source region or precursor emission species.

This study aims to address the gaps listed above and add to this growing list of literature by quantifying health impacts in a heavily populated region of the U.S., i.e. the Northeast and Mid-Atlantic U.S., broken down by five precursor emission species from five vehicle classes from 12 states and Washington D.C. and four metropolitan statistical areas (MSAs) in 2016. By quantifying impacts from these individual combinations of vehicle class/region/emissions precursor species, this study will be the first to quantify the $PM_{2.5}$ and O_3 premature mortalities in each of our 12 states, Washington D.C. and four MSAs attributable to precursor emissions from five distinct vehicle classes from each of the 12 states, Washington D.C. and four MSAs. We aim to quantify the largest total damages per ton estimates in each of these 12 states, Washington D.C. and four MSAs by vehicle class, precursor emission species, and emission source state/MSA. This study will utilize the decoupled direct method (DDM) forward sensitivity modeling technique in the community multiscale air quality model (CMAQ) that calculates sensitivities of $PM_{2.5}$ and O_3 concentrations to each of our variables of interest. This information is critical in developing effective emission control strategies, especially in a region as heavily populated as the Northeast and Mid-Atlantic U.S. which contains ~22% of the U.S. population.

The motivation for looking at this part of the U.S. and for utilizing this particular sensitivity methodology is research to support the Transportation, Equity, Climate, and Health Study (TRECH study). The TRECH study is an independent co-benefits study that looks to quantify potential health outcomes associated with a range of cap and invest scenarios under the proposed Transportation and Climate Initiative (TCI) (Transportation and Climate Initiative 2020), a set of policies aimed at mitigating climate impact by reducing emissions from on-road vehicles which is expected to have health benefits due to better air quality (Driscoll *et al* 2015, Mittal *et al* 2015, Buonocore *et al* 2016, 2018). This research, that quantifies damages per ton due to vehicle classes by source regions, is a key component of the TRECH study that aims to quantify the air quality and health-related impacts of on-road vehicle classes' emissions in each of the states that make up the TCI region as well as each state's impact on other states within the region.

2. Methods

2.1. Air quality modeling

The CMAQ model is used to quantify $PM_{2.5}$ and O_3 concentrations across the model domain using a 2016 air quality modeling platform. The DDM sensitivity analyis (Dunker 1984, Napelenok *et al* 2006, 2008, Koo *et al* 2007) as implemented in CMAQv5.2 (U.S. EPA Office of Research and Development 2017) with the Carbon Bond 6 revision 3 mechanism (CB6r3) (Luecken *et al* 2019) is used to calculate sensitivities of $PM_{2.5}$ and O_3 concentrations in each model grid cell to precursor emissions from on-road vehicle classes in each source region (Towns *et al* 2014). Sensitivities as calculated in the DDM framework describe the incremental change in pollutant concentrations with respect to model inputs

across the domain to estimate how sensitive pollutant concentrations are to a specific model input. Our modeling domain covers the eastern half of the U.S. with 12 × 12 km horizontal grid cell resolution (figure S1 (available online at stacks.iop.org/ERL/16/065008/mmedia)).

Precursor emissions to O_3 from on-road vehicles are nitrogen oxides (NO_X) and VOC; and precursor emissions to $PM_{2.5}$ are NO_X, VOC, sulfur dioxide (SO_2), ammonia (NH_3), and directly emitted primary $PM_{2.5}$ (henceforth referred to as PPM). On-road vehicle emissions are taken from the U.S. Environmental Protection Agency's (EPA) 2016v1 modeling platform based on the National Emissions Inventory (National Emissions Inventory Collaborative 2019, U.S. Environmental Protection Agency 2020). On-road emissions are generated using emission factors representative of all national fuel economy and GHG standards for vehicles as of October 2015 (U.S. Environmental Protection Agency 2015), county and source classification code (SCC)-specific activity data submitted by states for the year 2016 (U.S. Environmental Protection Agency 2020), and hourly meteorological data. Five distinct vehicle class emissions inventories were generated using the Sparse Matrix Operator Kernel Emissions (Baek and Seppanen 2018) modeling system according to SCC values (table S6) grouped by the MOtor Vehicle Emission Simulator (U.S. Environmental Protection Agency 2015) (MOVES2014a) vehicle types: light-duty autos (LDA), light-duty trucks (LDT), buses (BUS), medium-duty trucks (MDT), and heavy-duty trucks (HDT). Vehicle class emission inventories were generated for twelve states in the Northeast U.S. that make up the TCI region: Connecticut (CT), Delaware (DE), Maine (ME), Maryland (MD), Massachusetts (MA), New Hampshire (NH), New Jersey (NJ), New York (NY), Pennsylvania (PA), Rhode Island (RI), Vermont (VT), Virginia (VA); the District of Columbia (DC); and four large MSAs: the Boston-Cambridge-Newton, MA NH MSA (BOSMSA), New York-Newark-Jersey City, NY NJ PA (NYMSA), Philadelphia-Camden-Wilmington, PA NJ DE MD (PHILMSA), and the combined Baltimore-Columbia-Towson, MD and the Washington-Arlington-Alexandria, DC VA MD WV MSAs (BALMSA). Tables S1–S4 show the counties that comprise each of these source region MSAs. The resulting matrix of source-impact sensitivities are O_3 and $PM_{2.5}$ concentration sensitivities to the number of precursors × the number of vehicle classes × the number of source states/MSAs. Hence for O_3, the number of sensitivities = 2 × 5 × 17 = 170; and for $PM_{2.5}$, the number of sensitivities = 5 × 5 × 17 = 425. CMAQ-DDM simulations are run for January and July in 2016 to represent the winter and summer season, respectively. The results are then averaged to represent the annual contribution of precursor emissions from the selected vehicle classes and source regions to regional O_3 and $PM_{2.5}$ concentrations. Information regarding DDM calculations within CMAQ and model evaluation against observations can be found in the supporting information.

