CRC Report No. A-76-1

Effects of Light-duty Vehicle Emissions on Ozone and PM with Past, Present, and Future Controls

Final Report

February 2012

COORDINATING RESEARCH COUNCIL, INC.

3650 MANSELL ROAD·SUITE 140·ALPHARETTA, GA 30022

The Coordinating Research Council, Inc. (CRC) is a non-profit corporation supported by the petroleum and automotive equipment industries. CRC operates through the committees made up of technical experts from industry and government who voluntarily participate. The four main areas of research within CRC are: air pollution (atmospheric and engineering studies); aviation fuels, lubricants, and equipment performance, heavy-duty vehicle fuels, lubricants, and equipment performance (e.g., diesel trucks); and light-duty vehicle fuels, lubricants, and equipment performance (e.g., passenger cars). CRC's function is to provide the mechanism for joint research conducted by the two industries that will help in determining the optimum combination of petroleum products and automotive equipment. CRC's work is limited to research that is mutually beneficial to the two industries involved, and all information is available to the public.

Effects of Light-duty Vehicle Emissions on Ozone and PM with Past, Present, and Future Controls

CRC A-76-1
FINAL REPORT

Prepared for:
Coordinating Research Council, Inc.
3650 Mansell Road, Suite 140
Alpharetta, GA 30022

Prepared by:
ENVIRON International Corporation
773 San Marin Drive, Suite 2115
Novato, California, 94945
P-415-899-0700
F-415-899-0707

February 29, 2012

ENVIRON Project Number:
06- 27365B

EXECUTIVE SUMMARY

Light duty vehicle (LDV) emissions standards in the US have become increasingly stricter since the 1970s and more stringent vehicle emission regulations are being considered by agencies to attain compliance with national ambient ozone and fine particulate matter ($PM_{2.5}$) standards. We present a computer modeling study of the impact of past, present and potential future US Federal emissions standards for on-road gasoline-fueled LDVs (cars and light trucks) on ozone and $PM_{2.5}$ concentrations in the eastern US, with focus on four urban areas – Atlanta, Detroit, Philadelphia and St. Louis.

The Motor Vehicle Emission Simulator (MOVES, version 2010a) was used for deriving on-road vehicle emissions. This was complemented by a suite of other advanced emissions models for developing other source inventories. Air quality modeling was performed with the Comprehensive Air Quality Model with Extensions (CAMx, version 5.40). Modeling was conducted over a 12 km horizontal resolution domain in the eastern US nested within a 36 km continental US (CONUS) domain. In addition to a 2008 base case used for model performance evaluation, we modeled four hypothetical 2022 LDV emissions scenarios:

1. 2022 Tier 1 scenario (assume that only Tier 1 standards are implemented in 2022)
2. 2022 Tier 2 scenario (assume that no standards beyond Tier 2 are implemented in 2022)
3. 2022 LEV III scenario (the draft proposed California LEV III standard is adopted nationwide)
4. 2022 LDV zero-out scenario (assume there are no LDV emissions in 2022)

All simulations were conducted for a winter month (February) and summer month (July).

Model performance evaluations against atmospheric measurements in 2008 indicated that meteorological and air quality model performance was satisfactory.

MOVES was run in 220 representative counties in the US for calendar years 2008 and 2022 for vehicle ages 0-30 to develop on-road vehicle emissions for the 2008 base case and 2022 Tier 2 scenario. Tier 1 emission factors for the 2001-2022 vehicle model years do not exist by default in MOVES and were simulated as existing in 2022 by running multiple historic calendar years in MOVES keeping all other model assumptions the same as in 2022. Ratios of LEV III to LEV II emissions were calculated using EMFAC2007 and then used to adjust MOVES model LEV II emission rate input estimates to reflect LEV III emission rates. A complete set of per-mile emission rates by pollutant, county, Source Classification Code (SCC), and emission process was developed for February and July 2008 and 2022 (four scenarios). These emission rates were applied with 2008 and 2022 estimates of VMT in each month to develop by-county and SCC emissions of VOC, CO, NOx, PM_{10}, $PM_{2.5}$, SO_2 and NH_3. More detailed analysis was conducted for the Atlanta, Detroit and Philadelphia metropolitan areas using county specific age distributions obtained from the respective MPOs and I/M and fuels data available in MOVES. St.

Louis was modeled similar to the rest of the country due to non-availability of local data at the start of this project. We used the MOVES default fuel market share for 2008 but assumed 100% E10 usage for all counties in 2022 to avoid spatial differences in air quality model results. Emissions for other source categories were developed either from EPA's 2008 National Emissions Inventory (NEI), the 2020 NEI or using models such as NMIM for off-road sources and MEGAN for biogenic emissions. VOC speciation profiles were obtained from the EPA SPECIATE 4.3 database.

The on-road fraction of the national anthropogenic emissions inventory decreases by half or more from 2008 to the 2022 Tier 2 scenario for three key ozone and $PM_{2.5}$ precursors: NOx, VOC and primary $PM_{2.5}$ while the corresponding fractions of many other source categories are constant or increase. The on-road fractions for SO_2 and NH_3 are very small and hence LDV emissions of these two species have limited contributions to $PM_{2.5}$. When considering total emissions in each of the four metropolitan areas, wintertime LDV NOx emissions are highest in the Atlanta area in all 2022 scenarios. Wintertime VOC and primary $PM_{2.5}$ emissions are highest in the Detroit area due, in part, to the increase in cold start emissions. In summer, Atlanta has the highest LDV emissions of NOx, VOC and $PM_{2.5}$, due to a combination of higher ambient temperatures and higher VMT. LDV NOx emissions in all four metropolitan areas decrease by more than 70% from Tier 1 to Tier 2 and only by 4% from Tier 2 to LEV III. Similarly, VOC emissions decrease by approximately 60% or more from Tier 1 to Tier 2 and by 6–9% in the transition from Tier 2 to LEV III.

The modeling results show that large benefits in ozone and $PM_{2.5}$ (up to 16 ppb reductions in daily maximum 8-hr ozone, up to 10 ppb reductions in the monthly mean of daily maximum 8-hr ozone, up to 4.5 $\mu g/m^3$ reductions in maximum 24-hr $PM_{2.5}$ and up to 2.1 $\mu g/m^3$ reductions in the monthly mean of $PM_{2.5}$) accrued from the transition from the Tier 1 to Tier 2 LDV standards. However, the implementation of additional LDV controls similar to LEV III would result in very small additional improvements in air quality (up to 0.3 ppb reductions in daily maximum 8-hr ozone, up to 0.2 ppb reductions in the monthly mean of daily maximum 8-hr ozone, up to 0.1 $\mu g/m^3$ reductions in maximum 24-hr $PM_{2.5}$, and up to 0.1 $\mu g/m^3$ reductions in the monthly mean of $PM_{2.5}$) in 2022. Some additional improvements would be realized after LEV III phases in fully in 2028. The complete elimination of LDV emissions in 2022 is predicted to result in improvements in air quality (up to 7 ppb reductions in daily maximum 8-hr ozone, up to 4 ppb reductions in the monthly mean of daily maximum 8-hr ozone, up to 2.9 $\mu g/m^3$ reductions in maximum 24-hr $PM_{2.5}$, and up to 1.8 $\mu g/m^3$ reductions in the monthly mean of $PM_{2.5}$) from Tier 2 levels, that are generally smaller than the improvements obtained in switching from Tier 1 to Tier 2.

Journal Manuscript

Submitted to Atmospheric Environment

Effects of Light Duty Gasoline Vehicle Emission Standards in the United States on Ozone and Particulate Matter

Krish Vijayaraghavan*, Chris Lindhjem, Allison DenBleyker, Uarporn Nopmongcol, John Grant, Edward Tai, Greg Yarwood

ENVIRON International Corporation, 773 San Marin Drive, Suite 2115, Novato, CA 94998, USA

*Corresponding author: krish@environcorp.com, 415-899-0700

Abstract

More stringent motor vehicle emission standards are being considered in the United States to attain national air quality standards for ozone and $PM_{2.5}$. We modeled past, present and potential future US emission standards for on-road gasoline-fueled light duty vehicles (including both cars and light trucks) (LDVs) to assess incremental air quality benefits in the eastern US in 2022. The modeling results show that large benefits in ozone and $PM_{2.5}$ (up to 16 ppb reductions in daily maximum 8-hr ozone, up to 10 ppb reductions in the monthly mean of daily maximum 8-hr ozone, up to 4.5 $\mu g/m^3$ reductions in maximum 24-hr $PM_{2.5}$ and up to 2.1 $\mu g/m^3$ reductions in the monthly mean $PM_{2.5}$) accrued from the transition from Tier 1 to Tier 2 standards. However, the implementation of additional nationwide LDV controls similar to draft proposed California LEV III regulations would result in very small additional improvements in air quality by 2022 (up to 0.3 ppb reductions in daily maximum 8-hr ozone, up to 0.2 ppb reductions in the monthly mean of daily maximum 8-hr ozone, up to 0.1 $\mu g/m^3$ reductions in maximum 24-hr $PM_{2.5}$, and up to 0.1 $\mu g/m^3$ reductions in the monthly mean $PM_{2.5}$). The complete elimination of gasoline-fueled LDV emissions in 2022 is predicted to result in improvements in air quality (up to 7 ppb reductions in daily maximum 8-hr ozone, up to 4 ppb reductions in the monthly mean of daily maximum 8-hr ozone, up to 2.8 $\mu g/m^3$ reductions in maximum 24-hr $PM_{2.5}$, and up to 1.8 $\mu g/m^3$ reductions in the monthly mean $PM_{2.5}$) from Tier 2 levels, that are generally smaller than the improvements obtained in switching from Tier 1 to Tier 2.

Keywords: LEV, Tier 2, LDV, CAMx, MOVES, ozone, PM2.5

1. Introduction

Emissions from on-road motor vehicles in the United States (US) have decreased significantly over the past four decades even with increases in traffic volume. For example, highway vehicle emissions of volatile organic compounds (VOCs) decreased by approximately 75% from 1970 to 2005 and emissions of particulate matter (PM) and nitrogen oxides (NOx) decreased by over

50% though total Vehicles Miles Traveled (VMT) for highway vehicles increased more than two-fold (Kryak et al., 2010). These emissions reductions have been due, in large part, to increasingly stricter emissions and fuel standards for gasoline-fueled light duty vehicles (LDVs) in the US since the 1970s. The aim of these standards is to improve ambient air quality as emissions of VOCs, NOx and PM from LDVs are often key precursors to ambient ozone (O_3) and fine particulate matter ($PM_{2.5}$). With the potential lowering of the National Ambient Air Quality Standards (NAAQS) for 8-hour O_3 and $PM_{2.5}$, States would likely seek additional means to reach or stay in O_3 and PM attainment including possibly adopting more severe LDV emission standards. Therefore, it is of interest to understand the incremental O_3 and $PM_{2.5}$ benefits of past and current LDV emissions standards and the additional air quality benefits of potential future LDV emissions standards in the US.

While other modeling studies have analyzed the contribution of motor vehicles to O_3 and/or $PM_{2.5}$ concentrations and the impact of vehicle fuel and emissions controls on these concentrations (e.g., EPA, 1999; Mathes et al., 2007; Koffi et al., 2010; Nopmongcol et al., 2011; Roustan et al. 2011; Collet et al., 2012), the current work provides a cohesive analysis of the effect of historical, current, and potential future LDV emissions standards on O_3 and $PM_{2.5}$ in the US. We apply state-of-the-science emissions models and an advanced regional 3-D photochemical air quality model that simulates transport and dispersion, atmospheric chemical transformation, and deposition to the earth's surface of trace gases and aerosols, to estimate impacts of different LDV emissions standards on ozone and primary and secondary PM in the eastern US. A 2008 baseline is used for air quality model performance evaluation. Four future year emissions scenarios with increasingly stricter emission standards for gasoline-fueled LDVs are compared against each other to estimate the incremental and cumulative effect of LDV emissions controls on ambient air quality.

2. Methods

2.1 Modeling Domain and Emissions Scenarios

The air quality simulations were conducted with the Comprehensive Air Quality Model with Extensions (CAMx) (ENVIRON, 2011) using on-road emissions inventories derived using the Motor Vehicle Emission Simulator (MOVES) (EPA, 2010a) and other model inputs as discussed below. We applied version 5.40 of CAMx with the Carbon Bond 5 (CB05) chemical mechanism and version 2010a of MOVES.

The geographic region studied here includes part of the eastern US with focus on four of thirteen urban areas discussed in EPA's PM Risk Assessment analysis (EPA, 2010b). The four areas selected are Atlanta, Detroit, Philadelphia and St. Louis. The CAMx modeling domain extends over the continental US (CONUS) at 36 km horizontal resolution with an inner nested domain at 12 km resolution over part of the eastern US including the four urban areas of interest. The domain and four urban areas are shown in Fig. 1. The domain has a pressure-based vertical structure with 26 layers with the model top at 145 mb or approximately 14 km above mean sea level.

To study the effect of historical, current and additional LDV emissions controls, we modeled a 2008 base case and four 2022 LDV emissions scenarios. 2008 was chosen as the baseline modeling year due to the availability of emissions from the National Emissions Inventory (NEI) (EPA, 2011a). The 2008 base case is used for air quality model performance evaluation. The four 2022 LDV scenarios modeled are:

1. 2022 Tier 1 scenario (assume that only US Tier 1 standards are implemented through 2022)
2. 2022 Tier 2 scenario (assume that the current emissions standards, up to US Tier 2 standards, are implemented through 2022)
3. 2022 LEV III scenario (assume that the draft proposed California LEV III standard is adopted nationwide)
4. 2022 LDV zero-out (LDVZ) scenario (assume there are no gasoline-fueled LDV emissions in 2022)

All simulations were conducted for a winter month (February) and summer month (July).

The 2022 Tier 1 scenario aims to answer the question: "what if the US had not switched from Tier 1 to Tier 2 standards by 2022?" The 2022 Tier 2 case reflects a scenario with current Tier 2 emissions standards that are not revised through 2022. The 2022 LEV III scenario addresses the potential impact of further tightening LDV emission standards from Tier 2 to a nationwide LEV III standard. Emissions from all sources other than gasoline-fueled LDVs are held constant across the four 2022 scenarios.

The Tier 1 program instituted standards for Total Hydrocarbons, carbon monoxide (CO), NOx, and PM for 1994-2003 model year vehicles with a phase-in for the early years. Tier 2 applied to model years 2004 onwards and phased in completely in 2009. The draft proposed California LEV III standards will apply to vehicle model years 2015 to 2028. The exhaust emission standards for the Tier 1 and 2 programs for gasoline-fueled LDVs and the draft proposed California LEV III standards are shown in Table S1.1 (where "S" refers to Supplementary Data).

2.2 Meteorology

CAMx modeling for 2008 and the 2022 scenarios was driven by year 2008 meteorological fields from the Weather Research and Forecast (WRF) model – Advanced Research WRF (ARW) core (Skamarock et al., 2008). WRF output meteorological fields at 12 km horizontal resolution over the CONUS were obtained from the EPA (Gilliam, R., personal communication, 2011) and converted to CAMx input meteorological files for the nested 36 and 12 km resolution domains. Data in 34 WRF vertical layers extending up to 50 mb altitude were mapped to 26 layers in CAMx extending up to 145 mb. The WRF and CAMx vertical grid structure and mapping from WRF to CAMx layers are shown in Table S4.1. A limited performance evaluation of the WRF meteorological outputs and CAMx-ready meteorology showed satisfactory performance (see S4. in Supplementary Data for additional information).

2.3 On-road motor vehicle emissions

MOVES 2010a was used to prepare on-road emissions inventories in the CONUS for the 2008 base case and the four 2022 emissions scenarios. MOVES was run for calendar years 2008 and 2022 for vehicle ages 0-30 to develop on-road vehicle emissions for the 2008 base case and 2022 Tier 2 scenario. Tier 1 emission factors for vehicle model years after 2000 do not exist by default in MOVES and were simulated as existing in 2022 by running multiple historic calendar years in MOVES keeping all other model assumptions the same as they are in 2022. Ratios of LEV III to LEV II emissions calculated using simulations with the California Air Resources Board's Emissions Factor Model (EMFAC2007) (http://arb.ca.gov/msei/onroad/latest_version.htm, accessed August 2011) were used to adjust MOVES model LEV II emission rate input estimates to calculate emission rates for the 2022 LEV III scenario. On-road emissions in the zero-out LDV scenario were computed by setting emissions of Source Classification Codes (SCCs) corresponding to gasoline-fueled LDVs to zero in the 2022 Tier 2 emissions. Detailed information on the calculation of on-road emissions in the various scenarios is provided in the Supplementary Data.

The on-road emissions for winter and summer from MOVES for all emissions scenarios were speciated to CAMx model species, temporally allocated to hourly emissions, and spatially allocated to grid cells using version 2.7 of the Sparse Matrix Operator Kernel Emissions (SMOKE) model. Average day emissions were adjusted to account for day-of-week and hour-of-day effects based on SCC codes. Emission estimates for total VOC were converted to the CB05 chemical mechanism in CAMx using VOC speciation profiles derived from EPA's SPECIATE database, version 4.3 (EPA, 2011b) (see Table S5.1). PM emissions were speciated to CAMx model species, namely primary organic aerosol, primary elemental carbon, primary nitrate, primary sulfate, primary fine other PM, and coarse PM following methods outlined by Baek and DenBleyker (2010). On-road mobile sources generated using MOVES at the county level were allocated to CAMx 36 km and 12 km grid cells using spatial surrogates derived with the Spatial Surrogate Tool (http://www.epa.gov/ttn/chief/emch/spatial/spatialsurrogate.html, accessed August 2011).

2.4 Other emissions

Emissions from anthropogenic area and point sources in 2008 in the CONUS were developed from version 1.5 of the 2008 NEI (EPA, 2011a). Emissions from these source categories for the 2022 emissions scenarios were prepared from the 2020 NEI inventory (EPA, 2010c) and held constant from 2020 to 2022. The 2020 NEI was developed by EPA by projecting the 2005-based v4 modeling platform emissions to 2020. Anthropogenic area and point emissions for Canada for the 2008 base case and 2022 scenarios were prepared from the 2005 NEI (EPA, 2011c) and the 2020 NEI, respectively. Anthropogenic area and point emissions for Mexico for the 2008 base case were prepared from the 2005 NEI and held constant between the 2008 and 2022 scenarios due to lack of additional information.

We developed 2008 non-road mobile source emissions in the CONUS from the 2008 NEI. The NEI non-road emissions are based on the National Mobile Inventory Model (NMIM) using county specific fuel properties, meteorological parameters, and non-default local activity data for areas where such activity data has been provided to EPA as part of its NEI development efforts. We used the NMIM model to generate county level estimates of 2022 non-road emissions in the CONUS for February and July. 2008 emissions from locomotives/harbor craft, aircraft and commercial marine vessels were also obtained from the 2008 NEI. 2022 emissions from locomotives/harbor craft, aircraft and commercial marine vessels were obtained from the 2020 NEI and forecast two years through 2022 following forecast methods applied by EPA (2008a), FAA (2010) and EPA (2009), respectively.

Biogenic emissions in 2008 across the CONUS and the parts of Canada and Mexico in the CAMx 36 km domain were developed using the Model of Emissions of Gases and Aerosols from Nature (MEGAN v. 2.04; Guenther et al., 2006). MEGAN uses gridded emission factors that are based on global datasets for 11 species (CO, nitric oxide, isoprene and other VOCs) and 4 functional plant types, and plant leaf area index. Biogenic emissions were held constant from 2008 to 2022. Wildfire emission inventories of CO, NOx, VOCs, SO_2, NH_3 and PM in North America for 2008 were derived from the Blue Sky Framework SMARTFIRE database (http://www.getbluesky.org/smartfire) and processed using version 3.12 of the Emissions Processing System (EPS) tool (ENVIRON, 2009). Wildfire emissions were held constant in all emissions scenarios. Sea salt emissions inventories of particulate sodium, chloride and sulfate for 2008 were prepared using the meteorological fields driven by WRF (temperature, pressure, winds) and land cover information. Sea salt emissions were also not altered from the 2008 to 2022 scenarios.

The emissions inventories described above were converted to speciated, gridded, temporally varying emissions files suitable for air quality modeling with CAMx in the nested 36/12 km domains following standard emissions processing methods described in the literature (e.g., Morris et al., 2007, Morris et al. 2008).

2.5 Other model inputs

Boundary concentrations of O_3, PM components and precursors for February and July 2008 (in addition to a 15-day model spin-up in each case) for the CAMx 36 km domain were derived from the global chemical and transport model, Model for Ozone and Related Chemical Tracers (MOZART) version 4.6 (Emmons et al., 2010). Six-hourly model outputs in a latitude-longitude coordinate system with a spatial resolution of about 2.8^{o} for both latitude and longitude and 28 vertical layers were mapped onto the CAMx domain and speciated for the CB05 chemical mechanism.

The landuse/landcover (LULC) databases used in biogenic emissions inventory preparation and CAMx modeling were obtained from the National Land Cover Dataset (NLCD) (http://www.mrlc.gov/nlcd06_data.php, accessed July 2011). The data were processed and mapped to the 26 landuse categories in the dry deposition scheme of Zhang et al. (2003) used in CAMx. Photolysis rates required for ozone modeling were developed using the CAMx photolysis rate pre-processor, which incorporates the Tropospheric Ultraviolet and Visible (TUV) radiative transfer model (NCAR, 2011).

3. Results and Discussion

3.1 Emissions and Air Quality in 2008

3.1.1 Emissions

Fig. 2 presents the total anthropogenic emissions estimated in the CONUS and the fractions of the major source categories in February and July 2008. The sectors shown include area sources (comprising residential, commercial and small industrial sources), electric generating units (EGU), stationary point sources other than EGUs (abbreviated here as non-EGU Pt), off-road sources, LDVs and other on-road sources. The modeled emission totals across the CONUS are generally consistent with totals provided by EPA for the NEI (http://neibrowser.epa.gov, accessed September 2011); differences are mainly in the on-road sector. EPA developed the NEI on-road sector emissions data using the NMIM, which uses the MOBILE6 vehicle emissions model whereas this study uses the more current MOVES model. The on-road fraction (LDV plus others) of the total 2008 US anthropogenic inventory varies considerably across pollutants; it is high for CO (52–60%) and NOx (40–41%) and very low for SO_2 (< 0.5%). Pollutants exhibit seasonal effects. Total CO emissions decrease by 14% from winter to summer; this is primarily due to a 25% seasonal decrease in on-road emissions associated partly with fewer cold starts in summer. Total NH_3 emissions increase more than two-fold from winter to summer. This is due, in part, to higher dairy NH_3 emissions in summer than winter (Pinder et al., 2004). Primary $PM_{2.5}$ emissions from LDVs decrease from winter to summer due to the increase in ambient temperatures as discussed below.

The modeled spatial distribution of on-road emissions in the eastern US in the 2008 base case shows the urban signature of on-road emissions, in particular in Atlanta, Chicago, Detroit, Indianapolis, St. Louis and along the eastern seaboard (see Fig. S7.1). NOx emissions are higher in summer than winter (by 5–10% or more) because higher running exhaust NOx in summer more than compensate for higher cold start emissions in winter. However, on-road emissions of VOCs and $PM_{2.5}$ decrease from winter to summer by up to 20% to 30% in some urban areas such as in the New York/New Jersey. These seasonal trends are also evident in the 2008 emissions inventory for gasoline-fueled LDVs both across the CONUS (Table 1) and in the four urban areas of interest (Table 2).

Table 1 also shows the LDV fraction of total on-road and total anthropogenic emissions. Gasoline-fueled LDV emissions of NOx and VOC constitute ~20% of total anthropogenic emissions in 2008 and, hence, are important to studying the potential contribution of LDVs to ambient O_3 and $PM_{2.5}$. Due to their slow reactivity, CO emissions have a much more limited effect on O_3 concentrations. While primary $PM_{2.5}$ emissions from LDVs can directly affect ambient $PM_{2.5}$, these represent a very small fraction (2%) of the total anthropogenic inventory; there is a much larger PM contribution from stationary sources, wood-burning, non-road sources, road dust, and other sources. LDV emissions of NH_3 and SO_2 constitute a large fraction (70–90%) of total on-road emissions. However, they represent a very small fraction (0.3 –5%) of total anthropogenic emissions due to the dominance of other sources such as livestock farming and fuel combustion.

St. Louis has the highest NOx, VOC and $PM_{2.5}$ emissions among the four urban areas as shown in Table 2 (values shown represent the total across the counties in each metropolitan area). However, the use of MOVES default age distributions rather than local data on vehicle age distributions for this area could have introduced uncertainty in our estimates. For example, we determined that using local age distributions for Atlanta, Detroit and Philadelphia resulted in modeled VOC emissions that were approximately 10% lower (see Fig. S2.3). Atlanta has the highest NOx and VOC LDV emissions among the three urban areas where local vehicle age distributions were used in MOVES modeling. In all four urban areas, $PM_{2.5}$ emissions are higher in winter than summer by 75% or more. Vehicle testing in Kansas City has shown that PM emissions increase exponentially as temperature decreases with the effect more pronounced for cold starts (EPA, 2008b).

3.1.2 Air Quality in 2008

Fig. 3 shows the predicted monthly mean of daily maximum 8-hour average O_3 concentrations in winter and summer 2008 in the CONUS at 36 km model resolution and in the eastern US at 12 km resolution. As expected, O_3 levels are low (< 50 ppb) in February due to limited solar radiation and photochemical activity except in Colorado and other parts of the western US where O_3 formation may be enhanced by shallow inversion with limited mixing and snow cover with high albedo. In July, the predicted monthly mean of daily maximum 8-hr O_3 goes up to 95 ppb in the CONUS (with the highest value in the Los Angeles basin) and up to 91 ppb over the eastern US (near Washington, D.C.). The predicted monthly averages of daily maximum 8-hr O_3 in July 2008 are 83 ppb, 59 ppb, 82 ppb and 73 ppb in Atlanta, Detroit, Philadelphia and St. Louis, respectively.

Fig. 4 shows the predicted monthly mean concentrations of $PM_{2.5}$ mass in February and July 2008 in the CONUS and eastern US. Figures S8.1 and S8.2 show similar plots for $PM_{2.5}$ components in February and July, respectively. The exceptionally high $PM_{2.5}$ concentrations predicted in northern California (> 100 $\mu g/m^3$) in July 2008 are due to emissions from extreme wildfire events in this region. $PM_{2.5}$ sulfate concentrations are higher in the eastern US in summer than in winter due to enhanced formation from SO_2 emissions. With the exception of southern Georgia, organic carbon is generally higher in summer in the Southeast due, in part, to higher biogenic emissions. $PM_{2.5}$ nitrate is higher in winter in the upper Midwest caused, in part, by a stronger partitioning of total nitrate towards the aerosol phase at lower temperatures. Winter $PM_{2.5}$ concentrations exceed 30 $\mu g/m^3$ in Georgia, the Chicago metropolitan area and parts of the Northeast. The four urban areas of interest in this study all show comparable monthly averaged $PM_{2.5}$ concentrations of ~25–27 $\mu g/m^3$ in February 2008. Primary organic aerosol (POA) makes up the largest portion of predicted $PM_{2.5}$ mass in Atlanta in both winter and summer while nitrate is the major $PM_{2.5}$ component in Detroit, Philadelphia and St. Louis in both seasons.

The 2008 CAMx base case predictions of 1-hour and 8-hour average ozone concentrations were evaluated against measurements in the AIRS/AQS network (EPA, 2002) and the Clean Air Status and Trends Network (CASTNET, 2011). Model predictions of $PM_{2.5}$ mass and components were compared to daily (24-hour) average measurements in the AIRS/AQS and IMPROVE (IMPROVE, 1995) networks. Overall, model performance was good both for ozone and $PM_{2.5}$ mass and components. Details are provided in the Supplementary Data.

3.2. Emissions and Air Quality in 2022 Scenarios

3.2.1 Emissions

The total CONUS anthropogenic emissions and the relative contributions of the major source sectors in the 2022 Tier 2 scenario are shown in Fig. 5. Emissions from source sectors other than on-road sources are held constant between this scenario and all other 2022 scenarios. Between 2008 and 2022, total anthropogenic CONUS emissions are projected to decrease by

37% for NOx, 14% for VOC and 20% for CO (see Fig. 2 and 5). Differences between the 2020/2022 and 2008 inventories are due to both growth and control as well as differences in methodologies between the 2005 inventory (used by EPA to project to 2020) and the 2008 inventory. The reductions from 2008 to 2022 are achieved, in large part, due to large reductions in the on-road inventory reflecting a mature Tier 2 LDV program by 2022. When considering the average across February and July, the on-road fraction of the CONUS NOx anthropogenic inventory decreases from 41% in 2008 to 21% in 2022, while the corresponding fractions of many of the other source categories are constant or increase. For example, the off-road fraction of the CONUS anthropogenic NOx inventory stays constant at ~25% from 2008 to 2022. On-road emissions of other pollutants also show a more than proportional reduction from 2008 to 2022 (when compared with the reduction in the total inventory) without considering any controls beyond Tier 2 (e.g., on-road VOC emissions decrease by 63%, $PM_{2.5}$ by 59% and CO by 31%).

Table 1 shows estimates of emissions from gasoline-fueled LDVs in the CONUS in the 2022 Tier 1, Tier 2 and LEV III scenarios. The 2022 Tier 1 scenario represents a hypothetical scenario where no LDV controls beyond Tier 1 are implemented through 2022. LDV emissions decrease considerably from Tier 1 to Tier 2 and then decrease only slightly from the Tier 2 to LEV III scenarios. For example, on average across winter and summer, LDV NOx emissions are reduced by 75% from Tier 1 to Tier 2 and by only 4% from Tier 2 to LEV III. The LDV fraction of the total anthropogenic inventory also decreases considerably from Tier 1 to Tier 2 (e.g., by 32% to 10% for NOx and 17% to 8% for VOC on average across winter and summer) and subsequently only marginally from Tier 2 to LEV III (with the NOx fraction decreasing to 9.9% and VOC to 7.2%). The corresponding predicted spatial distributions of winter and summer weekday on-road emissions of NOx, VOC and $PM_{2.5}$ in the CAMx 12 km domain in the 2022 scenarios are presented in the Supplementary Data.

Table 3 shows the gasoline-fueled LDV emissions inventory in the four urban areas in the 2022 LDV emissions scenarios. Wintertime LDV NOx emissions are highest in Atlanta in all scenarios. Wintertime VOC and primary $PM_{2.5}$ emissions are highest in Detroit due, in large part, to the effect of colder weather on cold starts. In contrast, in summer, Atlanta has the highest LDV emissions of NOx, VOC and $PM_{2.5}$, due to a combination of higher ambient temperatures and higher VMT. LDV NOx emissions in all four areas decrease by more than 70% from Tier 1 to Tier 2 and then only by 4% from Tier 2 to LEV III. Similarly, VOC emissions decrease by ~60% or more from Tier 1 to Tier 2 and then by 6–9% in the transition to LEV III by 2022.

3.2.2 Air Quality

Model simulation results for O_3 are presented in Fig. 6 for the summer month (July), the time period of concern for O_3 in the eastern US. The incremental benefits of the LDV standards are examined using the spatial distribution of the monthly mean of daily maximum 8-hr O_3 concentrations and differences in these monthly means between pairs of 2022 LDV scenarios. The same quantities are listed in Table 4 for the four urban areas. Also shown in this table are the monthly maximum 8-hr O_3 concentrations in each area. All values tabulated for an urban area are those modeled in the CAMx 12 km resolution grid cell in the geographic center of each area reflecting the approximate impact on the local population.

If LDV emissions standards were no more stringent than the Tier 1 standard in 2022, the monthly mean of daily maximum 8-hr O_3 could be as high as 88 ppb in the portion of the eastern US within the CAMx 12 km domain with values exceeding 60 ppb in most of the eastern US and parts of Georgia and the New York/New Jersey/D.C. corridor experiencing more than 80 ppb. Among the four urban areas analyzed here, the monthly mean of daily maximum 8-hr O_3 ranges from 57 ppb at Detroit to 78 ppb at Philadelphia and the highest 8-hr O_3 predicted in the month ranges from 83 ppb in Detroit to 111 ppb in Atlanta.

Strengthening the standard from Tier 1 to Tier 2 results in a reduction of over 6 ppb in the monthly mean of daily 8-hr maxima in large parts of the eastern US and up to 10 ppb in Georgia (see Fig. 6). When considering only the four areas, Tier 2 ozone benefits are strongest in Atlanta with the monthly mean of the daily 8-hr O_3 maxima decreasing by 9 ppb (11%) from 2008 to 2022 and the monthly highest O_3 decreasing by 16 ppb (14%) from 2008 to 2022. The NOx-limited environment present in Atlanta coupled with the enhanced formation of ozone from precursor NOx at warmer temperatures implies that reductions in summertime LDV emissions of this precursor due to the more stringent Tier 2 standard result in a prominent ozone benefit. When compared to Atlanta and Philadelphia, Detroit shows a small benefit (3–4 ppb) for the monthly mean of daily maximum 8-hr O_3 and the monthly highest 8-hr O_3. St. Louis shows a very small reduction (2 ppb) in the monthly highest 8-hr O_3 but a higher reduction of 5 ppb in the monthly mean suggesting that the highest 8-hr concentration here in the 2022 Tier 1 scenario (94 ppb) is mostly due to sources other than on-road vehicles. There are some areas on the western shore of Lake Michigan (Milwaukee and Chicago) that experience a slight increase (3 ppb) in the monthly mean of daily maximum 8-hr O_3 from the Tier 1 to Tier 2 scenarios. This is likely due to a VOC-limited environment in these urban areas in the Tier 1 scenario; NOx that was otherwise titrating ozone becomes unavailable due to the Tier 2 LDV emissions reductions.

The monthly mean of daily maximum 8-hr O_3 in the summer month shows up to a 0.2 ppb (~0.2%) reduction in the eastern US domain in 2022 if we switch from the Tier 2 to LEV III programs (see Fig. 6). When considering the four urban areas, the predicted reduction in the monthly mean value is ~0.1 ppb and the monthly highest 8-hr O_3 is reduced by 0.1–0.3 ppb (0.1–0.3%) (see Table 4). The model results suggest that there is a very small additional benefit in 2022 in strengthening the LDV standard from Tier 2 to one similar to the draft proposed California LEV III standard. These small benefits are consistent with the small reductions in ozone (<1.5%) modeled by Collet et al. (2012) for the transition from LEV II to a standard similar to LEV III in the California South Coast Basin. We note that the LEV III standard for NOx + non-methane organic gases will not be fully phased in until 2025. Thus, results shown represent the air quality benefits achievable by 2022. We expect some slight additional improvements in ozone from 2022 to 2025 with the planned complete phase-in of the LEV III standard.

Eliminating LDV emissions (in the zero-out LDV scenario) results in 2–4 ppb (3–5%) reductions in the monthly mean of summertime daily 8-hr maximum ozone and 3–7 ppb (3–8%) in the highest 8-hr ozone below 2022 Tier 2 levels in the four urban areas. The maximum reduction in the monthly mean of daily 8-hr maximum ozone in the eastern US domain is 4 ppb (~6%). The predicted reductions in ozone achieved with the complete zero-out of LDV emissions from the 2022 levels with the current (i.e., Tier 2) standard are generally less than the reductions achieved in moving from the Tier 1 to Tier 2 standards.

Model simulation results for $PM_{2.5}$ mass are shown in Fig. 7 for February and in Fig. 8 for July. We present the spatial distribution of the monthly mean $PM_{2.5}$ concentrations and differences in these monthly means between 2022 LDV scenarios. Table 4 shows similar information for monthly mean $PM_{2.5}$ and monthly maximum 24-hr $PM_{2.5}$ in the four urban areas. Table 5 shows the monthly mean concentrations of key $PM_{2.5}$ components in the four areas and differences between the scenarios.

Wintertime monthly mean concentrations of $PM_{2.5}$ in the 2022 Tier 1 scenario exceed 15 $\mu g/m^3$ (the annual mean standard for $PM_{2.5}$) in large parts of Georgia, the Carolinas, the Northeast and the Upper Midwest (see Fig. 7). Similar spatial patterns are seen in the 2022 Tier 2 scenario but the elevated concentrations are less widespread. Monthly mean $PM_{2.5}$ decreases by more than 1 $\mu g/m^3$ from Tier 1 to Tier 2 levels in broad swaths across the eastern US and by 2 $\mu g/m^3$ (~10%) in large urban areas such as Chicago, D.C., Detroit, Philadelphia, Raleigh, New York and major highway corridors such as Interstate-95. Among the four urban areas analyzed here, the Tier 1 wintertime mean $PM_{2.5}$ concentration ranges from 14 $\mu g/m^3$ in Atlanta to 19 $\mu g/m^3$ in Philadelphia and the maximum 24-hr $PM_{2.5}$ ranges from 22 $\mu g/m^3$ in Atlanta to 48 $\mu g/m^3$ in Philadelphia (see Table 4). Wintertime Tier 2 $PM_{2.5}$ benefits are strongest in Philadelphia with the mean $PM_{2.5}$ reduced by 1.9 $\mu g/m^3$ (10%) from Tier 1 levels and maximum 24-hr $PM_{2.5}$ reduced by 4.5 $\mu g/m^3$ (9%). The reductions in $PM_{2.5}$ due to Tier 2 are driven by reductions in nitrate in all four urban areas (see Table 5). Because nitrate constitutes a very small fraction of primary PM emissions, the reduction in nitrate has to be due to the large reduction in LDV NOx emissions (see Table 3), which impacts secondary nitrate formation. This is also consistent with relatively high reductions predicted in PM ammonium (compared to the other PM components) which would have otherwise been associated with PM nitrate.

Reductions in $PM_{2.5}$ concentrations between Tier 1 and Tier 2 scenarios are generally lower in summer (Fig. 8) than winter with the mean $PM_{2.5}$ in Philadelphia reduced by 0.9 $\mu g/m^3$ (6%) from Tier 1 levels and maximum 24-hr $PM_{2.5}$ reduced by 1.5 $\mu g/m^3$ (6%). The Tier 2 $PM_{2.5}$ benefits in summer are lower primarily due to less formation of PM nitrate from NOx emissions in summer due to enhanced volatilization from the particulate phase. Also, larger reductions in PM sulfate are predicted in summer (0.1 – 0.2 $\mu g/m^3$ reduction in monthly mean) than winter (Table 5). The reductions in PM sulfate from Tier 1 levels are due to Tier 2 mandated reductions in gasoline sulfur to 30 ppm.

Switching from the Tier 2 to LEV III results in less than 0.1 $\mu g/m^3$ reduction in monthly mean $PM_{2.5}$ in the eastern US domain in 2022 in both summer and winter and up to 0.14 $\mu g/m^3$ (0.5%) reduction in monthly maximum 24-hr $PM_{2.5}$ in the four urban areas (see Table 4). These small changes suggest that little additional $PM_{2.5}$ benefit is obtained by strengthening the LDV standard from Tier 2 to a LEV III standard. This is consistent with the relatively small change in $PM_{2.5}$ precursor emissions between the Tier 2 and LEV III scenarios and the fact that Tier 2 LDV emissions of $PM_{2.5}$ precursors constitute a relatively small fraction (0.2-10%) of the total inventory (see Table 1). Because the PM component of the draft LEV III standard will not be fully phased in until 2028, some additional improvements in PM are expected from 2022 to 2028.

Modeling results suggest that elimination of gasoline-fueled LDVs in the four urban areas would result in 0.3–1.5 $\mu g/m^3$ (3–11%) reductions in the monthly mean $PM_{2.5}$ and 0.3–2.9 $\mu g/m^3$ (2–7%) in the monthly maximum 24-hr $PM_{2.5}$ below 2022 Tier 2 levels. The maximum reduction in the monthly mean $PM_{2.5}$ in the eastern US domain is 1.8 $\mu g/m^3$ (~8%). The predicted reductions in total $PM_{2.5}$ mass due to the complete removal of gasoline-fueled LDV emissions from 2022 Tier 2 levels are generally less than the reductions achieved in progressing from the Tier 1 to Tier 2 standards.

3.3. Summary

For the four urban areas considered here, the largest Tier 2 ozone benefit (compared to Tier 1 levels) is seen in Atlanta and the largest $PM_{2.5}$ benefit in Philadelphia. In both cases, reductions in NOx emissions have the largest contribution to ozone and $PM_{2.5}$ reductions, the former due to decreased ozone formation with NOx reductions in NOx-limited environments such as in Atlanta and the latter due to reduced secondary PM nitrate formation such as in Philadelphia.

Overall, the modeling results suggest that large improvements in ambient ozone and $PM_{2.5}$ concentrations resulted from the switch from Tier 1 to Tier 2 standards. However, very small additional reductions in 2022 ozone and $PM_{2.5}$ levels are predicted to result from the transition to a Federal standard similar to the draft proposed California LEV III standard. These results are consistent with the relatively small change in emissions between the Tier 2 and LEV III scenarios compared to the change between Tier 1 and Tier 2 scenarios and the fact that Tier 2 LDV emissions of ozone and $PM_{2.5}$ precursors constitute a relatively small fraction of the total inventory. Predicted improvements in ozone and $PM_{2.5}$ due to the complete elimination of gasoline-fueled LDV emissions are generally smaller than the improvements due to the transition from Tier 1 to Tier 2 standards.

The main limitation of this study is introduced by the lack of complete phase-in of the LEV III standard by 2022, the basis year for comparing emission standards. Some additional improvements in ozone from 2023 to 2025 and in PM from 2023 to 2028 are expected as the LEV III standard fully matures. Other sources of uncertainty include use of the 2020 NEI as a surrogate for 2022 anthropogenic area and point emissions, differences between the 2005 base year (which was used to derive the 2020 inventory) and the 2008 base year and assumed growth and control factors. There are also limitations in the data used to develop VOC speciation profiles. The benefits of the vehicle emissions standards have been determined using 2008 meteorology and global background concentrations. Other meteorological and background conditions might yield somewhat different results. We have focused on specific past, present and potential future Federal standards applied to the eastern US. Future work should examine whether similar results are obtained for urban areas in other parts of the country and consider additional vehicle standards.

Acknowledgements

This work was supported by the Coordinating Research Council Atmospheric Impacts Committee. We thank Robert Gilliam, EPA for data on 2008 WRF meteorology and Alison Eyth, EPA for NEI data.

Supplementary Data

Additional emissions data and model performance evaluation can be found in the online version, at doi: xxx.

References

Baek B.H., DenBleyker, A., 2010. User's Guide for the SMOKE-MOVES Integration Tool. Prepared for the U.S. EPA OAQPS. July. http://www.smoke-model.org/smoke_moves_tool/SMOKE-MOVES_Tool_Users_Guide.pdf, accessed April 2011.

CASTNET, 2011. Clean Air Status and Trends Network. http://epa.gov/castnet/, accessed August 2011.

Collet, S., Kidokoro, T., Sonoda, Y., Lohman, K., Karamchandani, P., Chen, S.Y., Minoura, H., 2012. Air quality impacts of motor vehicle emissions in the south coast air basin: Current versus more stringent control scenario. Atmos. Environ. 47, 236-240.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., Kloster, S., 2010. Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). Geosci. Model Dev., 3, 43-67.

ENVIRON, 2009. User's Guide Emissions Processor, version 3. August. ftp://amdaftp.tceq.texas.gov/pub/HGB8H2/ei/EPS3_manual/EPS3UG_UserGuide_200908.pdf, accessed August 2011.