2.2. Health impact assessment

Exposure to elevated levels of ground-level O_3 and $PM_{2.5}$ concentrations have been associated with increased adverse health effects (Bell *et al* 2004, Laden *et al* 2006, Jerrett *et al* 2009, Krewski *et al* 2009) leading to the development of concentration response functions (CRFs) that quantify the increased risk of adverse health effects occurring per unit increase in pollutant concentration. To perform the health impact assessments for all of the simulations that were done, we built a health impact assessment tool BenMAPR. Similar to the U.S. EPA's Benefits Mapping and Analysis Program (BenMAP) (Sacks *et al* 2018), BenMAPR is a geospatial air pollution health impact assessment modeling platform that links air pollution exposures to data on exposed populations and their background health. It then calculates the health impacts of these exposures using CRFs from the epidemiological literature.

For quantifying $PM_{2.5}$-attributable premature mortalities, we make use of a CRF from a recently published meta-analysis (Vodonos *et al* 2018) that found a 1.29% (95% CI 1.09–1.5) increase in all-cause mortality per 10 $\mu g\ m^{-3}$ increase in $PM_{2.5}$. For O_3-attributable premature mortalities, we use a CRF associating all-cause mortality to long-term O_3 exposure with a hazards ratio of 1.02 (95%CI 1.01–1.04) per 10 ppb increase in O_3 (Turner *et al* 2016). For both $PM_{2.5}$ and O_3-attributable premature mortalities, source region and vehicle-class specific contributions were calculated for adults 25 and over. We used linear approximations for each CRF as that has been found to be appropriate for U.S.—relevant changes in concentrations (Schwartz *et al* 2008, Vodonos *et al* 2018, Gilmore *et al* 2019).

We obtained population data at U.S. Census tract level split by age from the U.S. Census American Community Survey for the year 2018, the most recent year available. To obtain stable and generalizable mortality rate estimates, we used an average of county level baseline mortality rates from 1999 to 2016, the most recent years available from the U.S. Centers for Disease Control Wide-ranging Online Database for Epidemiologic Research. Baseline hospitalization and other adult morbidity data was obtained from BenMAP (Sacks *et al* 2018), which sources morbidity data from the Agency for Healthcare Research and Quality Healthcare Cost and Utilization Project (Agency for Healthcare Research and Quality, Rockville, MD 2020).

2.3. Emissions

Table 1 shows the emission totals in the TCI region by vehicle class for the months of January and July.

Table 1. Emissions of NO_X, VOC, SO_2, NH_3, and PPM in tons/month for January and July 2016 by each vehicle class.

	January					July				
Source vehicle	NO_X	VOC	SO_2	NH_3	PPM	NO_X	VOC	SO_2	NH_3	PPM
LDA	8638	15 258	186	579	417	9862	14 130	242	740	203
LDT	14 140	16 629	218	550	484	17 185	15 756	284	697	271
MDT	4875	1466	17	35	259	6186	1758	22	44	304
HDT	8840	900	24	42	363	11 118	1073	32	53	456
BUS	1594	342	3	7	68	2017	394	5	8	86

Table 2. Population-weighted sensitivities of $PM_{2.5}$ ($\mu g\ m^{-3}$) and O_3 (ppb) to precursors from each vehicle class averaged across the TCI region.

	$PM_{2.5}$					O_3	
Source vehicle	NO_X	VOC	NH_3	SO_2	PPM	NO_X	VOC
LDA	6.2×10^{-4}	1.7×10^{-3}	3.7×10^{-3}	1.7×10^{-4}	5.8×10^{-3}	3.3×10^{-2}	1.5×10^{-2}
LDT	1.4×10^{-3}	1.9×10^{-3}	3.3×10^{-3}	1.9×10^{-4}	6.0×10^{-3}	6.3×10^{-2}	1.6×10^{-2}
MDT	3.8×10^{-4}	2.4×10^{-4}	2.4×10^{-4}	1.5×10^{-5}	5.5×10^{-3}	2.2×10^{-2}	2.6×10^{-3}
HDT	9.3×10^{-4}	1.3×10^{-4}	3.1×10^{-4}	1.9×10^{-5}	7.2×10^{-3}	4.0×10^{-2}	1.4×10^{-3}
BUS	5.8×10^{-5}	6.0×10^{-5}	5.7×10^{-5}	3.2×10^{-6}	2.1×10^{-3}	4.7×10^{-3}	7.2×10^{-4}

Tables S8–S12 show the same for each source region. LDT are the largest source of NO_X, VOC, and SO_2 emissions in the 13 states (DC henceforth referred to as the 13th state) that make up the TCI region (excluding the MSAs as they are contained within one or multiple states) with 14 140 and 17 185 tons of NO_X, 16 629 and 15 756 tons of VOC, and 218 and 284 tons of SO_2 emitted in January and July, respectively. LDA are the largest source of NH_3 emissions with 579 tons in January and 740 tons in July. For PPM emissions, LDA are the largest source in January with 417 tons while HDT are the largest source in July with 456 tons.

Summing emissions from vehicle classes, Pennsylvania is the largest source of NO_X (8071 tons in January and 10 440 tons in July) and VOC (7463 tons in January and 7008 tons in July) emissions while New York is the largest source of SO_2 (99 tons in January and 134 tons in July), NH_3 (243 tons in January and 327 tons in July), and PPM (348 tons in January and 311 tons in July) emissions. In January, the individual largest emission sources for NO_X, SO_2, VOC, NH_3, and PPM are LDT from VA, LDT from PA, LDT from NY, LDA from NYMSA, and LDT from PA, respectively. In July, the individual largest emission sources for NO_X, SO_2, VOC, NH_3, and PPM are LDT from VA, LDT from VA, LDT from NY, LDA from NY, and HDT from PA, respectively.

Figures S2–S6 show the percentage of contribution to each of the total precursor emissions (from all sources) in each source region. Light-duty vehicles predominantly emit gas-phase precursors while heavy and medium duty trucks emit a larger portion of particle-phase emissions. Vehicular location and distribution will determine how precursor emissions from the vehicle classes in different source regions will impact $PM_{2.5}$ and O_3, both near the emission source and downwind.

3. Results

3.1. Population-weighted exposure

Table 2 shows the population-weighted annual mean $PM_{2.5}$ and O_3 sensitivities to each vehicle class and precursor in the states that make up the TCI region. $PM_{2.5}$ sensitivities are annually averaged and O_3 sensitivities are the annual average of the daily 8 h maximum. We continue to exclude MSAs as source regions here so as to avoid double counting the vehicle emissions contained in both the MSA and states within that MSA. $PM_{2.5}$ sensitivities to PPM and NH_3 are the largest amongst the precursors to $PM_{2.5}$ while O_3 sensitivities to NO_X are the largest amongst the precursors to O_3. LDA and LDT $PM_{2.5}$ sensitivities to VOC, NH_3, SO_2, and PPM are similar in magnitude, reflective of the magnitude and spatial distribution of the emissions of those vehicle classes. $PM_{2.5}$ sensitivities to VOC, NH_3, SO_2, and PPM are almost entirely positive across the domain for both months of simulation. $PM_{2.5}$ sensitivities to NO_X emissions in January are negative in some areas. Aerosol NO_3^- and NH_4^+ can have negative sensitivities to on-road NO_X emissions primarily in urban areas of our domain during the winter months due to the chemical system and feedback of emitted precursors and the formation of SO_4^{2-}-NO_3^--NH_4^+ components of $PM_{2.5}$. The main drivers of wintertime inorganic aerosol chemistry are oxidant availability, cloud water chemistry, and gas-particle partitioning which will influence how much SO_4^{2-} is formed from SO_2 and the partitioning of NH_3 to NH_4^+ and NO_X to NO_3^-. Recent studies in the Eastern U.S. (Shah *et al* 2018, Vasilakos *et al* 2018, Pye *et al* 2020) have characterized the response of inorganic aerosols to these drivers and help to explain the negative sensitivities of NO_3^- and NH_4^+ to on-road emissions of NO_X in our domain during winter months.

Table 3. Total premature mortalities in the TCI region due to long-term exposure to $PM_{2.5}$ attributable to each vehicle class and the percentage attributable to each precursor.

		Percentage from emission precursor (%)				
Source vehicle	Mortalities	NO_X	VOC	NH_3	SO_2	PPM
LDA	1153 (974–1341)	5	14	31	1	47
LDT	1234 (1042–1434)	12	15	26	1	46
MDT	601 (507–698)	6	4	4	0	85
HDT	829 (701–964)	11	1	4	0	83
BUS	206 (174–240)	3	3	3	0	91

Table 4. Total premature mortalities in the TCI region due to long-term exposure to O_3 attributable to each vehicle class and the percentage attributable to each precursor.