ENVIRON, 2011. User's Guide, Comprehensive Air Quality Model with Extensions (CAMx), Version 5.40. http://www.camx.com, accessed September 2011.

EPA, 1999. Technical Support Document for the Tier 2/Gasoline Sulfur Ozone Modeling Analyses. Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency. EPA420-R-99-031. December. http://ww.epa.gov/tier2/frm/ria/r99023.pdf, accessed August 2011.

EPA, 2002. User Guide: Air Quality System. Report prepared by the U.S. Environmental Protection Agency, Research Triangle Park, North Carolina. April. http://www.epa.gov/ttn/airs/aqs/softw/AQSUserGuide_v1.pdf, accessed August 2011.

EPA, 2008a. Regulatory Impact Analysis: Control of Emissions of Air Pollution from Locomotive Engines and Marine Compression Ignition Engines Less than 30 Liters Per Cylinder. Assessment and Standards Division, Office of Transportation and Air Quality, US Environmental Protection Agency. EPA420-R-08-001. March. http://www.epa.gov/otaq/regs/nonroad/420r08001a.pdf, accessed July 2011.

EPA, 2008b. Analysis of Particulate Matter Emissions from Light-Duty Gasoline Vehicles in Kansas City. Office of Transportation and Air Quality and Office of Research and Development, U. S. Environmental Protection Agency. Research Triangle Park, North Carolina. EPA420-R-08-

010. April. http://www.epa.gov/otaq/emission-factors-research/420r08010.pdf, accessed November 2011.

EPA, 2009. Regulatory Impact Analysis: Control of Emissions of Air Pollution from Category 3 Marine Diesel Emissions. Assessment and Standards Division, Office of Transportation and Air Quality, US Environmental Protection Agency. EPA-420-R-09-019. http://www.epa.gov/nonroad/marine/ci/420r09019.pdf, accessed July 2011.

EPA, 2010a. Technical Guidance on the Use of MOVES2010 for Emission Inventory Preparation in State Implementation Plans and Transportation Conformity. EPA-420-B-10-023. Office of Transportation and Air Quality, U.S. Environmental Protection Agency. April. http://www.epa.gov/otaq/models/moves/420b10023.pdf, accessed April 2011.

EPA, 2010b. Quantitative Health Risk Assessment for Particulate Matter. Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency. EPA-452/R-10-005, June.

EPA, 2010c. Purpose of OAQPS 2020 future-year inventories, from 2005-based v4 modeling platform. Prepared by Marc Houyoux, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency. April 15. ftp://ftp.epa.gov/EmisInventory/2005v4/2020_emissions_purpose_memo_final.doc, accessed March 2011.

EPA, 2011a. 2008 National Emissions Inventory: Data and Documentation. Clearinghouse for Inventories and Emissions Factors. http://www.epa.gov/ttnchie1/net/2008inventory.html, accessed July 2011.

EPA, 2011b. SPECIATE 4.3: Addendum to SPECIATE 4.2. Speciation Database Development Documentation. Prepared for the U.S. EPA Office of Research and Development. Prepared by TransSystems| E.H. Pechan & Associates. EPA/600/R-11/121. September. http://www.epa.gov/ttn/chief/software/speciate/speciate4/addendum4.2.pdf, accessed September 2011.

EPA, 2011c. 2005 National Emissions Inventory: Data and Documentation. Clearinghouse for Emissions Inventories. http://www.epa.gov/ttnchie1/net/2005inventory.html, accessed August 2011.

FAA, 2010. Federal Aviation Administration, Terminal Area Forecast (TAF). http://aspm.faa.gov/main/taf.asp, accessed July 2011.

Guenther, A., Karl, T., Harley, P.,Wiedinmyer, C., Palmer, Geron, C., 2006. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). Atmos. Chem Phys. 6, 3181-3210.

IMPROVE, 1995. IMPROVE Data Guide. University of California Davis, August, 1995. http://vista.cira.colostate.edu/improve/Publications/OtherDocs/IMPROVEDataGuide/IMPROVEDataGuide.htm, accessed August 2011.

Koffi, B., Szopa, S., Cozic, A., Hauglustaine, D., van Velthoven, P., 2010. Present and future impact of aircraft, road traffic and shipping emissions on global tropospheric ozone. Atmos. Chem. Phys. 10, 11681–11705.

Kryak, D., Black, K., Cook, R., Constantini, M., Baldauf, R., Vette, A., Costa, D., 2010. A source-to-outcome approach to address near-road air pollution. EM, Air & Waste Management Association, November.

Matthes, S., Grewe, V., Sausen, R., Roelofs, G.J., 2007. Global impact of road traffic emissions on tropospheric ozone. Atmos. Chem. Phys. 7, 1707–1718.

Morris, R.E., Koo, B., Wang, B., Stella, G., McNalley, D., Loomis, C., Chien, C., Tonnesen, G., 2007. Technical Support Document for VISTAS Emissions and Air Quality Modeling to Support Regional Haze State Implementation Plan (Draft). Prepared for the VISTAS Technical Analysis Committee. http://www.vistas-sesarm.org/documents/ENVIRON_Air_Quality_Modeling_Technical_Support_Document_11-14-07.pdf, accessed July 2011.

Morris, R.E., Koo, B., Sakulyanontvittaya, T., Stella, G., McNally, D., Loomis, C., Tesche, T.W., 2008. Technical Support Document for Association for Southeastern Integrated Planning (ASIP) Emissions and Air Quality Modeling to Support $PM_{2.5}$ and 8-Hour Ozone State Implementation Plans. Prepared for the ASIP Technical Analysis Committee. http://www.gaepd.org/Files_PDF/plans/sip/Appendix_L.1_ASIP_TSD_PM25-O3_FinalRept_3.24.09.pdf, accessed July 2011.

NCAR. 2011. The Tropospheric Visible and Ultraviolet (TUV) Radiation Model web page. National Center for Atmospheric Research, Atmospheric Chemistry Division, Boulder, Colorado. http://cprm.acd.ucar.edu/Models/TUV/index.shtml, accessed May 2011.

Nopmongcol, U., Griffin, W. M., Yarwood, G., Dunker, A. M., Maclean, H. L., Mansell, G., Grant, J., 2011. Impact of Dedicated E85 Vehicle Use on Ozone and Particulate Matter in the U.S. Atmos. Environ. 45, 7330-7340.

Pinder, R. W., Strader, R., Davidson, C. I., Adams, P. J., 2004. A temporally and spatially resolved ammonia emission inventory for dairy cows in the United States. Atmos. Environ. 38, 3747-3756.

Roustan, Y., Pausader, M., Seigneur, C., 2011. Estimating the effect of on-road vehicle emission controls on future air quality in Paris, France. Atmos. Environ. 45, 6828-6836.

Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Duda, M.G., Huang, X-Y., Wang, W., Powers, J.G., 2008. A Description of the Advanced Research WRF Version 3. NCAR Technical Note, NCAR/TN-45+STR (June 2008). http://www.mmm.ucar.edu/wrf/users/, accessed July 2011.

Zhang, L., Brook, J.R., Vet, R., 2003. A revised parameterization for gaseous dry deposition in air quality models. Atmos. Chem. Phys. 3, 2067–2082.

Table 1. Average-day emissions from gasoline-fueled LDVs in the continental US in model scenarios.

	Winter						Summer					
	NOx	**VOC**	**$PM_{2.5}$**	**CO**	**NH_3**	**SO_2**	**NOx**	**VOC**	**$PM_{2.5}$**	**CO**	**NH_3**	**SO_2**
2008												
LDV emissions (Mg/day)	8380	7039	242	92610	272	79	9661	6384	123	68064	337	99
% of all on-road	50%	86%	39%	90%	92%	72%	52%	83%	21%	89%	92%	71%
% of total anthropogenic	20%	21%	2%	54%	5%	0.3%	21%	18%	2%	46%	2%	0.4%
2022 Tier 1												
LDV emissions (Mg/day)	10422	6127	250	101288	457	48	12047	5319	131	73217	566	57
% of all on-road	77%	92%	73%	92%	94%	83%	80%	88%	55%	92%	94%	81%
% of total anthropogenic	31.0%	18.6%	2.9%	60%	8%	0.3%	32%	16%	2%	49%	4%	0.3%
2022 Tier 2												
LDV emissions (Mg/day)	2609	2269	188	69576	166	41	2897	2247	108	40745	206	48
% of all on-road	46%	81%	67%	89%	86%	80%	49%	75%	51%	87%	86%	79%
% of total anthropogenic	10.1%	7.8%	2.2%	51%	3%	0.2%	10%	8%	2%	35%	1%	0.2%
2022 LEV III												
LDV emissions (Mg/day)	2505	2134	173	65152	166	41	2781	2098	103	38120	206	48
% of all on-road	45%	80%	66%	89%	86%	80%	47%	74%	49%	86%	86%	79%
% of total anthropogenic	9.7%	7.4%	2.0%	49%	3%	0.2%	10%	7%	1%	33%	1%	0.2%

Table 2. Average-day emissions from gasoline-fueled LDVs in four urban areas in 2008.

Urban area	Pollutant	February 2008		July 2008	
		Emissions (Mg/day)	% of all on-road	Emissions (Mg/day)	% of all on-road
Atlanta	NOx	141.8	48%	157.1	51%
	VOC	103.2	85%	99.8	83%
	$PM_{2.5}$	3.9	34%	2.2	20%
Detroit	NOx	106.2	41%	107.2	41%
	VOC	102.6	84%	67.3	77%
	$PM_{2.5}$	5.0	40%	1.8	17%
Philadelphia	NOx	66.4	55%	73.0	57%
	VOC	53.1	81%	42.6	74%
	$PM_{2.5}$	2.5	48%	1.1	27%
St. Louis	NOx	155.8	50%	171.4	52%
	VOC	120.3	83%	99.8	77%
	$PM_{2.5}$	5.4	42%	2.4	22%

Table 3. Emissions from gasoline-fueled LDVs in four urban areas in 2022 LDV emissions scenarios[a].

Month	Area	Pollutant	LDV Tier 1 Emissions (Mg/day)	LDV Tier 2 Emissions (Mg/day)	LDV LEV III Emissions (Mg/day)	%Change from Tier 1 to Tier 2	%Change from Tier 2 to LEV III
February	Atlanta	NOx	161	39	37	-76%	-4%
		VOC	92	31	29	-66%	-6%
		$PM_{2.5}$	3.7	.8	2.6	-24%	-8%
	Detroit	NOx	152	33	32	-78%	-4%
		VOC	103	33	31	-68%	-6%
		$PM_{2.5}$	5.2	3.8	3.1	-27%	-18%
	Philadelphia	NOx	83	17	16	-80%	-4%
		VOC	51	18	17	-65%	-7%
		$PM_{2.5}$	2.6	1.9	1.8	-26%	-7%
	St. Louis	NOx	105	28	27	-73%	-4%
		VOC	67	25	23	-63%	-6%
		$PM_{2.5}$	3.0	2.2	2.0	-25%	-8%
July	Atlanta	NOx	182	42	41	-77%	-4%
		VOC	81	30	29	-63%	-6%
		$PM_{2.5}$	2.2	1.9	1.8	-17%	-5%
	Detroit	NOx	156	31	30	-80%	-4%
		VOC	66	26	23	-61%	-9%
		$PM_{2.5}$	1.9	1.6	1.5	-15%	-10%
	Philadelphia	NOx	91	17	16	-82%	-4%
		VOC	38	15	14	-60%	-7%
		$PM_{2.5}$	1.2	1.0	1.0	-17%	-4%
	St. Louis	NOx	117	30	29	-74%	-4%
		VOC	53	22	21	-58%	-7%
		$PM_{2.5}$	1.3	1.1	1.1	-17%	-5%

[a] LDV emissions are all zero in LDV zero-out scenario.

Table 4. Concentrations of O_3 and $PM_{2.5}$ in four urban areas in the 2022 LDV emissions scenarios.

Urban area	Month	Pollutant	Form of concentration reported	2022 Tier 1	2022 Tier 2	Change (%Change) from Tier 1 to Tier 2		Change (%Change) from Tier 2 to LEV-III		Change (%Change) from Tier 2 to zero-out		Change (%Change) from LEV III to zero-out	
Atlanta	**Feb.**	$PM_{2.5}$	Monthly average (μg/m^3)	14.2	12.9	-1.3	-9%	-0.04	-0.3%	-0.8	-7%	-0.8	-6%
			Highest 24-hr average (μg/m^3)	21.5	19.3	-2.2	-10%	-0.05	-0.3%	-1.7	-9%	-1.6	-8%
	July	Ozone	Monthly average of daily 8-hr max (ppb)	76.7	68.0	-8.7	-11%	-0.13	-0.2%	-3.6	-5%	-3.5	-5%
			Highest 8-hr average (ppb)	111.4	95.4	-16.0	-14%	-0.27	-0.3%	-7.4	-8%	-7.1	-7%
		$PM_{2.5}$	Monthly average (μg/m^3)	13.9	13.3	-0.6	-4%	-0.02	-0.1%	-0.3	-3%	-0.3	-2%
			Highest 24-hr average (μg/m^3)	21.0	19.6	-1.4	-6%	-0.02	-0.1%	-0.5	-2%	-0.4	-2%
Detroit	**Feb.**	$PM_{2.5}$	Monthly average (μg/m^3)	16.1	14.3	-1.8	-11%	-0.11	-0.7%	-1.5	-11%	-1.4	-10%
			Highest 24-hr average (μg/m^3)	28.4	25.6	-2.8	-10%	-0.14	-0.5%	-2.1	-8%	-1.9	-8%
	July	Ozone	Monthly average of daily 8-hr max (ppb)	57.4	54.3	-3.1	-5%	-0.06	-0.1%	-1.5	-3%	-1.4	-3%
			Highest 8-hr average (ppb)	83.0	79.2	-3.8	-5%	-0.14	-0.2%	-2.9	-4%	-2.7	-3%
		$PM_{2.5}$	Monthly average (μg/m^3)	10.9	10.4	-0.5	-4%	-0.02	-0.2%	-0.3	-3%	-0.3	-2%
			Highest 24-hr average (μg/m^3)	23.4	22.1	-1.4	-6%	-0.03	-0.1%	-0.6	-3%	-0.6	-3%
Philadelphia	**Feb.**	$PM_{2.5}$	Monthly average (μg/m^3)	19.4	17.5	-1.9	-10%	-0.05	-0.3%	-1.3	-8%	-1.3	-7%
			Highest 24-hr average (μg/m^3)	48.1	43.6	-4.5	-9%	-0.09	-0.2%	-2.9	-7%	-2.8	-6%
	July	Ozone	Monthly average of daily 8-hr max (ppb)	77.5	70.8	-6.7	-9%	-0.10	-0.1%	-2.6	-4%	-2.5	-4%
			Highest 8-hr average (ppb)	107.8	99.6	-8.2	-8%	-0.15	-0.2%	-3.5	-4%	-3.4	-3%
		$PM_{2.5}$	Monthly average (μg/m^3)	13.9	13.0	-0.9	-6%	-0.02	-0.1%	-0.4	-3%	-0.4	-3%
			Highest 24-hr average (μg/m^3)	24.9	23.5	-1.5	-6%	-0.02	-0.1%	-0.6	-3%	-0.6	-3%
St. Louis	**Feb.**	$PM_{2.5}$	Monthly average (μg/m^3)	16.0	14.8	-1.3	-8%	-0.04	-0.3%	-0.9	-6%	-0.9	-6%
			Highest 24-hr average (μg/m^3)	34.2	31.8	-2.4	-7%	-0.06	-0.2%	-2.1	-7%	-2.1	-7%
	July	Ozone	Monthly average of daily 8-hr max (ppb)	70.2	65.3	-4.9	-7%	-0.09	-0.1%	-2.1	-3%	-2.1	-3%
			Highest 8-hr average (ppb)	93.7	91.9	-1.9	-2%	-0.12	-0.1%	-2.6	-3%	-2.4	-3%
		$PM_{2.5}$	Monthly average (μg/m^3)	12.2	11.8	-0.4	-3%	-0.01	-0.1%	-0.3	-2%	-0.3	-2%
			Highest 24-hr average (μg/m^3)	17.8	17.3	-0.5	-3%	-0.02	-0.1%	-0.3	-2%	-0.3	-2%

Table 5. Monthly average concentrations of key $PM_{2.5}$ components ($\mu g/m^3$) in four urban areas in the 2022 LDV emissions scenarios.

Urban area	Month	Pollutant	2022 Tier 1	Change from Tier 1 to Tier 2	Change from Tier 2 to LEV III	Change from Tier 2 to Zero-out
Atlanta	Feb.	$SO_4^=$	3.48	-0.03	0.00	0.00
		NO_3^-	2.30	-0.79	-0.01	-0.33
		NH_4^+	1.68	-0.29	0.00	-0.15
		EC	1.06	-0.02	-0.01	-0.06
		OC	5.19	-0.12	-0.03	-0.29
	July	$SO_4^=$	3.99	-0.14	0.00	-0.06
		NO_3^-	0.32	-0.18	0.00	-0.04
		NH_4^+	1.40	-0.11	0.00	-0.04
		EC	0.71	-0.01	0.00	-0.02
		OC	7.22	-0.13	-0.01	-0.18
Detroit	Feb.	$SO_4^=$	4.13	0.10	0.00	0.01
		NO_3^-	4.55	-1.26	-0.01	-0.69
		NH_4^+	2.54	-0.40	0.00	-0.26
		EC	0.86	-0.04	-0.02	-0.10
		OC	3.48	-0.19	-0.08	-0.43
	July	$SO_4^=$	4.63	-0.10	0.00	-0.04
		NO_3^-	0.57	-0.21	0.00	-0.06
		NH_4^+	1.76	-0.13	0.00	-0.06
		EC	0.64	0.00	0.00	-0.02
		OC	3.22	-0.04	-0.01	-0.09
Philadelphia	Feb.	$SO_4^=$	4.34	0.03	0.00	0.00
		NO_3^-	6.16	-1.34	-0.01	-0.68
		NH_4^+	3.09	-0.39	0.00	-0.21
		EC	0.95	-0.03	-0.01	-0.08
		OC	4.22	-0.15	-0.03	-0.36
	July	$SO_4^=$	4.92	-0.17	0.00	-0.06
		NO_3^-	1.03	-0.43	0.00	-0.12
		NH_4^+	1.89	-0.20	0.00	-0.06
		EC	0.81	-0.01	0.00	-0.02
		OC	4.81	-0.07	-0.01	-0.11
St. Louis	Feb.	$SO_4^=$	4.16	0.09	0.00	0.00
		NO_3^-	4.67	-0.91	-0.01	-0.41
		NH_4^+	2.88	-0.26	0.00	-0.14
		EC	0.72	-0.02	-0.01	-0.06
		OC	2.58	-0.12	-0.03	-0.29
	July	$SO_4^=$	4.22	-0.10	0.00	-0.04
		NO_3^-	0.36	-0.12	0.00	-0.04
		NH_4^+	1.51	-0.08	0.00	-0.04
		EC	0.74	-0.01	0.00	-0.02
		OC	5.17	-0.09	-0.01	-0.13

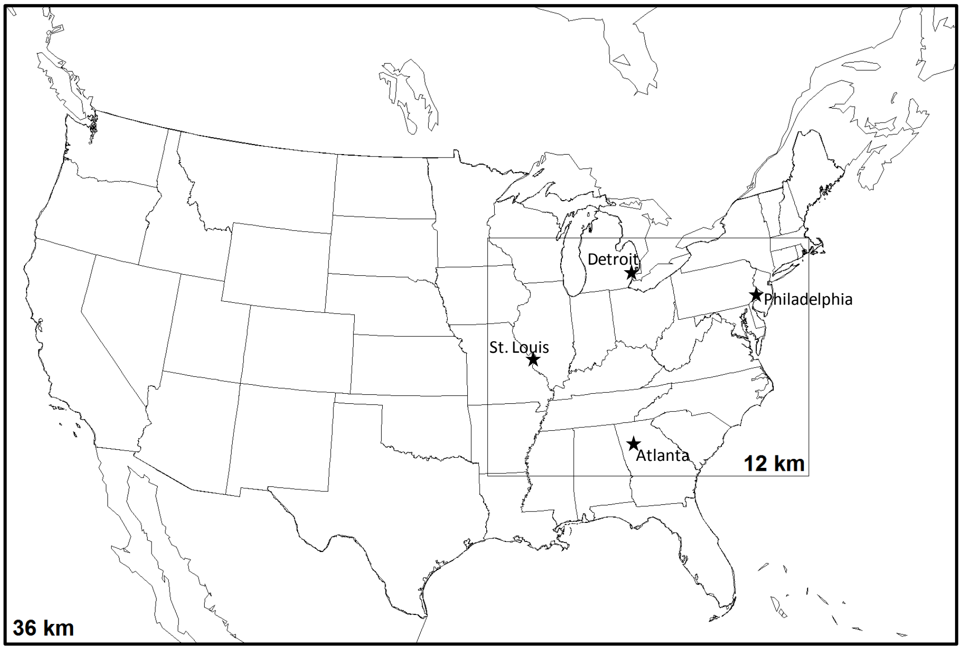


Fig. 1. Air quality modeling domain and urban areas analyzed.