		Percentage from emission precursor (%)	
Source vehicle	Mortalities	NO_X	VOC
LDA	728 (364–1456)	69	31
LDT	1129 (615–2459)	80	20
MDT	395 (197–790)	90	10
HDT	636 (318–1272)	97	3
BUS	91 (45–181)	88	12

MDT and HDT $PM_{2.5}$ population-weighted sensitivities are largest with respect to PPM emissions consistent with what we see from the emission magnitudes as compared to LDT and LDA as well as emission percentages in each source region. BUS $PM_{2.5}$ population weighted sensitivities are the smallest of the vehicle classes with the only significant source of emissions from BUS coming from NY and VA.

O_3 population weighted sensitivities to NO_X emissions are larger than sensitivities to VOC for each vehicle class. O_3 sensitivities to LDT NO_X emissions are the largest at 6.3×10^{-2} ppb across the TCI region followed by sensitivities to HDT NO_X emissions at 4.0×10^{-2} ppb. O_3 sensitivities to LDA and LDT VOC emissions are much larger than sensitivities to MDT, HDT, and BUS VOC emissions reflecting the magnitude of VOC emissions from each vehicle class.

3.2. Health impacts

Tables 3 and 4 show the total $PM_{2.5}$ and O_3-attributable premature mortalities across the TCI region by vehicle class and precursor. Percentage of premature mortalities from each precursor follows similar trends to the magnitudes of population weighted exposures. The largest source of both $PM_{2.5}$ and O_3-attributable premature mortalities are LDT at 1234 and 1229 mortalities, respectively. LDT PPM emissions are responsible for 46% of $PM_{2.5}$ mortalities, and LDT NO_X emissions are responsible for 80% of O_3 mortalities. Tables S15–S26 show the breakdown of the total premature mortalities by source state. Looking at the largest $PM_{2.5}$ mortalities for each vehicle class, the largest percentage of mortalities occur in NY except for HDT where the largest percentage occurs in PA. When considering BUS emissions, 64.9% of premature mortalities occur in NY, with 62.1% of those mortalities from PPM emissions. This indicates that BUS PPM emissions in NY outweigh any other precursor and source region emissions from BUS. While not included in the total $PM_{2.5}$-attributable mortalities from states that make up the TCI region (table 3 and tables S15–S19), mortalities in the MSAs we considered were similar in amount to NY, and in some instances exceeded total mortalities in NY. $PM_{2.5}$ mortalities in NYMSA were 388.7, 354.1, 302.7, 251.8, and 137.1 for LDA, LDT, MDT, HDT, and BUS emissions, respectively, showing that more $PM_{2.5}$-attributable premature mortalities are occurring in the NYMSA due to each vehicle class rather than any individual state. This seems reasonable given the population density and vehicle activity in the counties that comprise the NYMSA. $PM_{2.5}$ mortalities for all four MSAs are in table S20.

Looking at the O_3 mortalities for each vehicle class, the largest percentage of mortalities occur in PA, except for BUS, where the largest percentage occurs in NJ (although the impacts are much smaller than the other vehicle classes due to the relatively small percentage of NO_X and VOC emitted from BUS). Total $PM_{2.5}$-attributable mortalities exceed O_3-attributable mortalities in the TCI region, however the largest individual impact is O_3-attributable mortalities in PA by LDT emissions.

Figures 1 and 2 allocate the $PM_{2.5}$ and O_3-attributable premature mortalities in each receptor state/MSA in the TCI region to each source state's/MSA's vehicle emissions. Tables S27 and S28 show the exact values for each source and receptor where in each column, the number in bold represents the largest impact from a given source state/MSA. Here we have summed mortalities by precursor to give an estimate of the total premature mortalities due