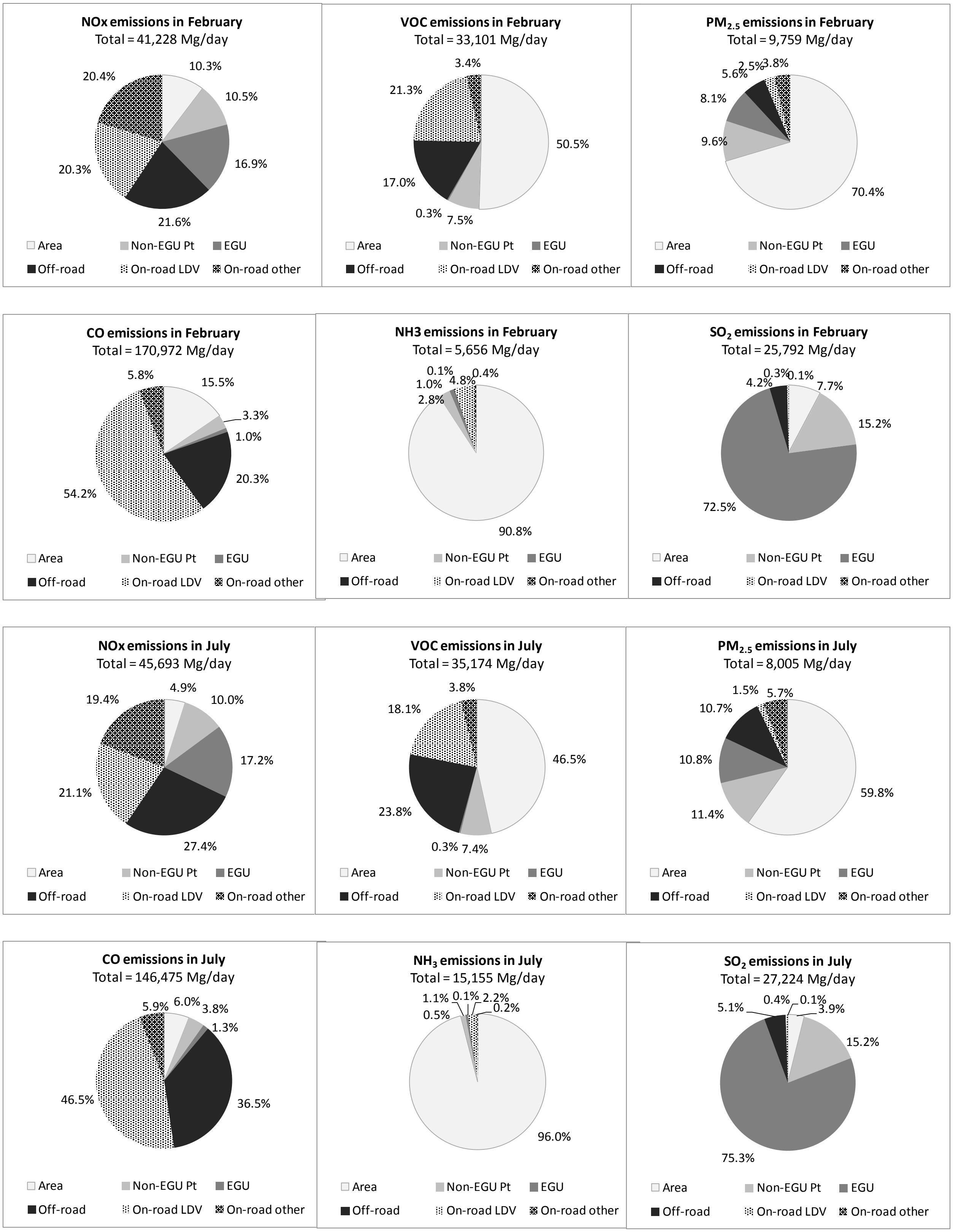


Fig. 2. Estimated anthropogenic emissions in the continental US in February and July, 2008.

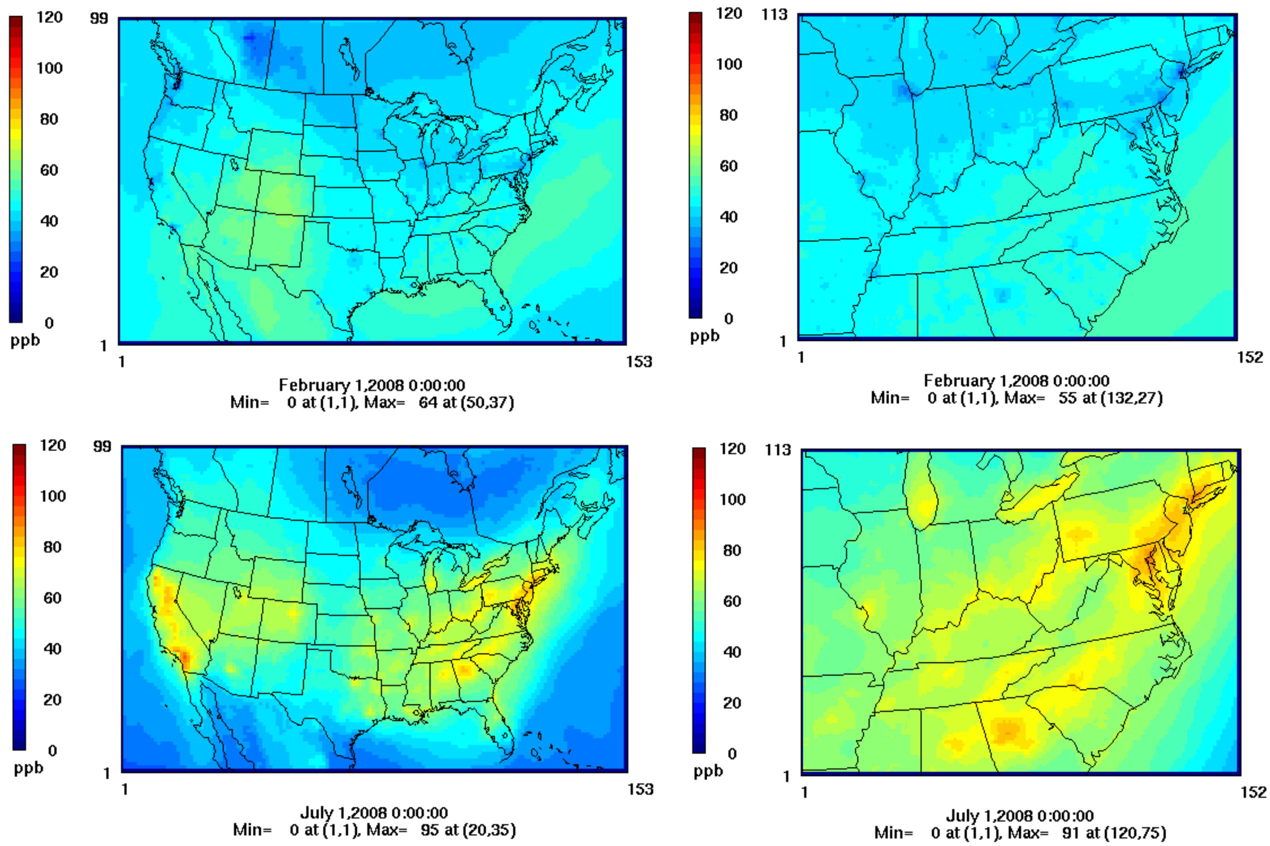


Fig. 3. Monthly mean of daily maximum 8-hr ozone concentrations in the 36 km domain (left) and 12 km domain (right) in February (top) and July (bottom), 2008.

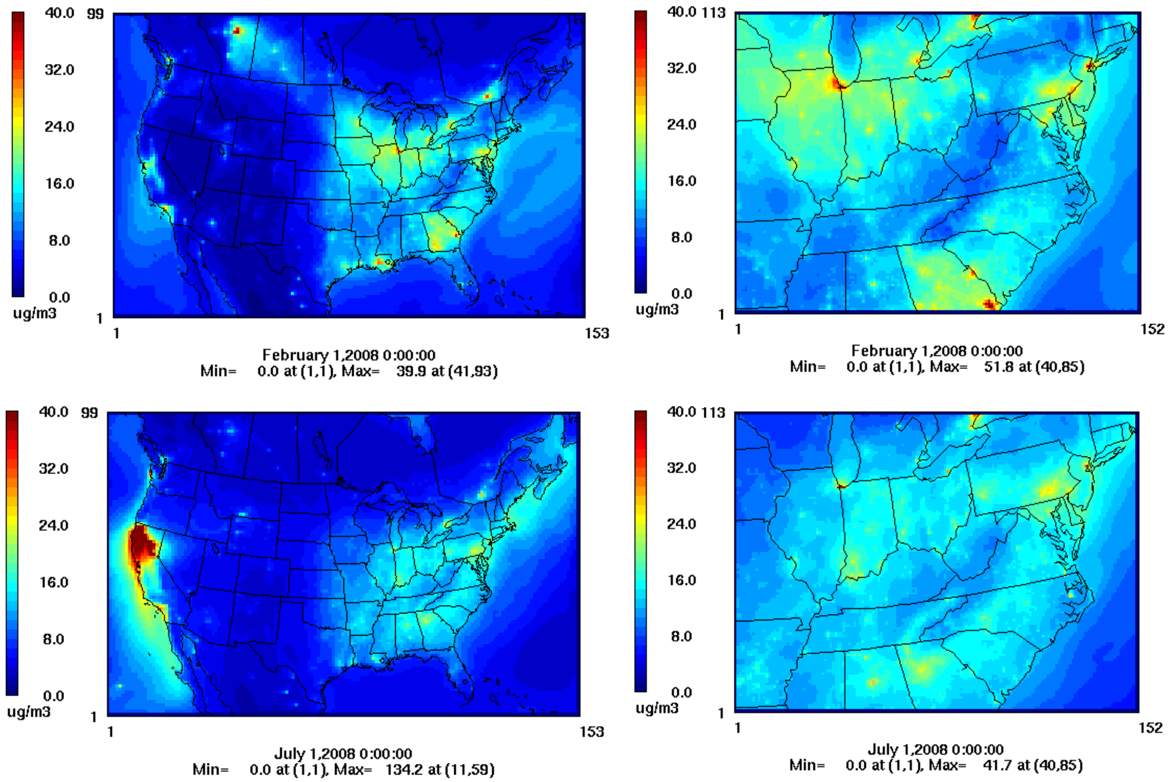


Fig. 4. Monthly mean concentrations of $PM_{2.5}$ in the 36 km domain (left) and 12 km domain (right) in February (top) and July (bottom), 2008.

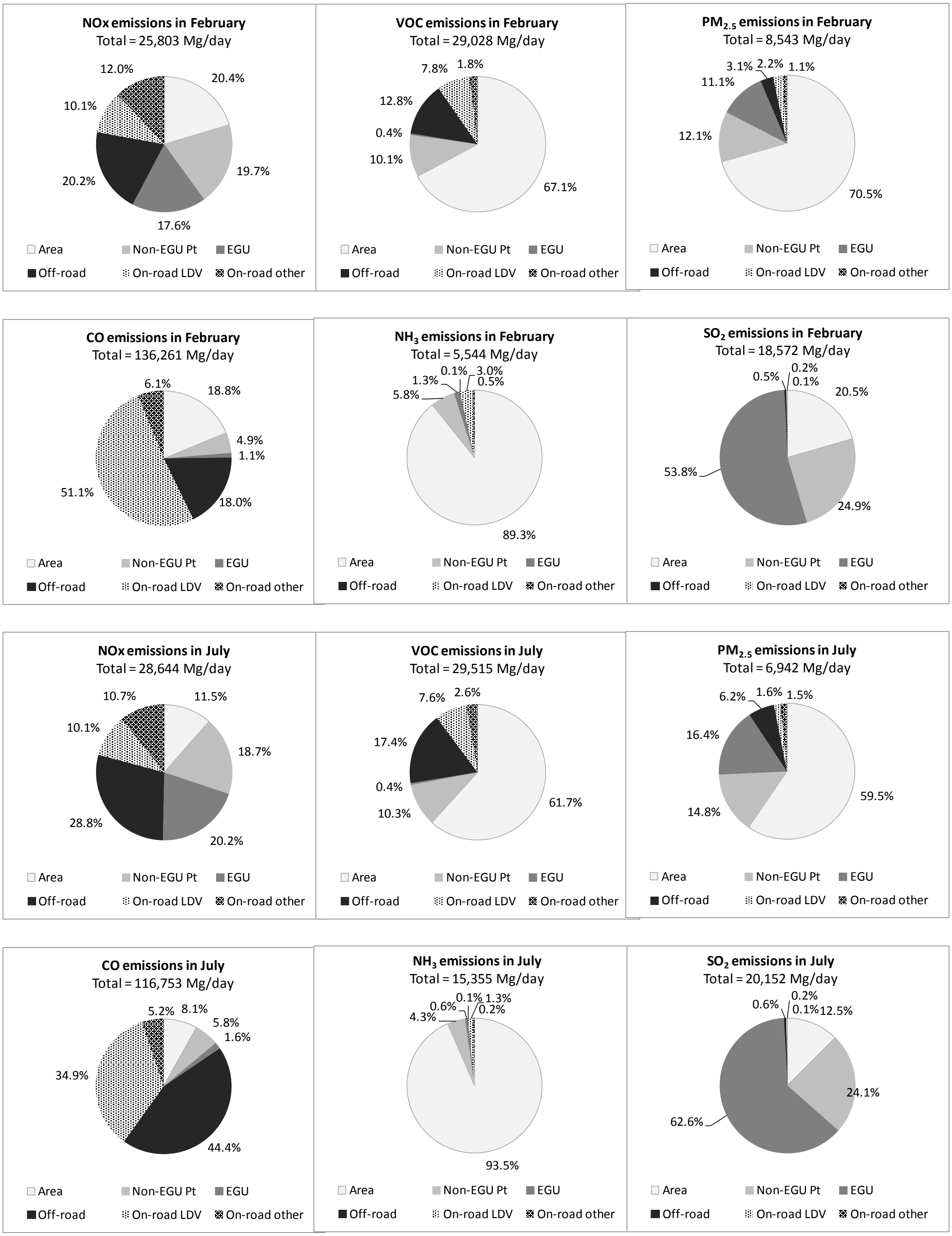


Fig. 5. Estimated anthropogenic emissions in the continental US in the 2022 Tier 2 scenario.

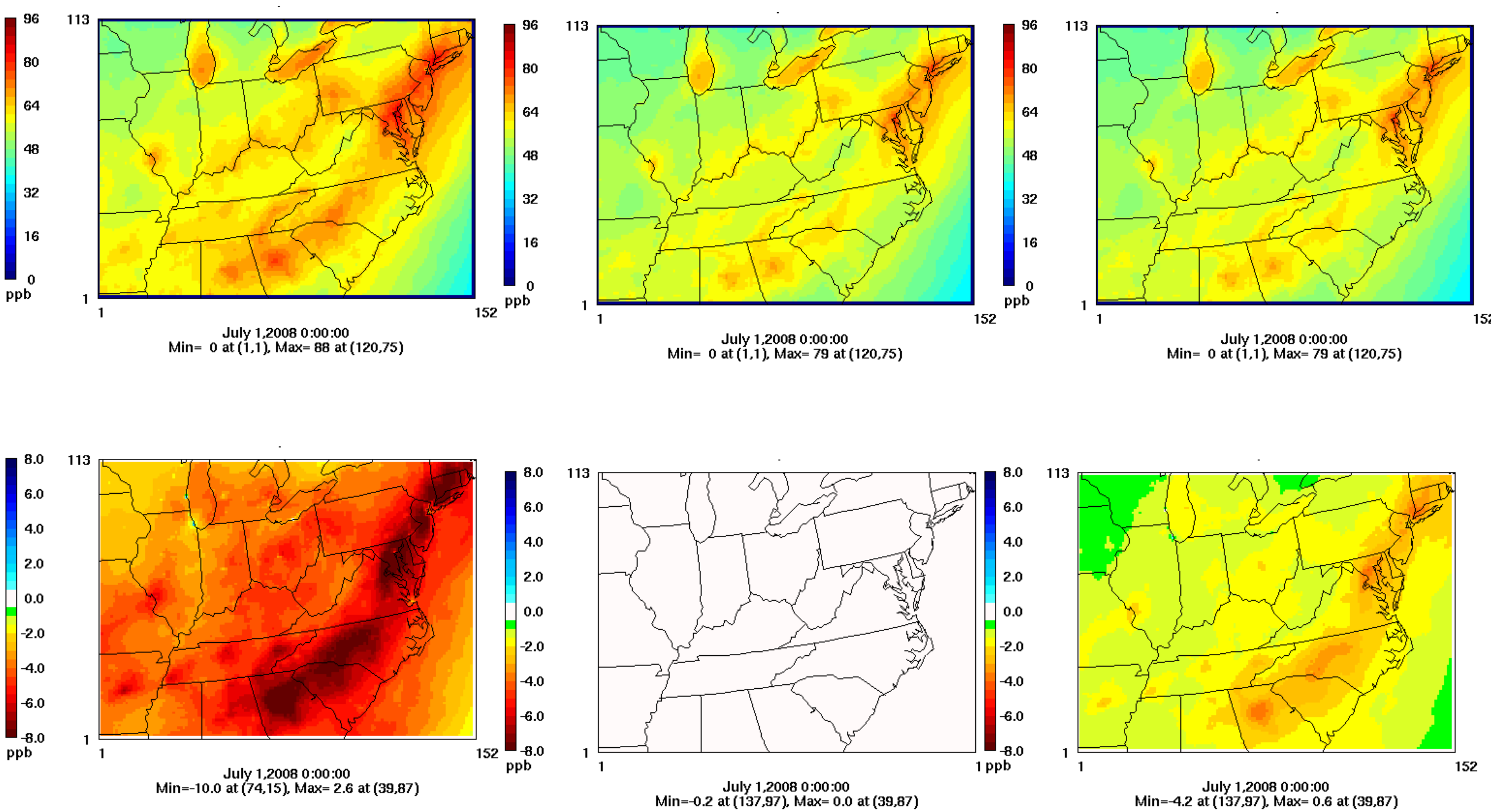


Fig. 6. Monthly mean and differences in monthly mean of daily maximum 8-hr ozone concentrations in July in 2022 scenarios: Tier1 (top left), Tier2 (top center), LEV III (top right), Tier 2 – Tier 1 (bottom left), LEV III – Tier 2 (bottom center), and LDV zero-out – Tier 2 (bottom right).

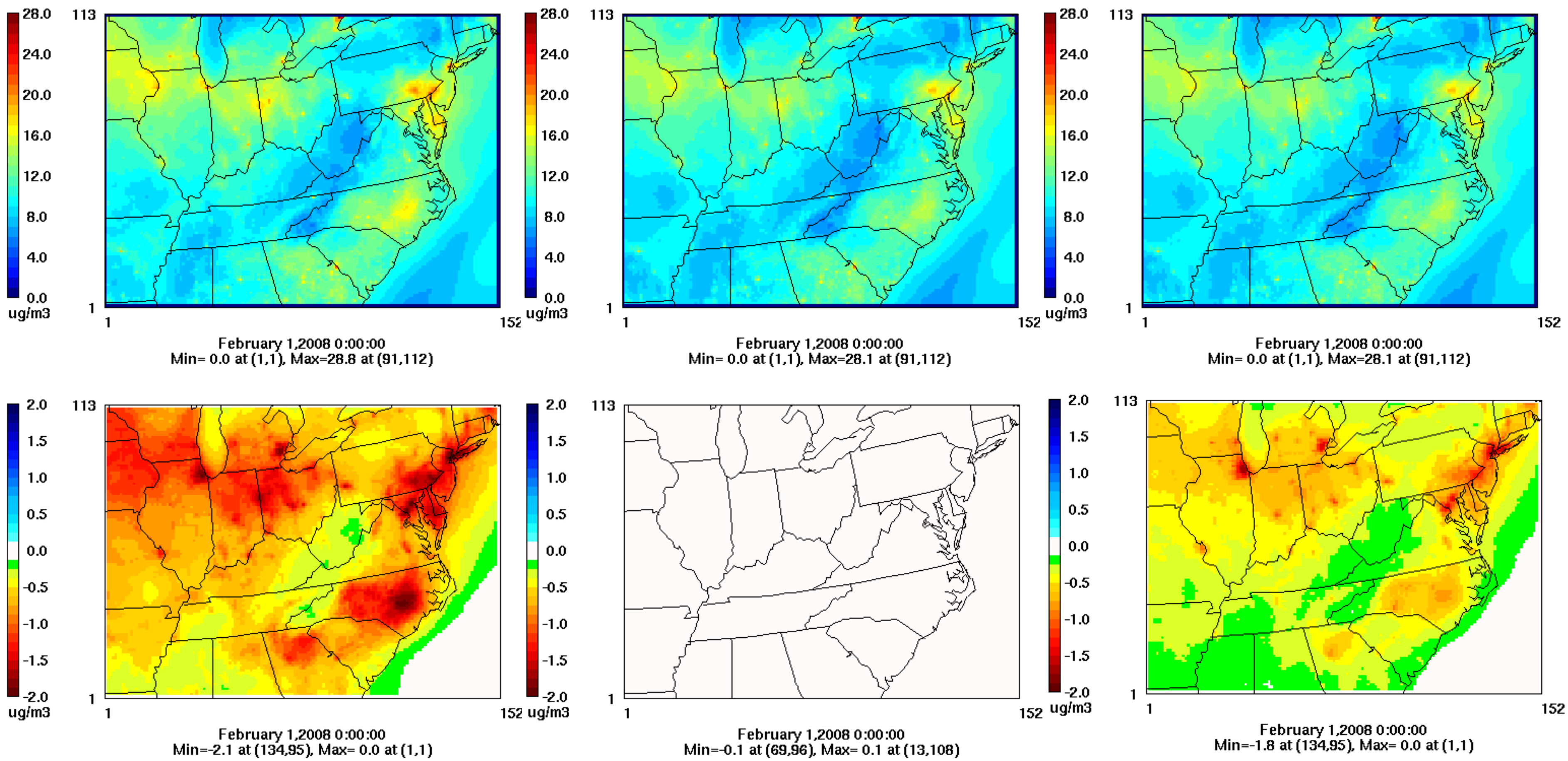


Fig. 7. Monthly mean and differences in monthly mean of hourly $PM_{2.5}$ concentrations in February in 2022 scenarios: Tier1 (top left), Tier2 (top center), LEV III (top right), Tier 2 – Tier 1 (bottom left), LEV III – Tier 2 (bottom center), and LDV zero-out – Tier 2 (bottom right).

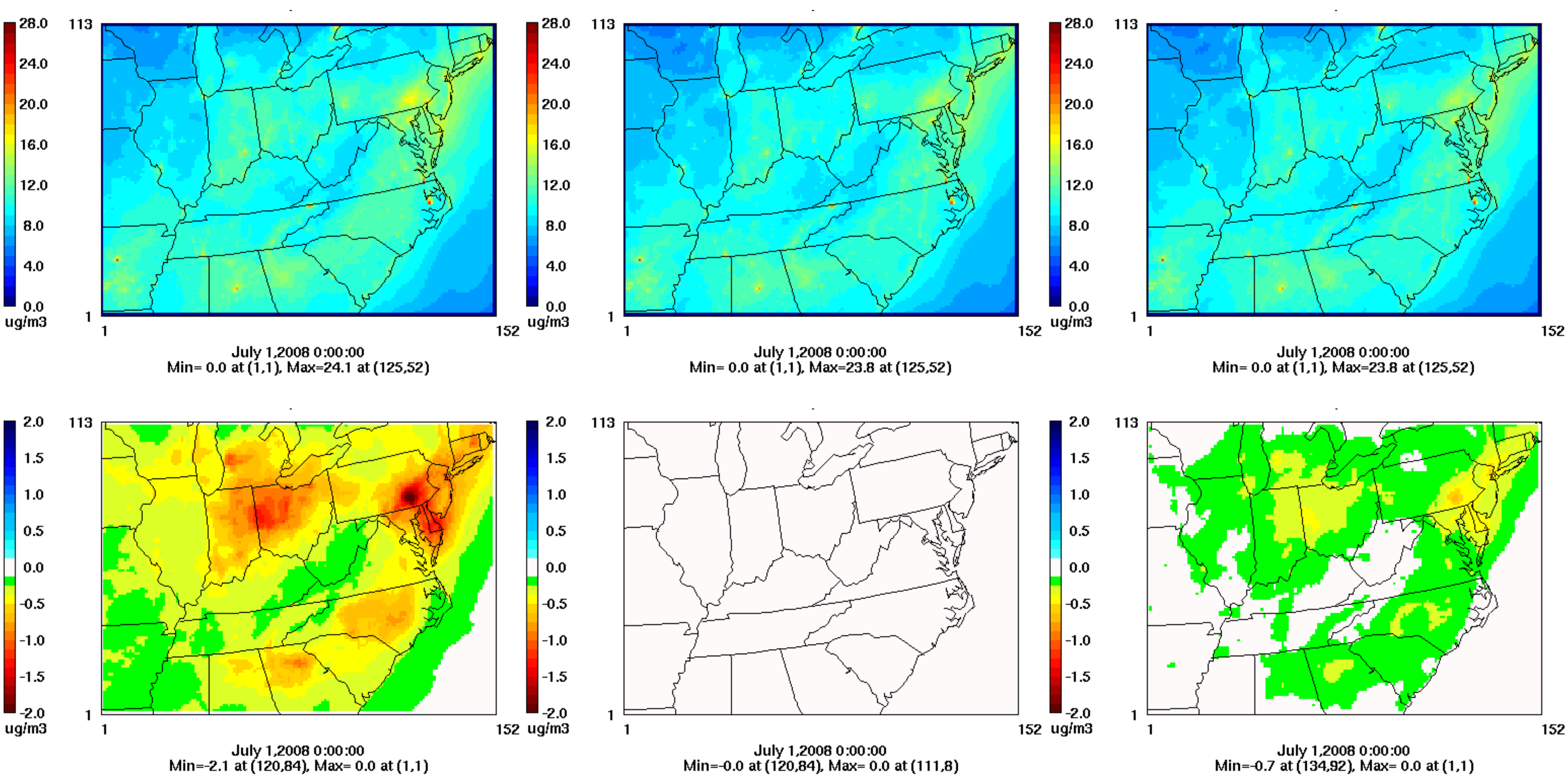


Fig. 8. Monthly mean and differences in monthly mean of hourly $PM_{2.5}$ concentrations in July in 2022 scenarios: Tier1 (top left), Tier2 (top center), LEV III (top right), Tier 2 – Tier 1 (bottom left), LEV III – Tier 2 (bottom center), and LDV zero-out – Tier 2 (bottom right).

SUPPLEMENTARY DATA

Effects of Light Duty Gasoline Vehicle Emission Standards in the United States on Ozone and Particulate Matter

Krish Vijayaraghavan, Chris Lindhjem, Allison DenBleyker, Uarporn Nopmongcol, John Grant, Edward Tai, Greg Yarwood

ENVIRON International Corporation, 773 San Marin Drive, Suite 2115, Novato, CA 94998, USA

S1. Emission standards

Table S1.1 Light duty gasoline vehicle exhaust emission standards[a,b] (g/mi at 10 years/100,000 miles[c])

Standard	Model Years	THC	NMHC	NMOG	CO	NOx	PM	NMOG+NOx
Tier 1	1994-2003	-	0.31	-	4.2	0.6	0.1	-
Tier 2	2004+[d]	-	-	0.0-0.09	0.0-4.2	0.07[e]	0.0-0.02	-
LEV III	2015	-	-	-	-	-	-	0.100
	2016	-	-	-	-	-	-	0.093
	2017	-	-	-	-	-	0.006	0.086
	2018	-	-	-	-	-	0.005	0.079
	2019	-	-	-	-	-	0.004	0.072
	2020	-	-	-	-	-	0.003	0.065
	2021	-	-	-	-	-	0.003	0.058
	2022	-	-	-	-	-	0.003	0.051
	2023	-	-	-	-	-	0.003	0.044
	2024	-	-	-	-	-	0.003	0.037
	2025	-	-	-	-	-	0.0025	0.030
	2026	-	-	-	-	-	0.002	-
	2027	-	-	-	-	-	0.0015	-
	2028	-	-	-	-	-	0.001	-

[a] Tier 1 and Tier 2 data source: www.epa.gov/greenvehicles/detailedchart.pdf, accessed June 2011
[b] LEV III data source: http://www.arb.ca.gov/msprog/levprog/leviii/meetings/071911/071911_lev_zev_eplabel_scoping.pdf, accessed July 2011
[c] Except LEV III for which 150,000 mile standard is shown
[d] 2004 to 2007 phase-in, ranges presented for all but NOx emission rates due to phase-in with emission standards that vary by vehicle definition bin
[e] Fleet wide standard

S2. On-Road Motor Vehicle Emissions Calculations

MOVES 2010a was run for calendar years 2008 and 2022 for vehicle ages 0-30 to develop on-road vehicle emissions for the 2008 base case and 2022 Tier 2 scenario. To model the 2022 Tier 1 and LEV III scenarios, multiple MOVES runs were made with adjustments were made to the fleet and model parameters as described below. A complete set of per-mile emission rates by pollutant, county, Source Classification Code (SCC), and emission process was developed for February and July 2008 and 2022. These emission rates were applied with 2008 and 2022 estimates of VMT in each month to develop by-county and SCC emissions of VOC, carbon monoxide (CO), NOx, PM_{10}, $PM_{2.5}$, sulfur dioxide (SO_2) and ammonia (NH_3).

MOVES was run for a subset of approximately 220 representative counties ("rep-counties") to manage MOVES' challenging computational requirements (a single MOVES run for one day over the 220 counties requires approximately 3 days on eight 2.6 GHz computers), yet still capture the important variations in emissions that occur geographically. The selection of rep-counties was made by grouping counties based on meteorology, fuels, and inspection and maintenance programs (I/M) that the counties share in common. A mapping file to associate rep-county to actual county was developed based on each actual county's fuels, I/M, and meteorological data similarity to the rep-county. The mapping used in this application is shown in Table S2.1.

The focus of the current study is on the eastern US and, in particular, on four urban areas – Atlanta, Detroit, Philadelphia and St. Louis. County specific VMT distributions were applied in MOVES for all counties in the US, including those in these four areas as shown in Table S2.2. In addition, more detailed analysis was conducted for the Atlanta, Detroit and Philadelphia metropolitan areas using county specific age distributions obtained from the respective Metropolitan Planning Organizations (MPOs) and Inspection/Maintenance (I/M) and fuels data available in MOVES.

The rep-county emissions in the 220 counties were divided by the appropriate corresponding MOVES default VMT by SCC, month and county in order to develop representative emission rates. We used the latest available state-reported VMT activity data collected for the 2008 National Emissions Inventory (NEI) (EPA, 2011). The base year 2008 VMT contained in the EPA National County Database (NCD, version ncd20101201) was used directly for the 2008 baseline inventory, and projected to 2022 for the future year inventory. The projection of NEI VMT from 2008 to 2022 was performed using the MOVES assumptions of annual national VMT growth, vehicle sales, and scrappage rates over 2008 to 2022. The annual VMT from the NCD was apportioned to February and July using the MOVES monthly VMT distribution which reflects higher proportion of VMT in summer months than winter (as shown in Fig. S2.1). The emission rates for rep-counties were merged with the VMT for all counties to develop the national inventories for 2008 and 2022. We used the MOVES default fuel market share (of conventional and ethanol-blended gasoline) for 2008 but assumed 100% E10 (10% ethanol blend) usage for all counties in 2022 to avoid spatial differences in air quality model results due to differences in E10 usage across counties.

Fig. S2.2 shows the model-derived travel fraction distributions in 2022 in Atlanta, Detroit, Philadelphia and other parts of the nation including St. Louis. Travel fractions represent the relative fraction of travel from each vehicle age category of a given vehicle class. Because distance traveled is the basis of emission factors (grams per mile), they are the appropriate weighting fractions to aggregate over model year. An important characteristic of these distributions is that newer vehicles represent a disproportionately large fraction of the travel due to both higher number vehicles present in the fleet and more miles driven per vehicle. Detroit represents an extreme case of a high number of newer vehicles as seen in Fig. S2.2. The significance of differing age distributions (and thus travel fractions) is that an urban area with a high fleet turnover rate will receive a higher benefit of cleaner emission control technologies during early years of a phase-in schedule.

Travel fractions were calculated as product of relative mileage accumulation rate and age distribution. The relative mileage accumulations by age are stored in the MOVES database, with no geographic variation by county, and the relative mileages reflect the fact that newer vehicles tend to drive more miles annually than older vehicles. The age distributions were derived from 2005 vehicle registration data developed by MPOs in Atlanta, Detroit and Philadelphia. Because the MPOs developed the data primarily for MOBILE6 and not MOVES, the newest age category shows a “dip” due to a half-year age distribution in vehicle model year 2022, a requirement of MOBILE6 to use July age distribution that is not a feature in MOVES which assumes full year registration.

In order to estimate the effect on vehicle emissions of adherence to LEV III standards for VOC, CO, and NOx, LEV III to LEV II emissions ratios were estimated by pollutant and mode based on simulations with the California Air Resources Board’s (ARB) Emissions Factor Model (EMFAC2007) (http://arb.ca.gov/msei/onroad/latest_version.htm, accessed July 2011). The LEV III to LEV II emissions ratios were then used to adjust MOVES model LEV II emission rate input estimates to reflect LEV III emission rates. The EMFAC2007 model was run to estimate emissions from a calendar year and model year 2010 LEV II vehicle and a calendar year and model year 2016 Partial Zero Emissions Vehicle (PZEV) reflecting Super Ultra Low Emission Vehicle (SULEV) emission rates, the latter adhering to standards of non-methane organic gases (NMOG) + NOx similar to a LEV III vehicle. Total hydrocarbon (THC) LEV III to LEV II ratios were used as estimates of VOC LEV III to LEV II ratios. The emission rates and LEV III to LEV II ratios are shown in Table S2.3 by mode along with LEV III to LEV II ratios by pollutant and mode. It is clear that start emissions are preferentially reduced compared with running emissions, most notably for THC (or VOC). Table S2.4 provides the assumed LEV III phase-in schedule based on preliminary data from ARB regarding the LEV III phase-in schedule.

Light duty LEV III exhaust PM adjustments were estimated based on the ratio of LEV III to Tier 2 emission standards applied to MOVES Tier 2 emission factors in 2022 by vehicle model year. MOVES was run for 2022 to determine the 6-year old Tier 2 light duty vehicle exhaust PM emission factor of 6.9 mg/mi which was used as a surrogate for the Tier 2 fleet wide light duty PM standard. The vehicle age of six years was chosen because in MOVES the average light duty vehicle has nearly reached an accumulation of 120,000 miles which is the Tier 2 durability standard. Table S2.5 shows by vehicle model year the estimated Tier 2 exhaust PM multiplicative adjustment factor for estimating LEV III exhaust PM emission rates.

Per preliminary information from ARB regarding LEV III evaporative standards, LEV III standards would require all light duty vehicles be subject to an updated zero evaporative emissions standard similar to the zero evaporative emission standard to which vehicles can be certified under the LEV II program to acquire Zero Emission Vehicle (ZEV) credits. The EMFAC2007 model was run for a vehicle certified to LEV II zero evaporative emissions standards and a vehicle certified to LEV II near zero standards to estimate the ratio of near zero evaporative to zero evaporative standard emission rates. The ratio of near zero evaporative to zero evaporative standard emission rates were applied to MOVES Tier II vehicular emissions to estimate LEV III emissions, mapping EMFAC resting loss ratios to MOVES permeation and leaks processes and the sum of EMFAC diurnal, running, and hot soak losses to the MOVES vapor venting process. As indicated in Table S2.6, relatively larger emission reductions were estimated for diurnal emissions and resting losses with very small reductions to running losses and modest reductions to hot soak emissions. The model year phase-in of zero evaporative emission standards was assumed per preliminary rulemaking information from ARB: 60% in 2018 and 2019, 80% in 2020 and 2021, and 100% for 2022+.

The 2022 Tier 1 scenario represents the on-road emissions scenario that would occur in 2022 if light duty vehicles had never transitioned from Tier 1 to cleaner emission control technology. The Tier 1 scenario was generated similar to the LEV III scenario, in that scaling factors were applied to the 2022 Tier 2 inventory to adjust the light duty gasoline vehicle emissions. The LEV III scaling factors reduced emissions, while the Tier 1 scaling factors increase the Tier 2 emissions scenario. The Tier1/Tier2 multiplicative factors were applied on a fleet wide (average vehicle age) basis but were developed on a by-model-year basis.

MOVES underlying emission factors for each vehicle model year represent the technology in use in the given year and Table S2.7 below shows the schedule assumed by the model. Tier 1 emission factors for the 2001-2022 vehicle model years do not exist by default in MOVES. Rather, they are simulated as existing in 2022 by running multiple historic calendar years in MOVES keeping all other model assumptions the same as they are in 2022. MOVES contains 5 model years with 100% Tier 1 emission factors, 1996 through 2000. Thus running MOVES for calendar year 2000 provides 2000-1996 model year Tier 1 vehicle emission factors that are deteriorated 0 to 5 years. If such technology existed in 2022, vehicle model years 2022-2018 would have this same technology and deterioration combination. In order to piece together a full Tier 1 scenario fleet, MOVES was run for 2000, 2005, 2010 and 2020. The Tier 1 by model year fleet then contained Tier 1 emission factors for 2022-1996, and the historic pre-Tier 1 schedule shown in Table S2.7. The by model year emission rates for “as-is” 2022 fleet and for the constructed Tier 1 fleet were aggregated over model year by applying a weighted average of travel fraction, which is the product of registration distribution and relative mileage accumulation over the 30 model years. Then Tier 1/Tier 2 ratios by SCC, pollutant, emission process were applied to appropriate vehicle classes in the 2022 Tier 2 national inventory resulting in the Tier 1 scenario inventory.

On-road emissions for the zero-out LDV scenario were calculated by setting emissions for the following SCCs to zero in the 2022 Tier 2 inventory: SCC 2201001 for Light Duty Gasoline Vehicles, SCC 2201020 for Light Duty Gasoline Trucks 1 and 2, and SCC 2201040 for Light Duty Gasoline Trucks 3 and 4.

Table S2.1 Representative county criteria, binning, and basis in MOVES.

Parameter	Bin	Data Source
State	One for each state	FIPS code
Ethanol fuel as EtOHvol*EtOHMktShare (Ethanol volume percent * Ethanol market share)	0-3, 3-8, 8+	fuelsupply and fuelformulation tables (movesdb20100830 database)
Gasoline RVP	0-7.3, 7.3-8.2, 8.2-9.2, 9.2+	fuelsupply and fuelformulation table (movesdb20100830 database)
Sulfur	0-50, 50-100, 100-110, 110+	fuelsupply and fuelformulation table (movesdb20100830 database)
July monthly average maximum temperature (20-year historical average)	<=-20F, 10F bins from -20F to 100F, 100F+	ZoneMonthHour table (movesdb20100830 database)
February monthly average minimum temperature (20-year historical average)	<=-20F, 10F bins from -20F to 100F, 100F+	ZoneMonthHour table (movesdb20100830 database)
I/M	One for each unique I/M program	IMCoverage table (movesdb20100830 database)
Altitude	High or low	County table (movesdb20100830 database)

Table S2.2. Input data for on-road emission estimates.

Input Data	Atlanta, Detroit and Philadelphia Urban Areas	St. Louis Urban Area and Other Counties in US
VMT	Unique to county	Unique to county
Meteorology	Unique to urban area	Rep-county
I/M & Fuels	Unique to urban area	Rep-county
Age Distribution	Unique to urban area	National scale

Table S2.3. LEV II and PZEV THC, CO, and NOx emission rates with estimated LEV III to LEV II ratios.

Vehicle Technology	THC Exhaust Fractions		CO Fractions		NOx Fractions	
	Run	Start	Run	Start	Run	Start
Passenger Cars						
MY2010 (LEV II)	0.0026	0.0011	0.0672	0.0148	0.0030	0.0002
PZEV-Tech31 (LEV III)	0.0007	0.0002	0.0371	0.0065	0.0017	0.0001
LEV III/LEV II Ratio	0.29	0.14	0.55	0.44	0.58	0.38
Light Duty Trucks						
MY2010 (LEV II)	0.0035	0.0014	0.0655	0.0134	0.0034	0.0002
PZEV-Tech31 (LEV III)	0.0008	0.0002	0.0399	0.0070	0.0019	0.0001
LEV III/LEV II Ratio	0.23	0.12	0.61	0.52	0.55	0.36

Table S2.4. Phase-in of LEV III exhaust NMOG + NOx emission standards.

Model Year	LDT2	PC/LDT1
2014	0%	0%
2015	9%	9%
2016	18%	18%
2017	28%	27%
2018	37%	36%
2019	46%	45%
2020	55%	55%
2021	64%	64%
2022	73%	73%
2023	83%	82%
2024	92%	91%
2025	100%	100%

Table S2.5. LEV III to Tier 2 exhaust PM adjustment ratios.

Vehicle Model Year	2016 and earlier	2017	2018	2019	2020	2021	2022
LEV III/Tier 2	1	0.87	0.73	0.58	0.44	0.44	0.44

Table S2.6. EMFAC 2007 evaporative reactive organic gas emission rates (g/mi) and estimated LEV III to LEV II ratio.

Vehicle Technology	Diurnal	Hot soak	Running loss	Resting loss
Ev015 (near zero)	0.38	0.62	10.19	0.33
Ev017 (PZEV)	0.03	0.33	9.85	0.06
LEV III to LEV II ratio	8%	53%	97%	17%

Table S2.7. Vehicle model year schedule assumed in the calculation of 2022 Tier 1 emissions.

Year	MOVES2010a Emission Standard
2022	100% Tier 2
…	100% Tier 2
2007	100% Tier 2
2006	75% Tier 2, 25% LEV
2005	50% Tier 2, 50% LEV
2004	25% Tier 2, 75% LEV
2003	100% LEV
2002	20% TLEV, 80% LEV
2001	40% TLEV, 60% LEV
2000	100% Tier 1
…	100% Tier 1
1996	100% Tier 1
1995	80% Tier 1, 20% Tier 0
1994	40% Tier 1, 60% Tier 0
1993	100% Tier 0
1992	100% Tier 0

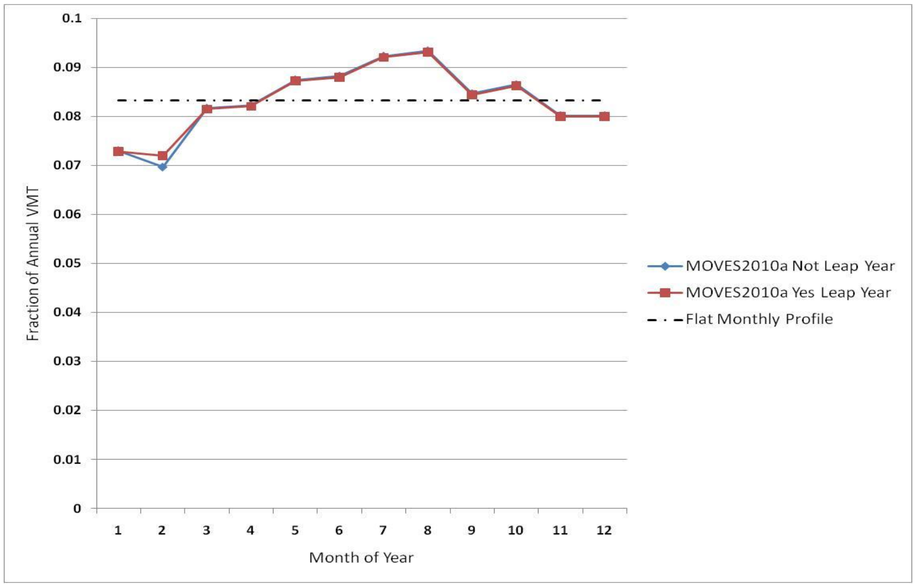


Fig. S2.1. Monthly VMT Distribution from MOVES

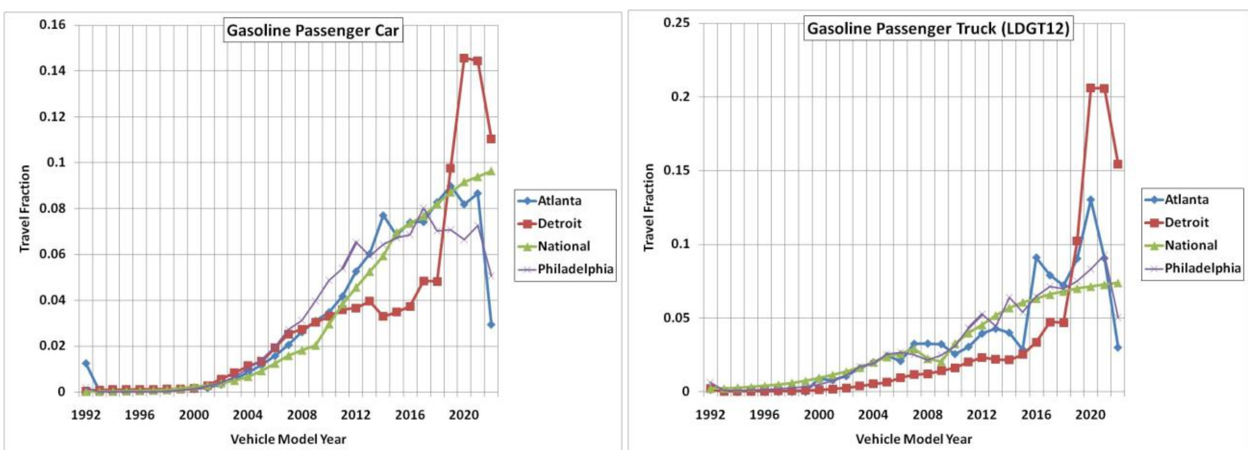


Fig. S2.2. Estimated Travel Fraction Distributions in 2022.