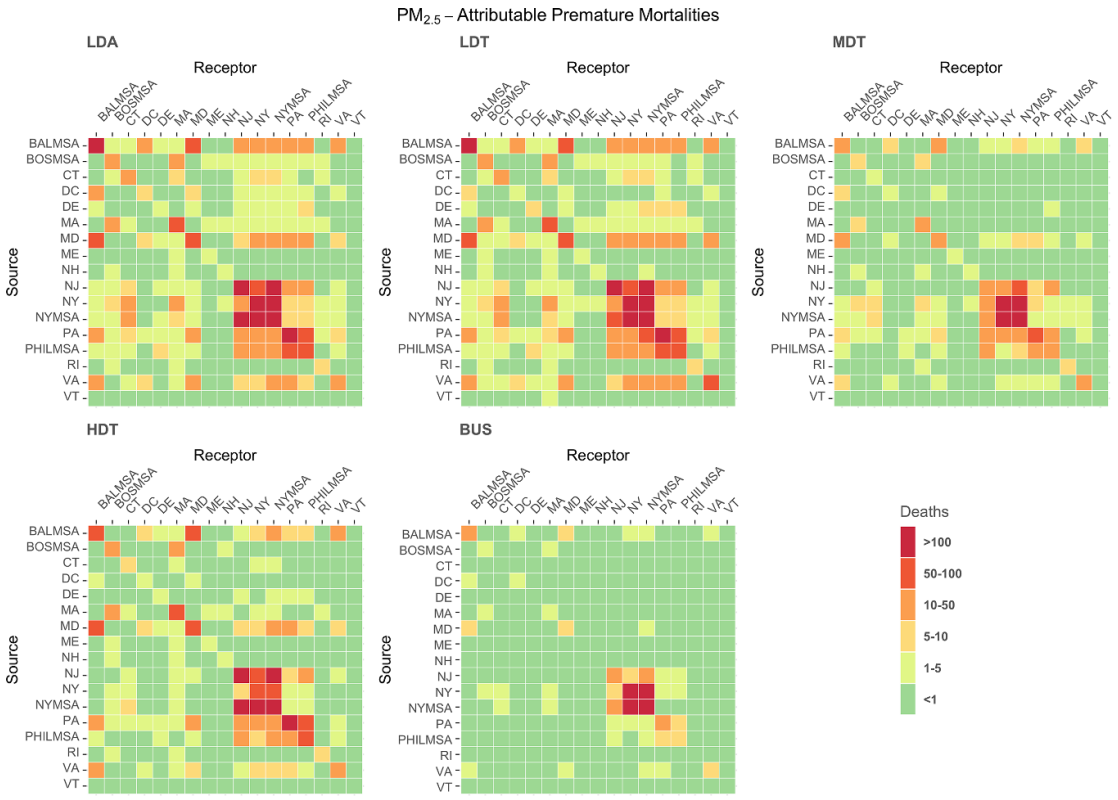

Figure 1. Premature deaths in each receptor state/MSA due to long-term exposure to $PM_{2.5}$ attributable to each source state/MSA and vehicle class.

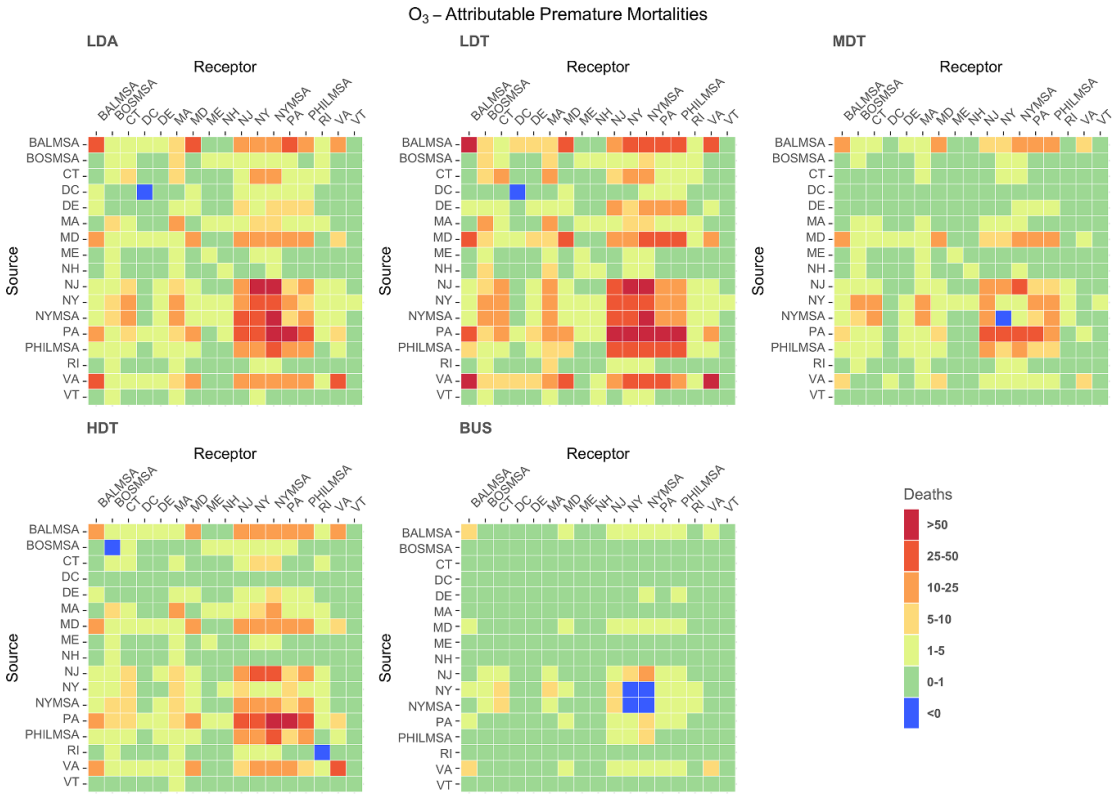

Figure 2. Premature deaths in each receptor state/MSA due to long-term exposure to O_3 attributable to each source state/MSA and vehicle class.