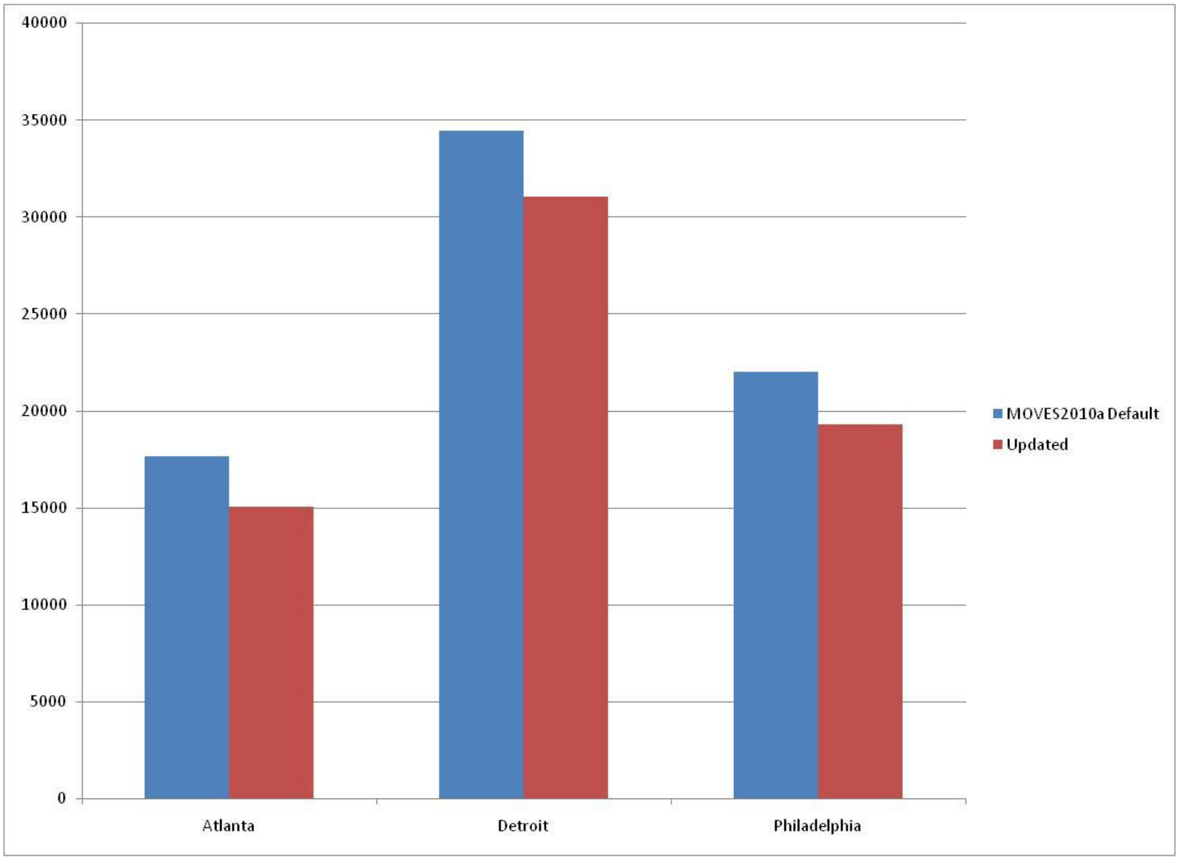


Fig. S2.3. VOC emissions (lbs/day) in three urban areas with MOVES default and local vehicle age distributions.

S3. Statistical indicators for meteorological and air quality model evaluation

Mean Observation (M_o) (in units of observed quantity):

$$M_o = \frac{1}{IJ}\sum_{j=1}^{J}\sum_{i=1}^{I} C_{o,j}^{i}$$

where $C^{i}_{o,j}$ is the individual observed quantity at site *i* and time *j*, and the summations are over all sites (*I*) and over time periods (*J*).

Mean Prediction (M_p) (in units of predicted quantity):

$$M_p = \frac{1}{IJ}\sum_{j=1}^{J}\sum_{i=1}^{I} C_{m,j}^{i}$$

where $C^{i}_{m,j}$ is the individual predicted quantity at site *i* and time *j*.

Mean Bias (MB) (in units of predicted quantity):

$$MB = \frac{1}{N}\sum_{i=1}^{N}\left(C_m - C_o\right)$$

In this equation and hereafter, C_m and C_o are modeled (i.e., predicted) and observed concentrations and N is the total number of data points.

Mean Error (ME) (in units of predicted quantity):

$$ME = \frac{1}{N}\sum_{i=1}^{N}\left|C_m - C_o\right|$$

Mean Normalized Bias (MNB) (%):

$$MNB = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{C_m - C_o}{C_o}\right)$$

Mean Normalized Gross Error (MNGE) (%)

$$MNGE = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{C_m - C_o}{C_o}\right|$$

Mean Fractional Bias (MFB) (%)

$$MFB = \frac{1}{N}\sum_{i=1}^{N}\frac{\left(C_m - C_o\right)}{\left(\frac{C_o + C_m}{2}\right)}$$

Mean Fractional Error (MFE) (%)

$$MFE = \frac{1}{N}\sum_{i=1}^{N}\frac{\left|C_m - C_o\right|}{\left(\frac{C_o + C_m}{2}\right)}$$

S4. Meteorology

Table S4.1 Vertical grid structure for meteorological and air quality modeling

WRF Vertical Layers					CAMx Vertical Layers			
k (MM5)	sigma	Press (mb)	Height (m)	Depth (m)	k (CAMx)	sigma	Height(m)	Depth (m)
34	0.000	50	19532	3330				
33	0.050	97.5	16202	2257				
32	**0.100**	**145**	**13945**	**1739**	**26**	**0.100**	**13945**	**1739**
31	**0.150**	**192.5**	**12206**	**1426**	**25**	**0.150**	**12206**	**1426**
30	**0.200**	**240**	**10780**	**1216**	**24**	**0.200**	**10780**	**1216**
29	**0.250**	**287.5**	**9564**	**1063**	**23**	**0.250**	**9564**	**1063**
28	**0.300**	**335**	**8501**	**947**	**22**	**0.300**	**8501**	**1802**
27	0.350	382.5	7554	855				
26	**0.400**	**430**	**6699**	**781**	**21**	**0.400**	**6699**	**1500**
25	0.450	477.5	5919	719				
24	**0.500**	**525**	**5200**	**667**	**20**	**0.500**	**5200**	**1290**
23	0.550	572.5	4532	623				
22	**0.600**	**620**	**3910**	**584**	**19**	**0.600**	**3910**	**1134**
21	0.650	667.5	3326	550				
20	**0.700**	**715**	**2775**	**419**	**18**	**0.700**	**2775**	**419**
19	**0.740**	**753**	**2357**	**303**	**17**	**0.740**	**2357**	**303**
18	**0.770**	**781.5**	**2054**	**293**	**16**	**0.770**	**2054**	**293**
17	**0.800**	**810**	**1761**	**191**	**15**	**0.800**	**1761**	**191**
16	**0.820**	**829**	**1570**	**187**	**14**	**0.820**	**1570**	**187**
15	**0.840**	**848**	**1383**	**184**	**13**	**0.840**	**1383**	**184**
14	**0.860**	**867**	**1199**	**180**	**12**	**0.860**	**1199**	**180**
13	**0.880**	**886**	**1018**	**177**	**11**	**0.880**	**1018**	**177**
12	**0.900**	**905**	**841**	**87**	**10**	**0.900**	**841**	**174**
11	0.910	914.5	754	87				
10	**0.920**	**924**	**667**	**86**	**9**	**0.920**	**667**	**171**
9	0.930	933.5	581	85				
8	**0.940**	**943**	**496**	**84**	**8**	**0.940**	**496**	**84**
7	**0.950**	**952.5**	**412**	**84**	**7**	**0.950**	**412**	**84**
6	**0.960**	**962**	**328**	**83**	**6**	**0.960**	**328**	**83**
5	**0.970**	**971.5**	**245**	**82**	**5**	**0.970**	**245**	**82**
4	**0.980**	**981**	**163**	**41**	**4**	**0.980**	**163**	**41**
3	**0.985**	**985.75**	**122**	**41**	**3**	**0.985**	**122**	**41**
2	**0.990**	**990.5**	**81**	**41**	**2**	**0.990**	**81**	**41**
1	**0.995**	**995.25**	**40**	**40**	**1**	**0.995**	**40**	**40**
0	**1.000**	**1000**	**0**	**40**	**0**	**1.000**	**0**	**40**

<u>Meteorology Evaluation</u>

The 2008 WRF meteorology has been previously evaluated by EPA against hourly surface observations of temperature and wind speed over the CONUS and good model performance has been noted (Gilliam, R., unpublished results, personal communication, 2011): mean bias (MB) for temperature = 0.3 K in winter and 0.0 K in summer, MB for wind speed = -0.1 m/s in winter and -0.1 m/s in summer, mean error (ME) for temperature = 2.0 K in winter and 1.6 K in summer, ME for wind speed = 1.4 m/s in winter and 1.3 m/s in summer, Index of Agreement (IOA) for temperature = 1.0 in winter and 1.0 in summer, and IOA for wind speed = 0.7 in winter and 0.7 in summer. The formulae for these and other statistical measures calculated in this study are presented in Section S3 in the Supplementary Data.

As an additional check on the reliability of the meteorology used for O_3 and $PM_{2.5}$ modeling, we evaluated the CAMx meteorology developed from the 2008 EPA WRF meteorology against observations at stations in the ds472.0 database (RDA, 2011). The evaluation was conducted for hourly surface temperature, wind speed, wind direction and humidity in February and July 2008 at approximately 650 stations in the CAMx 12 km domain which comprises the region of interest in this study. Table S4.2 presents a summary of the statistical metrics. Overall, the performance of the meteorological fields input to CAMx is good with low bias and error and high IOA.

Table S4.2. Statistical evaluation of meteorological fields used in air quality modeling in the eastern US.

Month	**Parameter**	**Metric**	**Units**	**Value**
February 2008	Temperature	Mean Bias (MB)	K	0.18
		Mean Error (ME)	K	0.92
		Index of Agreement (IOA)		0.96
	Wind Speed	Mean Bias (MB)	m/s	0.05
		Mean Error (ME)	m/s	0.05
		Index of Agreement (IOA)		0.74
	Wind Direction	Mean Bias (MB)	degrees	8.33
		Mean Error (ME)	degrees	13.65
	Humidity	Mean Bias (MB)	g/kg	0.25
		Mean Error (ME)	g/kg	0.26
		Index of Agreement (IOA)		0.91
July 2008	Temperature	Mean Bias (MB)	K	0.22
		Mean Error (ME)	K	0.4
		Index of Agreement (IOA)		0.86
	Wind Speed	Mean Bias (MB)	m/s	0.18
		Mean Error (ME)	m/s	0.29
		Index of Agreement (IOA)		0.64
	Wind Direction	Mean Bias (MB)	degrees	8.35
		Mean Error (ME)	degrees	21.44
	Humidity	Mean Bias (MB)	g/kg	0.48
		Mean Error (ME)	g/kg	0.54
		Index of Agreement (IOA)		0.83

S5. Speciation Profiles

The following speciation profiles from SPECIATE 4.3 were used to develop model-ready on-road emissions.

Table S5.1 VOC speciation profiles for on-road emissions

Source Type	Profile ID	Profile Description
Gasoline		
Start Exhaust	8760	Gasoline Exhaust - Cold Start - Tier 2 light-duty vehicles using 10% Ethanol - Composite Profile
Running Exhaust (CY 2008)	8751	Gasoline Exhaust - E10 ethanol gasoline (Pre-Tier 2)
Running Exhaust (CY 2022)	8757	Gasoline Exhaust - Tier 2 light-duty vehicles using 10% Ethanol
Evaporative: permeation	8769	Diurnal Permeation Evaporative Emissions from Gasoline Vehicles using 10% Ethanol – Combined
Evaporative: non-permeation	8754	Gasoline Vehicle - Evaporative emission - E10 ethanol gasoline
Stage I&II Refueling	8763	Gasoline Headspace Vapor using 10% Ethanol - Composite Profile
Diesel		
Exhaust	4674	Diesel exhaust - medium duty trucks
Evaporative	4547	Gasoline Headspace Vapor - Circle K Diesel

*We note that the running exhaust profiles did not incorporate influence of age distributions (i.e., pre- versus post-Tier 2). The pre-Tier 2 profile is used in the 2008 base year and the Tier 2 profile is used in the 2022 future year.

S6. CAMx 2008 Base Case Performance Evaluation

Tables S6.1 and S6.2 show the CAMx model performance statistics for 1-hour and 8-hour ozone, respectively. The statistical measures shown in this table are listed by EPA (2007) and defined in Section S3 in the supplementary information. A threshold value of 40 ppb for the observed hourly ozone concentrations was used to remove the influence of low observed concentrations on the performance statistics.

The EPA no longer recommends numerical goals for statistical metrics, but the previously recommended performance goals of ±20% unpaired peak accuracy (UPA), ±15% mean normalized bias (MNB) and ±35% mean normalized gross error (MNGE) (EPA, 1991) are still widely used to evaluate ozone model performance. The UPA goals are met here in all cases except in the 36 km domain in February for AIRS/AQS and in the 12 km domain in July for CASTNET. MNB goals are met in February in the 36 km and 12 km domains for both AIRS/AQS and CASTNET. However ozone concentrations are over-predicted in July for both networks; this may be due to one or more of the following factors: excess NOx emissions (see discussion below) in a VOC-rich environment, exceptional events such as fires and excess biogenic VOC emissions. The MNGE goals are met for all cases in Table S6.1. The model performance for 8-hour ozone is similar with good UPA and MNGE and slightly high MNB. Overall, model performance for ozone is reasonable.

Tables S6.3 and S6.4 show model performance evaluation statistics against NOx and NOy measurements in the AIRS/AQS network. Performance for NOx is much better in the eastern US domain (the area of focus in this study) than in the CONUS domain at 36 km resolution. NOx concentrations are slightly over-predicted (MNB of 11%) in July in the eastern US. NOy concentrations are also over-predicted in the same period (see Table S6.4). While a direct comparison of the NOx and NOy performance is not possible because the statistics are computed across different numbers of stations, the results suggest that there may be excess NOx emissions in some regions in the inventory. In general, model performance for NOx and NOy is satisfactory compared to prior modeling studies (e.g., Biswas et al., 2001; Liang et al., 2004; Hu et al., 2010).

The mean fractional bias (MFB) and mean fractional error (MFE) of simulated versus observed values were calculated to assess the model performance. MFB and MFE are useful for evaluating $PM_{2.5}$ predictions because they give equal weight to under- and over-predictions, both have an ideal value of 0, and both require no minimum threshold in order to be appropriate for evaluation (Seigneur et al., 2000). Model performance results for 24-hour $PM_{2.5}$, presented in Table S6.5, show MFB ranging from 2 to 50% and MFGE from 33 to 55% in the eastern US; these results are satisfactory compared to previous studies (e.g., Baker and Scheff, 2007; Gaydos et al., 2007; Russell, 2008).

Figures S6.1 – S6.3 show bugle plots of MFB and MFE versus observed values of $PM_{2.5}$ mass and components in the eastern US (12 km domain) along with the corresponding model performance goal and criterion, following Boylan and Russell (2006). The four data points shown in each plot are the monthly mean of 24-hr averaged $PM_{2.5}$ statistics in February (winter) and July (summer) in the AIRS/AQS and IMPROVE networks. Model performance goal and criterion are defined as 'the best the model can achieve' and 'acceptable model performance', respectively, and both are set based on current knowledge and technological capabilities to simulate the value of interest (Boylan and Russell, 2006). Total $PM_{2.5}$ mass shows very good performance in summer with the MFB and MFE lying within the goal envelope. $PM_{2.5}$ mass is over-predicted in winter; this is due, in large part, due to the over-prediction of fine nitrate. However, $PM_{2.5}$ performance in winter is still within the criterion envelope demonstrating satisfactory model performance. The MFB and MFE for all $PM_{2.5}$ components except organic carbon (OC), fall within the goal envelope showing very good model performance. The statistics for OC fall within the criterion envelope exhibiting satisfactory model performance.

Overall, the air quality model performance for the 2008 base case is good.

Table S6.1 Statistical metrics for 1-hour ozone*

	AIRS/AQS network				**CASTNET network**			
	February 2008		**July 2008**		**February 2008**		**July 2008**	
Metric	**36 km** (591 stations)	**12 km** (103 stations)	**36 km** (1186 stations)	**12 km** (532 stations)	**36 km** (79 stations)	**12 km** (41 stations)	**36 km** (78 stations)	**12 km** (40 stations)
Mean observed (ppb)	40.6	33.9	56.9	59.4	41.2	37.7	56.4	56.8
Mean predicted (ppb)	49.5	43.5	68.5	72.4	49.7	48.6	65.5	71.3
Unpaired peak accuracy (UPA) (%)	-26.0	-0.1	-16.6	12.4	0.4	7.1	-9.6	26.5
Mean Normalized Bias (MNB) (%)	8.0	5.6	20.4	22.9	7.3	7.4	16.6	24.5
Mean Normalized Error (MNE) (%)	16.8	13.6	25.8	28.1	13.9	14.5	22.8	26.8

* Number of stations with valid data is approximate as it varies daily.
Threshold of 40 ppb used to calculate UPA, MNB and MNE.

Table S6.2. Statistical metrics for 8-hour ozone*

Metric	AIRS/AQS network				CASTNET network			
	February 2008		July 2008		February 2008		July 2008	
	36 km (591 stations)	**12 km** (103 stations)	**36 km** (1186 stations)	**12 km** (532 stations)	**36 km** (79 stations)	**12 km** (41 stations)	**36 km** (78 stations)	**12 km** (40 stations)
Mean observed (ppb)	36.2	30.6	50.4	53.1	38.7	34.9	51.4	51.6
Mean predicted (ppb)	46.1	39.9	62.8	67.1	47.5	45.8	62.5	66.9
Unpaired peak accuracy (UPA) (%)	-11.9	2.9	-7.3	22.3	4.7	9.7	-4.8	28.4
Mean Normalized Bias (MNB) (%)	7.8	5.6	29.1	25.6	7.6	7.7	17.7	26.7
Mean Normalized Error (MNE) (%)	15.5	11.3	36.1	28.6	13.3	12	22.8	27.9

* Number of stations with valid data is approximate as it varies daily.

Threshold of 40 ppb used to calculate UPA, MNB and MNE.

Table S6.3. Statistical metrics for hourly NOx*

Metric	AIRS/AQS network			
	February 2008		July 2008	
	36 km (330 stations)	**12 km** (97 stations)	**36 km** (335 stations)	**12 km** (103 stations)
Mean observed (ppb)	60.9	64.7	27.2	32.7
Mean predicted (ppb)	21.6	41.5	16.4	28.4
Unpaired peak accuracy (UPA) (%)	-81.3	-46.4	-67.9	-29.7
Mean Normalized Bias (MNB) (%)	-41.8	-10.3	-28.9	11.2
Mean Normalized Error (MNE) (%)	63.1	59.9	58.3	66.1

* Number of stations with valid data is approximate as it varies daily.

Threshold of 10 ppb used to calculate UPA, MNB and MNE.

Table S6.4. Statistical metrics for hourly NOy*

Metric	AIRS/AQS network			
	February 2008		July 2008	
	36 km (25 stations)	12 km (11 stations)	36 km (41 stations)	12 km (22 stations)
Mean observed (ppb)	33.7	22.9	18.9	22.9
Mean predicted (ppb)	20.3	25.5	19.4	25.5
Unpaired peak accuracy (UPA) (%)	-62.3	9.0	-29.9	9.0
Mean Normalized Bias (MNB) (%)	-6.9	26.6	13.9	26.6
Mean Normalized Error (MNE) (%)	61.4	54.3	54.3	54.3

* Number of stations with valid data is approximate as it varies daily.

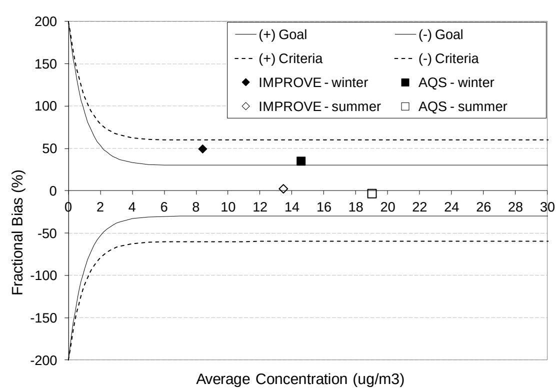
Threshold of 10 ppb used to calculate UPA, MNB and MNE.

Table S6.5. Statistical metrics for daily (24-hr) $PM_{2.5}$*

Metric	AIRS/AQS network				IMPROVE network			
	February 2008		July 2008		February 2008		July 2008	
	36 km (187 stations)	**12 km** (115 stations)	**36 km** (187 stations)	**12 km** (115 stations)	**36 km** (145 stations)	**12 km** (26 stations)	**36 km** (145 stations)	**12 km** (26 stations)
Mean observed ($\mu g/m^3$)	13.9	14.6	16.7	19.0	4.2	8.4	8.5	13.5
Mean predicted ($\mu g/m^3$)	15.8	20.6	15.3	18.0	7.1	13.4	9.0	13.4
Mean Fractional Bias (MFB) (%)	15.4	35.2	-8.1	-3.4	63.2	49.6	3.8	2.2
Mean Fractional Gross Error (MFE) (%)	44.3	45.4	37.8	34.4	69.0	54.7	39.6	32.8

* Number of stations with valid data is approximate as it varies daily. No threshold used.

(a) $PM_{2.5}$ fractional bias



(b) $PM_{2.5}$ fractional error

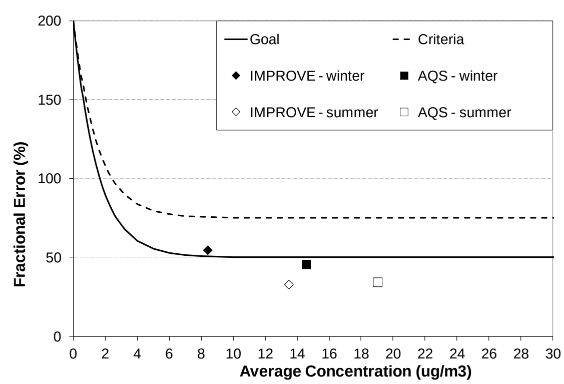
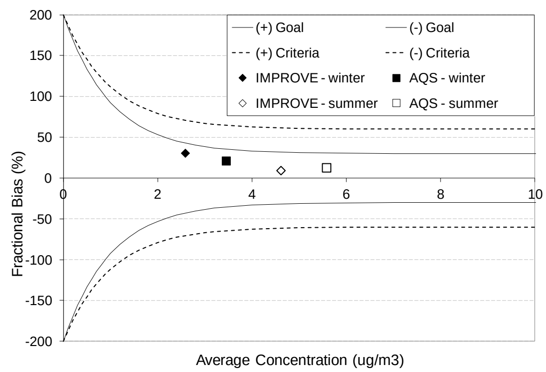
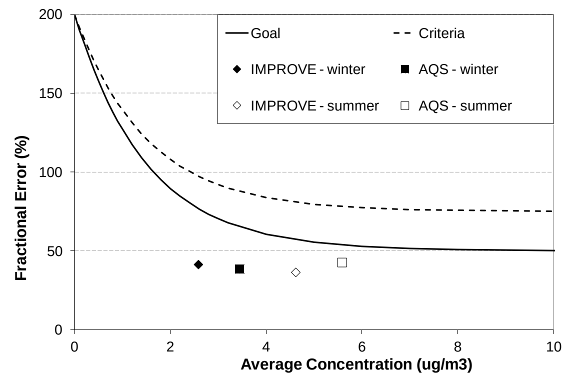


Fig. S6.1. Bugle plots of monthly mean fractional bias and fractional error of daily concentrations of $PM_{2.5}$ in the eastern US (12 km domain) in February and July, 2008.

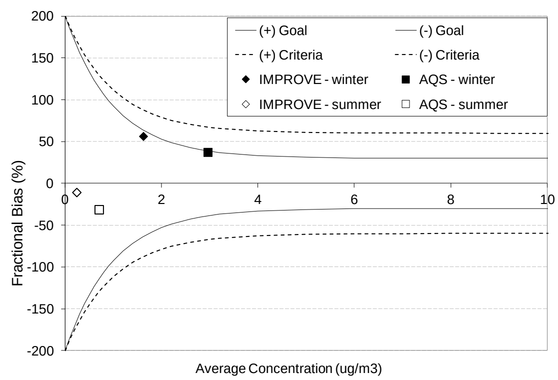
(a) Sulfate fractional bias



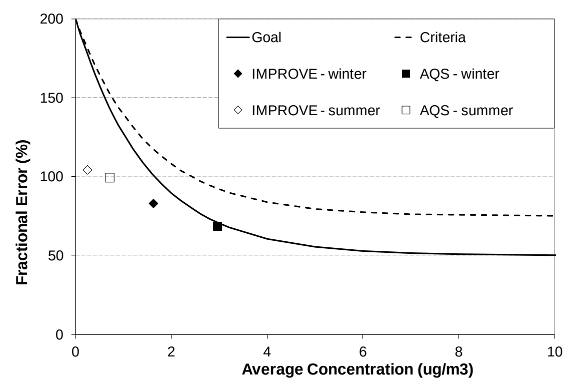
(b) Sulfate fractional error



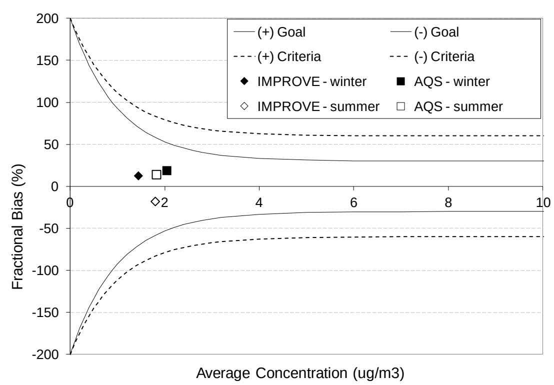
(c) Nitrate fractional bias



(d) Nitrate fractional error



(e) Ammonium fractional bias



(f) Ammonium fractional error

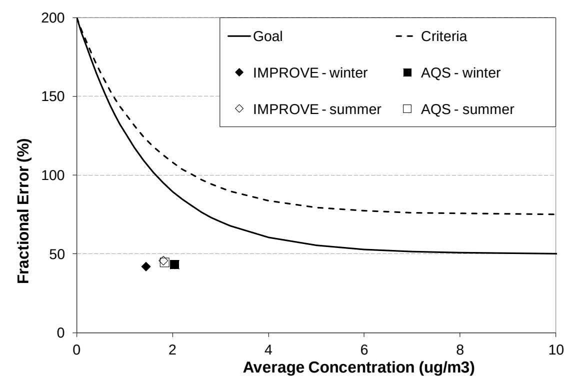
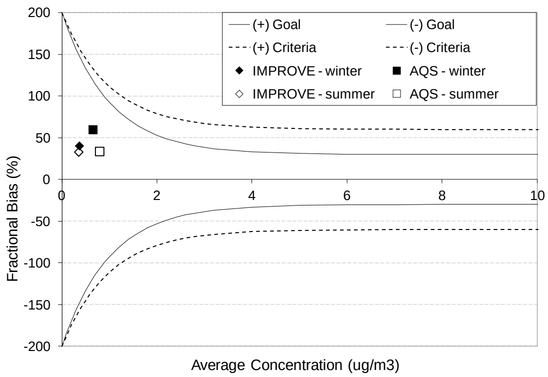
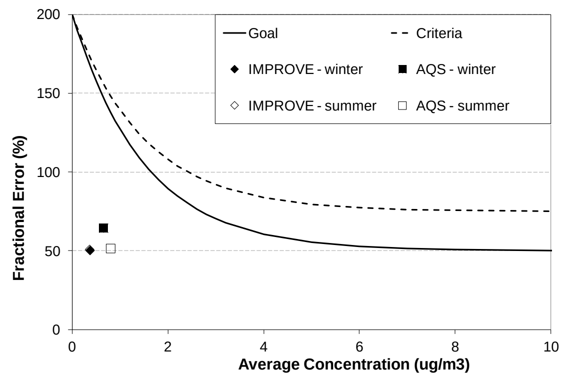


Fig. S6.2. Bugle plots of monthly mean fractional bias and fractional error of daily concentrations of $PM_{2.5}$ sulfate, nitrate and ammonium in the eastern US (12 km domain) in February and July, 2008.

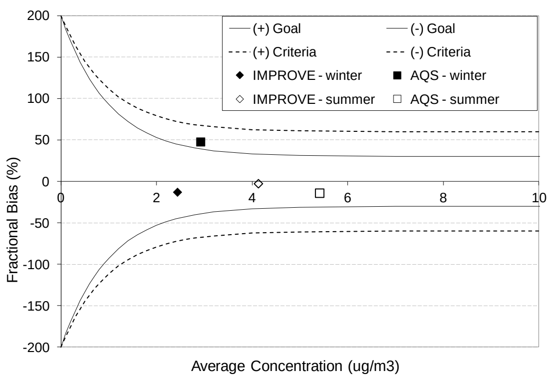
(a) Elemental carbon fractional bias



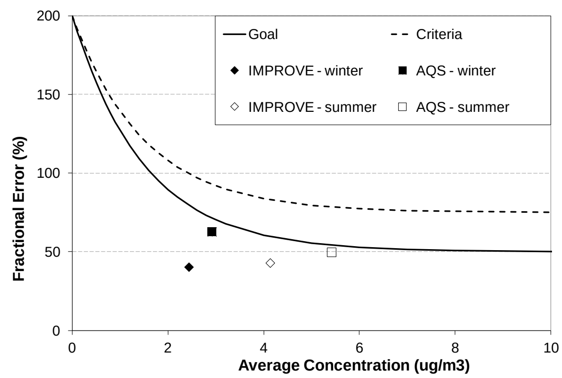
(b) Elemental carbon fractional error



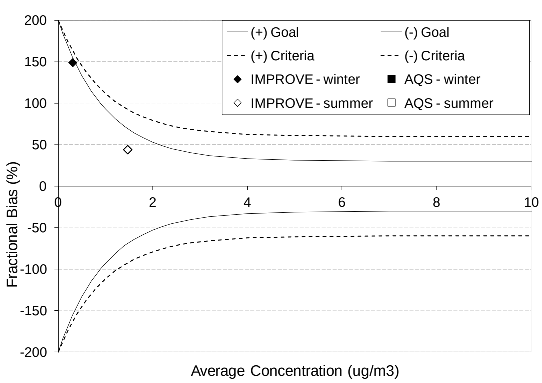
(c) Organic carbon fractional bias



(d) Organic carbon fractional error



(e) Soil fractional bias



(f) Soil fractional error

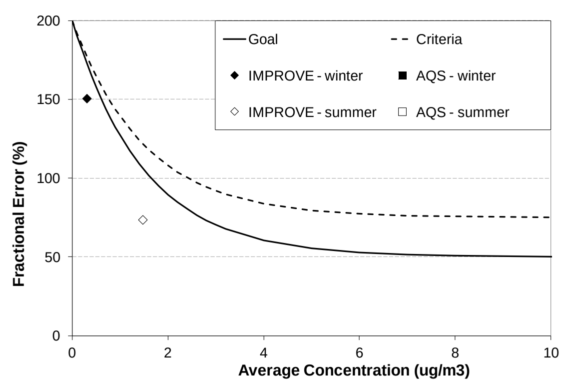


Fig. S6.3. Bugle plots of monthly mean fractional bias and fractional error of daily concentrations of $PM_{2.5}$ elemental carbon, organic carbon and soil material in the eastern US (12 km domain) in February and July, 2008.

S7. Spatial Distributions of Predicted On-road Emissions

The predicted spatial distributions of weekday on-road emissions of NOx, VOC and $PM_{2.5}$ (in short tons/day; 1 short ton is ~0.907 Mg) in the eastern US 12 km domain in February and July in the 2008 base case are shown in Fig. S7.1. The spatial distributions of winter weekday on-road emissions of NOx, VOC and $PM_{2.5}$ in the 12 km domain in the 2022 scenarios are presented in Figures S7.2 to S7.4. Similar plots for summer are shown in Figures S7.5 to S7.7. The trend for on-road emissions to decrease substantially from Tier 1 to Tier 2 scenarios and then only slightly with LEV III controls is predicted broadly across the eastern US and particularly in urban areas. In addition, the transition from the 2022 Tier 2 program to complete elimination of LDV emissions results in emissions reductions that are smaller than those obtained in progressing from the Tier 1 to the Tier 2 programs.

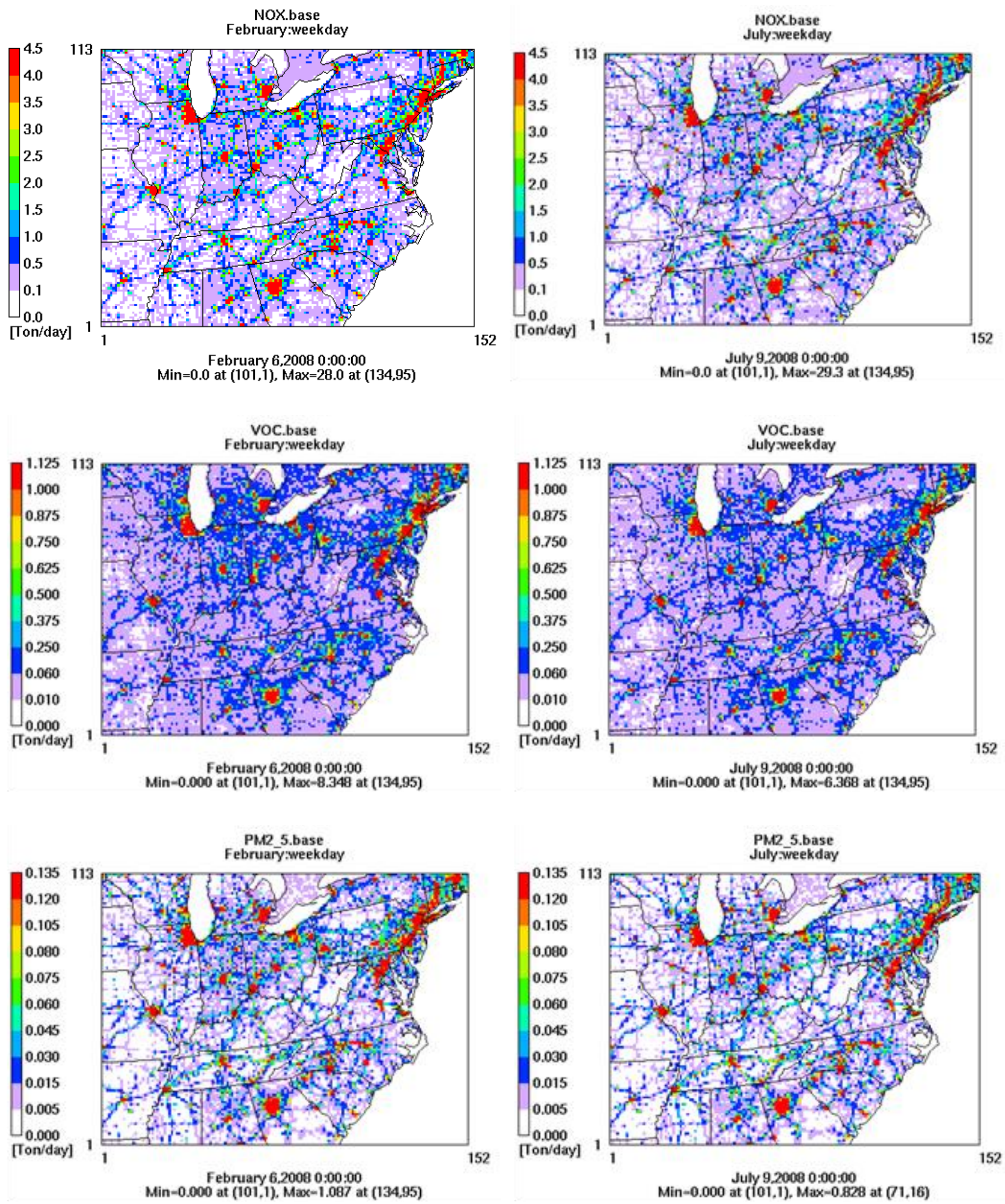


Fig. S7.1. Estimated on-road emissions of NOx (top), VOC (center) and $PM_{2.5}$ (bottom) in February (left) and July (right), 2008.

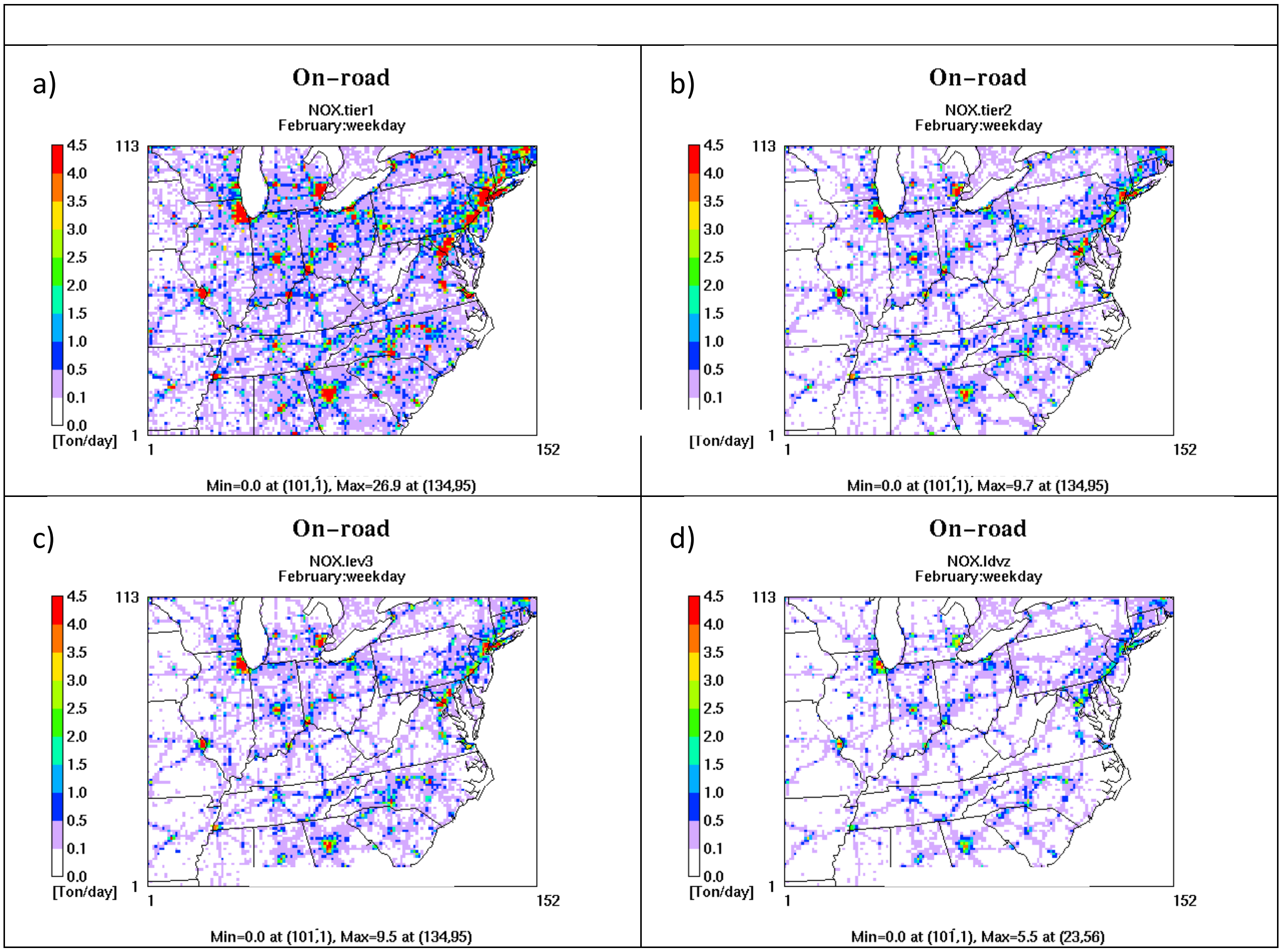


Fig. S7.2. Winter on-road NOx emissions: a) 2022 Tier1, b) 2022 Tier2, c) 2022 LEV III, d) 2022 zero-out LDV.