to the source state's/MSA's vehicle class. The largest amount of premature mortalities (335), from $PM_{2.5}$ or O_3, occurs in the NYMSA by emissions from LDA in NYMSA. As a source for $PM_{2.5}$-attributable mortalities, vehicles from MSAs make up 40% (34 of 85 possible instances) of the largest impacts seen in each receptor state/MSA. As a source for O_3-attributable mortalities, this number drops to 17.6% (15 of 85 possible instances). This is due to $PM_{2.5}$ impacts remaining localized to the emission source region, driven by large population-weighted sensitivities to NH_3 and PPM emissions, and O_3 impacts largely occurring downwind of the source. Large NO_X emissions in MSAs and highly populated regions can cause O_3 depletion due to the photochemical regime, which can be seen with small negative impacts (approximately −1 premature mortalities) in DC from all vehicle class in DC, in BOSMSA from MDT and BUS in BOSMSA, and in RI from MDT and HDT in RI; and can be seen with large negative impacts (less than −10 premature mortalities) in NY from MDT in NYMSA, and in NY and NYMSA from BUS in NY and NYMSA. The most impacted region by $PM_{2.5}$-attributable mortalities is NYMSA for each vehicle class from NYMSA. The most impacted region by O_3-attributable mortalities from LDA, MDT, HDT, and BUS is also NYMSA with LDA and BUS emissions from NJ and MDT and HDT emissions from PA. For LDT, PA is the most impacted with emissions from PA.

To monetize the value of avoided mortalities we apply a value of statistical life approach as recommended by the EPA (U.S Environmental Protection Agency 2010) by multiplying the number of $PM_{2.5}$ and O_3 attributable premature mortalities by a 2016 USD ($) income-adjusted value of $10.3 million. The monetized value of premature mortalities in each receptor state/MSA is divided by the emission amounts from each source region and vehicle class precursor to approximate the economic damages per ton of precursor emitted. Figure 3 shows the rank-ordered top 10 damages per ton for each receptor state/MSA, where total damages reflect the sum of $PM_{2.5}$ and O_3 attributable premature mortalities. By selecting only the top 10 damages per ton estimates in each receptor state/MSA, we are able to quantify the largest economic value of avoided deaths in each region with respect to any source region/vehicle class/precursor. Looking at precursors, we can see that the largest monetized value of health-related benefits can be achieved by reducing PPM and NH_3. As PPM and NH_3 are mainly responsible for localized impacts, each receptor state/MSA has itself as one of its largest contributing source regions in terms of damages per ton. The largest damages per ton is in NYMSA from BUS PPM in NYMSA at a little over $4 million. Figures S19–S25 show the top 5 damages per ton for each of the five precursors for $PM_{2.5}$ and two for O_3. The top 5 damages per ton for NO_X emissions to $PM_{2.5}$ range from $500 to $4500; VOC emissions to $PM_{2.5}$ range from $100 to $16 000; and SO_2 emissions to $PM_{2.5}$ range from $1300 to $81 000.

4. Discussion

On-road emission sector's impacts have been widely studied across the U.S.. Caiazzo *et al* (2013) quantified premature mortalities in the U.S. from road transportation and attributed 52 800 $PM_{2.5}$ and 5250 O_3 mortalities in 2005 from 2005 emissions, while a follow up to that study (Dedoussi and Barrett 2014) found 47 780 premature $PM_{2.5}$ mortalities. A recent study (Davidson *et al* 2020) attributed 9666 $PM_{2.5}$ and 1939 O_3 mortalities from 2011 emissions, compared to our findings of 4023 $PM_{2.5}$ and 2979 O_3 mortalities in 2016 from 2016 emissions in the TCI region. Davidson *et al* further evaluated outcomes based on vehicle class fuel types, where they attributed 2300 $PM_{2.5}$ and 410 O_3 mortalities to gas cars and motorcycles, and 2500 and 700 to light duty gas trucks. This compares to our findings that attribute 1153 $PM_{2.5}$ and 728 O_3 mortalities due to LDA, and 1234 $PM_{2.5}$ and 1129 O_3 mortalities due to LDT in the TCI region. Larger differences in O_3 mortalities in our study compared to Davidson *et al* can also be explained by different CRFs being used. Davidson *et al* utilized a seasonal O_3 mortality CRF while we use an annual O_3 mortality CRF. As a further comparison with Davidson *et al,* table S14 shows the total O_3 mortalities by vehicle class in the TCI region calculated with an annual O_3 mortality CRF (Turner *et al* 2016) and a pooled estimate from two studies (Zanobetti and Schwartz 2008, Levy *et al* 2012), that has been used in health benefits assessments for climate policies (Driscoll *et al* 2015, Buonocore *et al* 2016). Since we have used an annual O_3 mortality CRF, we made sure to assess the model performance against hourly O_3 measurements for both January and July with monitoring networks that had an adequate number of measurements for both months in 2016 in the TCI region (See figures S9–S11 and table S13). Dedoussi and Barrett (2014) evaluated results based on source states and found that in 2005 2827, 3982, and 3702 $PM_{2.5}$ mortalities were attributable to PA, NY, and NJ, respectively, compared to our findings which show 854, 1085, and 731 $PM_{2.5}$ mortalities from those same states in 2016. Estimates from Dedoussi and Barrett (2014) are larger than those estimated in this study likely due to 2005 mobile source emission inventories being larger than 2016. Dedoussi *et al* (2020) quantified total $PM_{2.5}$ and O_3 mortalities in each state due to on-road emissions from each other state. In 2011, they found on-road emissions from NY to be responsible for 1003 mortalities in NY and 115 in neighboring CT while we find the sum of our impacts from our five vehicle classes from NY to be responsible for 929 mortalities in NY and 112 in CT. Caiazzo *et al* evaluated outcomes in

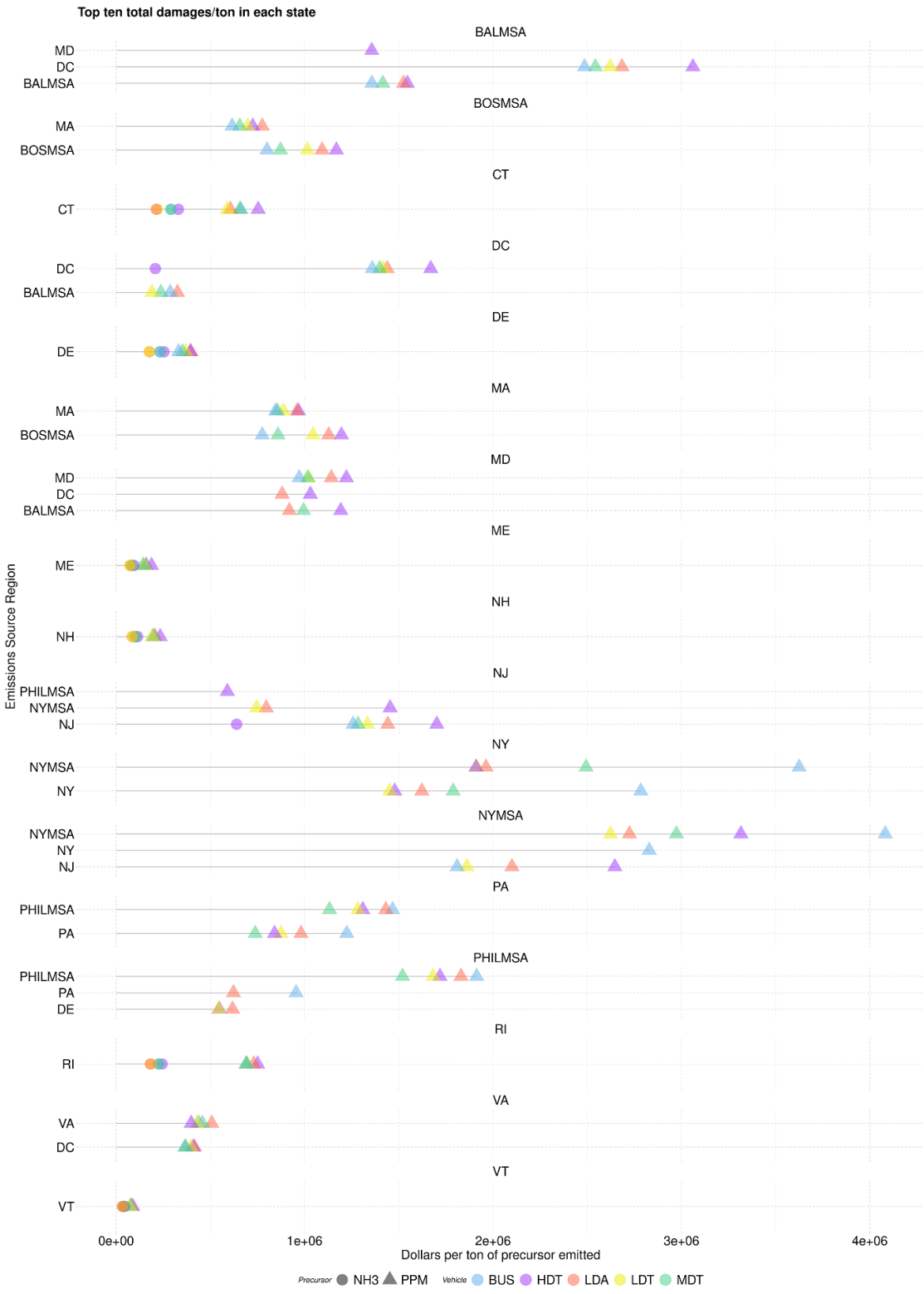

Figure 3. Top 10 damages per ton values for each receptor state/MSA.

some major metropolitan areas and found that 3615 $PM_{2.5}$ mortalities and 3.76 O_3 mortalities in NYMSA were attributable to on-road emissions. If we sum across vehicle class emissions and consider NYMSA vehicles only, we attribute 1283 $PM_{2.5}$ mortalities and 120.7 O_3 mortalities in NYMSA to on-road emissions. Wolfe *et al* (2019) monetizes $PM_{2.5}$ damages per ton of directly emitted $PM_{2.5}$, SO_2 /pSO_4, and NO_X from the same vehicle classes as Davidson *et al*. They find the same trend as we do in the TCI region, with NO_X emissions having the lowest damages per ton and PPM having the largest (table S34) with PPM estimates being two orders of magnitude larger than NO_X estimates for all vehicle classes. Although difficult to

compare across studies due to differences in study designs, Wolfe *et al* found that the average damages per ton of PPM from light duty gas trucks in the Eastern U.S. in 2025 to be $450 000. We estimated average LDT damages per ton of PPM in the TCI region to be $97 000. Goodkind *et al* (2019) calculated the total $PM_{2.5}$ damages in the U.S. in 2011 from light gas vehicles to be $94.1 billion while we found total $PM_{2.5}$ damages in 2016 in the TCI region from LDA and LDT to be $25 billion.

While Wolfe *et al* and Caiazzo *et al* look at impacts by precursors, they exclude on-road emissions of NH_3 in their analyses. We show that on-road emissions of NH_3 are the second largest contributor to regional on-road attributable—$PM_{2.5}$ concentrations, absolute health impacts (table 3), and damages per ton (figure 3) in the TCI region. NH_3 is emitted from vehicles as a by-product of catalytic technologies used in gasoline light duty vehicles (Suarez-Bertoa *et al* 2014, Li *et al* 2020) and diesel particle filters in heavy-duty diesel trucks (Preble *et al* 2019). Recent studies have assessed the importance of vehicular NH_3 as a contributor to $PM_{2.5}$ concentrations in urban regions (Chang *et al* 2016, 2019, Sun *et al* 2017, Osada *et al* 2019, He *et al* 2020). Dedoussi and Barrett (2014) found that reducing 1 Tg of NH_3 emissions from road transportation is ∼20 times greater in terms of damages reductions than a 1 Tg reduction of NO_X across the U.S.. We find that in the TCI region reducing 1 ton of NH_3 emissions from LDT is ∼75 times and from HDT is ∼90 times greater in terms of damages reductions than a 1 ton reduction of NO_X. Hence, any strategy aimed at reducing the on-road vehicle sector's impact on air pollution in urban areas must consider reducing vehicular NH_3 emissions.

In this study we have accounted for uncertainties in the premature mortality calculations by including 95% confidence intervals for each CRF. The confidence intervals reflect the variability in the results from the epidemiological studies used to construct the CRFs. We also accounted for uncertainty in the valuation of the health impacts by including lower and upper bounds for the 2016 USD ($) income-adjusted value of $10.3 million as given in S5. Tables S29–S33 show the valuation results with these lower and upper bounds for each vehicle class and source state's impact on the TCI region. We have chosen not to include uncertainty estimates, either from the confidence intervals of the CRFs or from the bounds on the valuation amount, in the main results of this study which are the individual source/receptor impacts in figures 1–3 (tables S27 and S28); as the focus of these results is the intercomparison between precursor emissions by vehicle class and source region. The correlated uncertainties associated with each of these source/receptor estimates do not vary between the variables we are studying. We did not account for uncertainty in the CMAQ model output or emission inventory estimates as this would have been too computationally demanding given the study framework. However, this can be an extension for a future study.

We make use of DDM in this study to represent the impacts from entire emission sources. This is not the same as source apportionment and brute force methods. For an emissions-to-pollutant system that is highly nonlinear, DDM and brute force methods will give varying results. However, the on-road vehicle sector has been shown to be approximately linear up to a 100% perturbation for primary PM, secondary inorganic PM, and secondary organic aerosols (Koo *et al* 2009). Another point to consider with DDM is that by summing results from individual emission sectors we may be missing any inherent nonlinearity of the interactions between emission sectors. However, when summing health impact results we are confident the nonlinearities from summing emission sources are within the confidence intervals of the CRFs. The benefit of DDM is that sensitivities allow for additional perturbations in individual emission sectors to model the expected concentration change without re-running the model. We have chosen to run each simulation for two months of the year to represent seasonal variations similar to work done by Penn *et al* using CMAQ-DDM to quantify impacts from residential combustion and electricity generating unit emissions (Penn *et al* 2017) and aviation emissions (Penn *et al* 2017b); as the computational requirements to run CMAQ-DDM to compute O_3 and $PM_{2.5}$ sensitivities for each source region/vehicle class/precursor emission species at this grid resolution are immense. Future work will utilize these sensitivities to model impacts of emission reduction strategies as dictated by illustrative policies in the TCI region, as well as to develop new policies for consideration. This study developed a comprehensive source-receptor impact analysis for each state in the TCI region to determine the full impact from multiple vehicle classes and precursor emissions. The results in this study can help inform the design of effective emission perturbation strategies aimed at reducing air pollution and adverse health effects in individual states and MSAs, and further provide guidance for similar analyses to be performed for other source sectors.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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