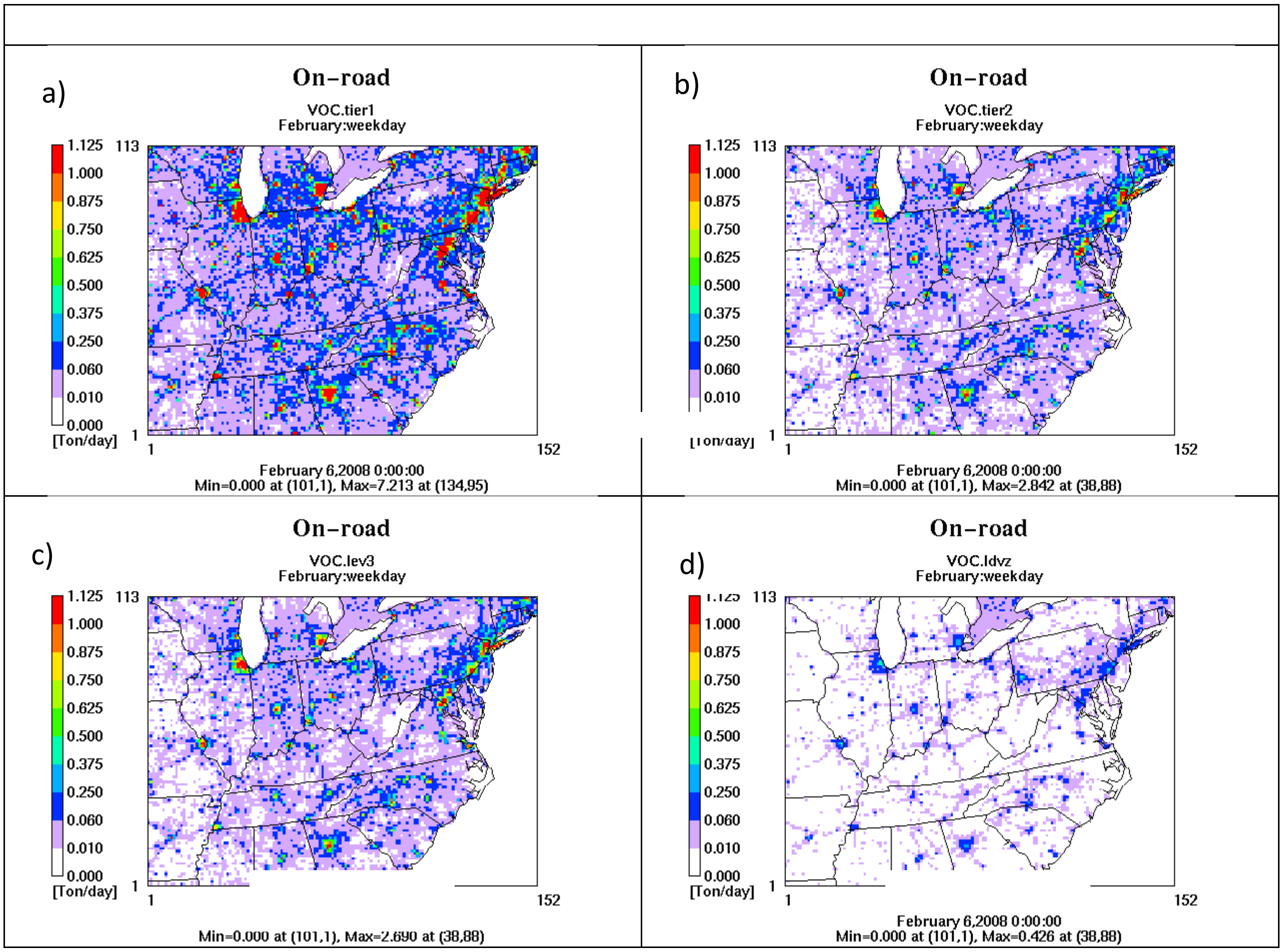


Fig. S7.3. Winter on-road VOC emissions: a) 2022 Tier1, b) 2022 Tier2, c) 2022 LEV III, d) 2022 zero-out LDV.

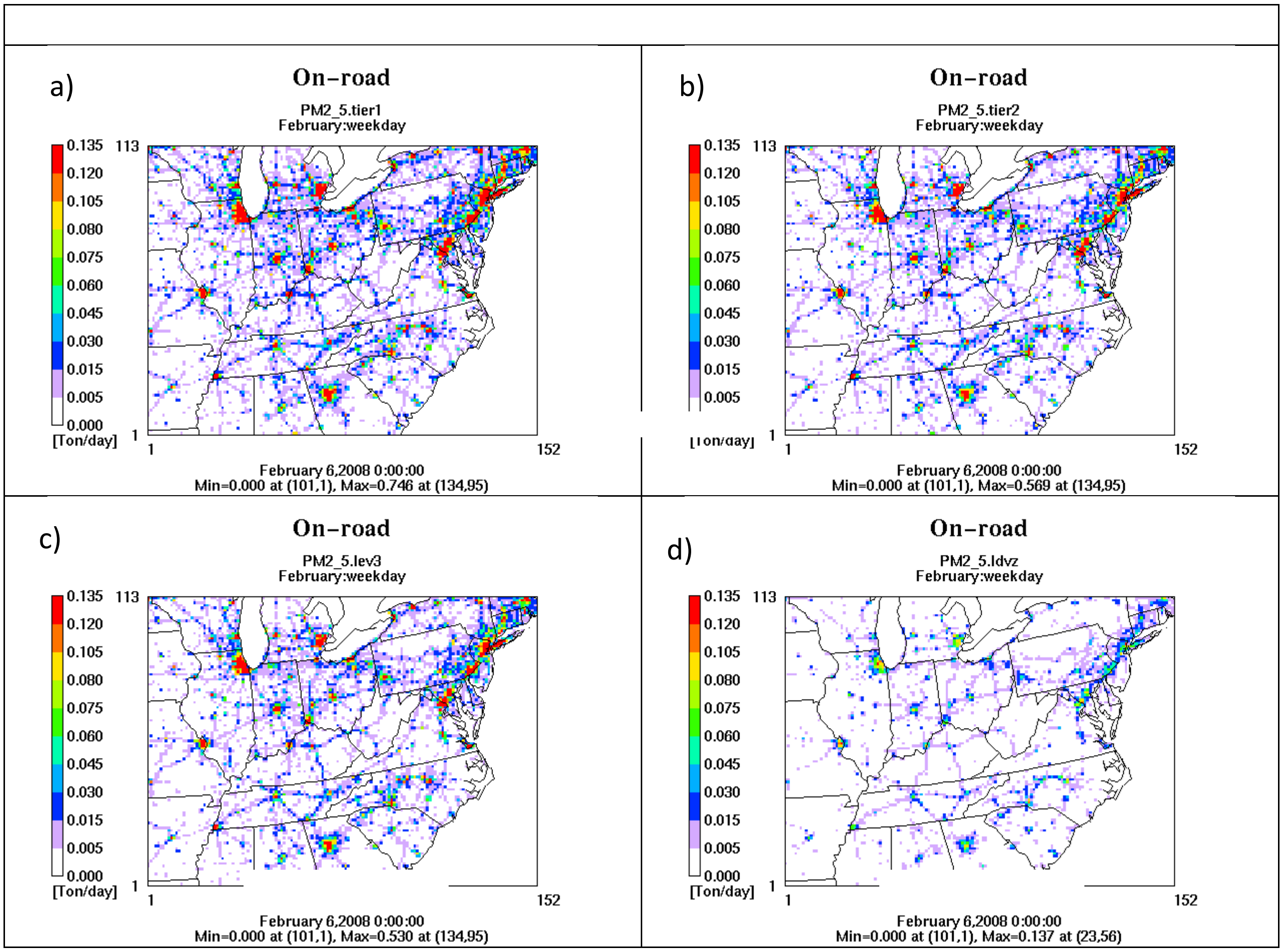


Fig. S7.4. Winter on-road $PM_{2.5}$ emissions: a) 2022 Tier1, b) 2022 Tier2, c) 2022 LEV III, d) 2022 zero-out LDV.

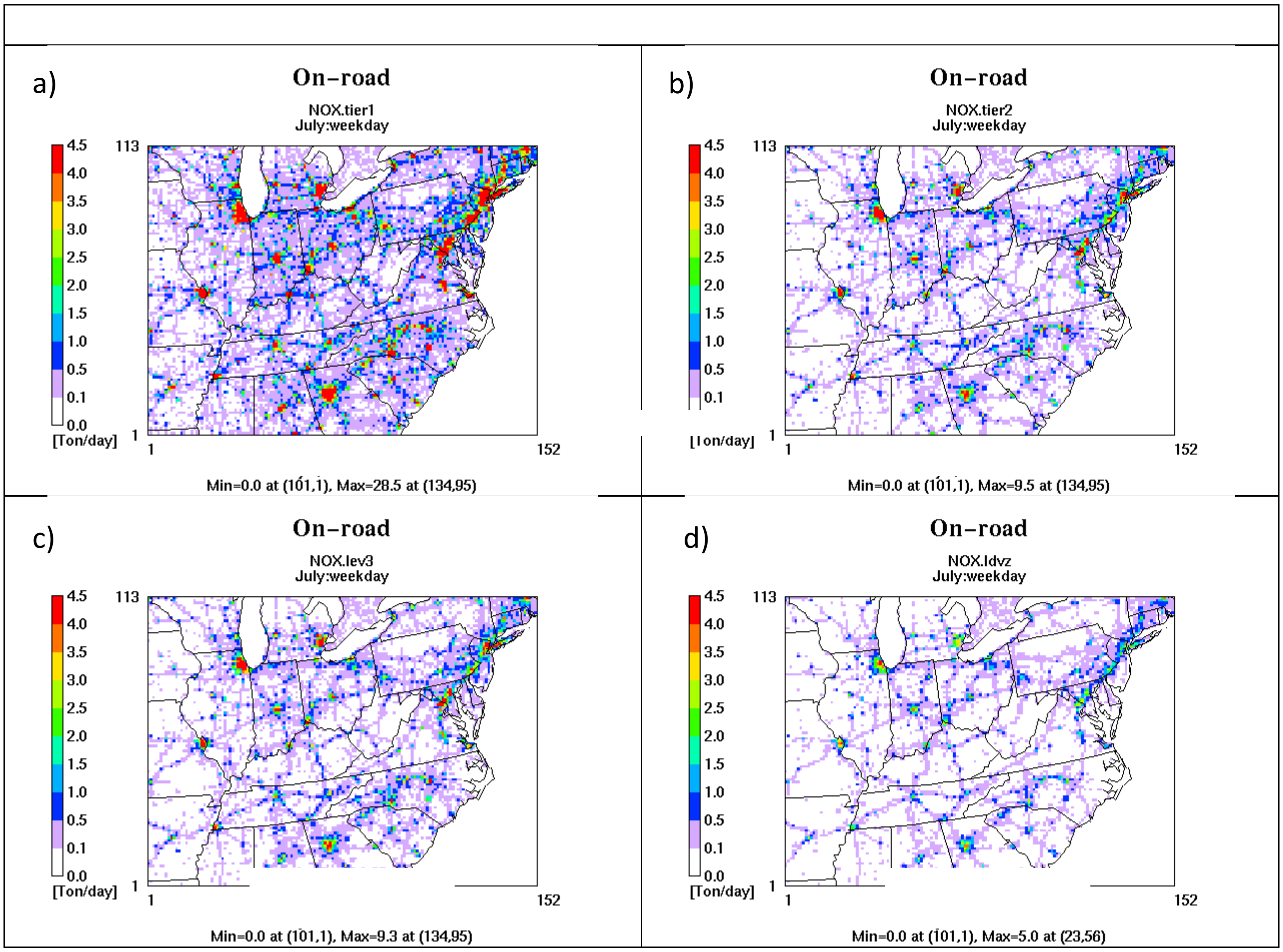


Fig. S7.5. Summer on-road NOx emissions: a) 2022 Tier1, b) 2022 Tier2, c) 2022 LEV III, d) 2022 zero-out LDV.

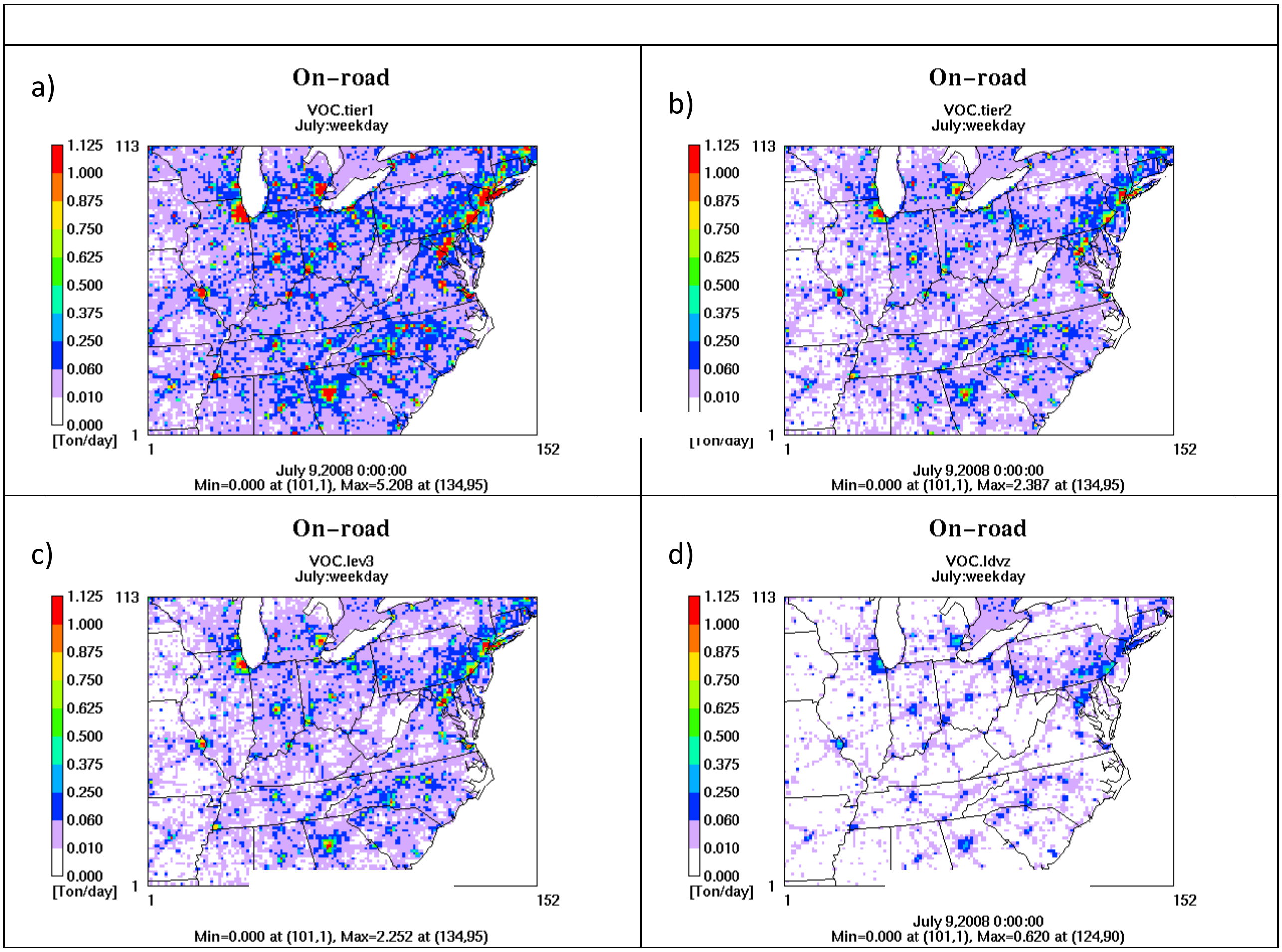


Fig. S7.6. Summer on-road VOC emissions: a) 2022 Tier1, b) 2022 Tier2, c) 2022 LEV III, d) 2022 zero-out LDV.

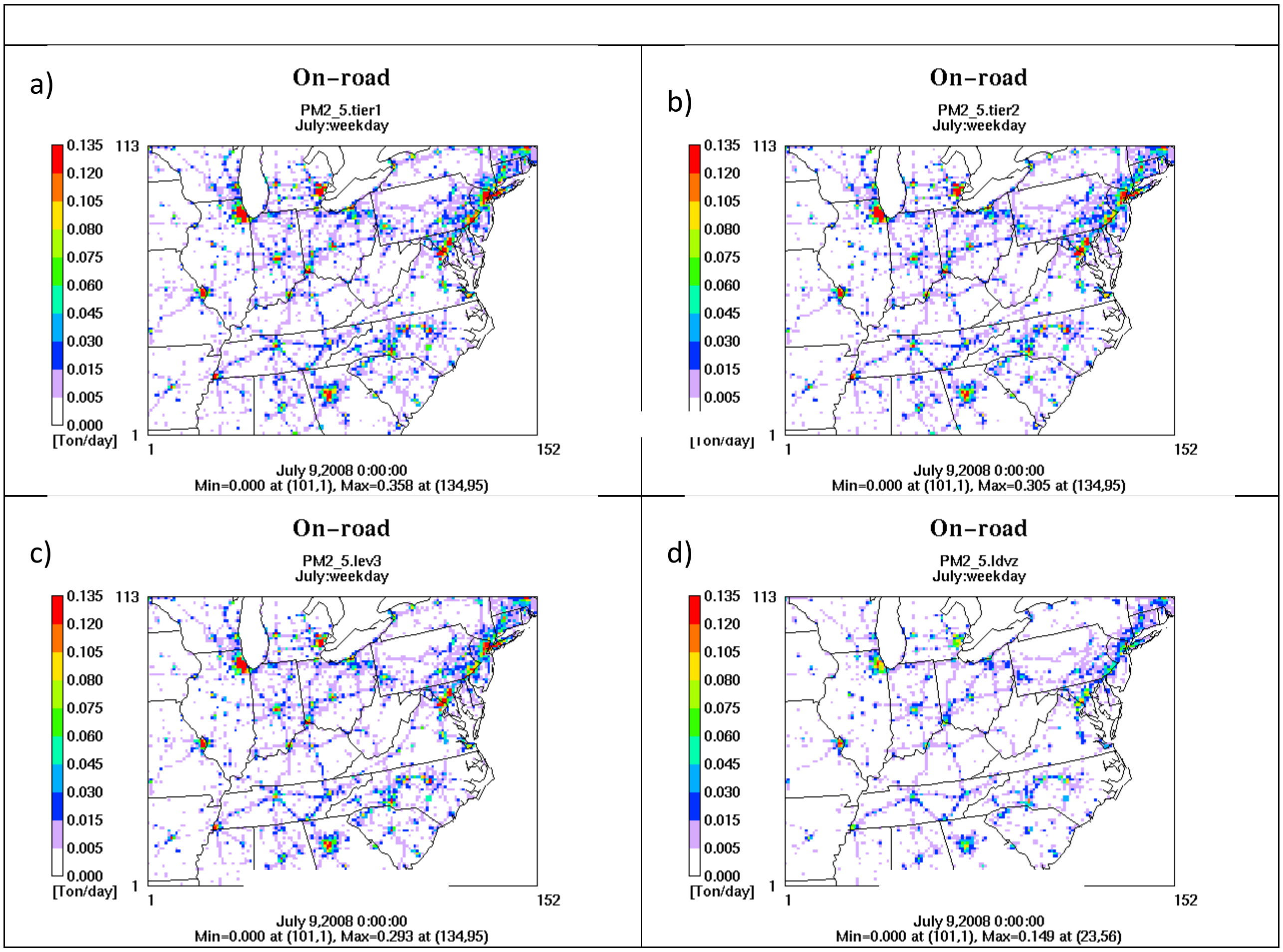
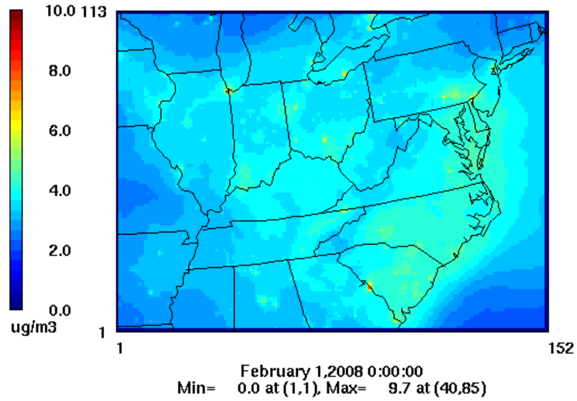


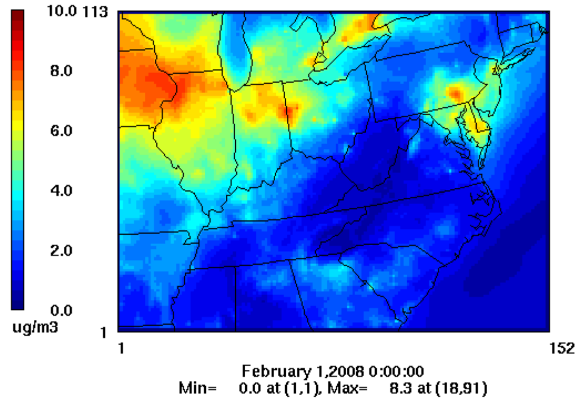
Fig. S7.7. Summer on-road $PM_{2.5}$ emissions: a) 2022 Tier1, b) 2022 Tier2, c) 2022 LEV III, d) 2022 zero-out LDV.

S8. Predicted spatial distributions of $PM_{2.5}$ components in 2008.

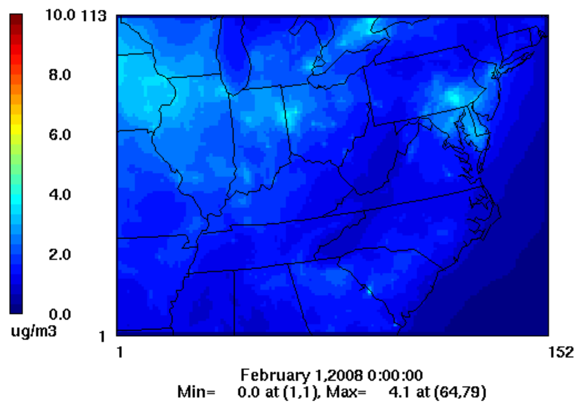
(a) Sulfate



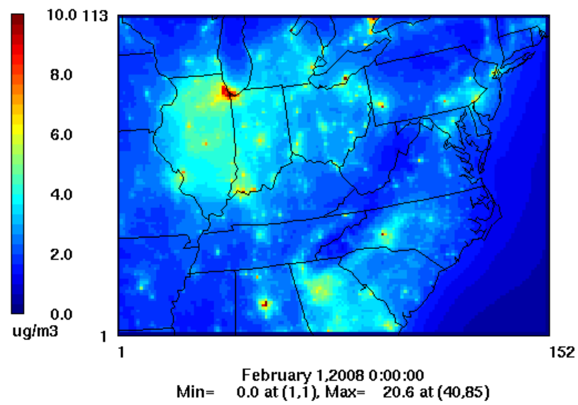
(b) Nitrate



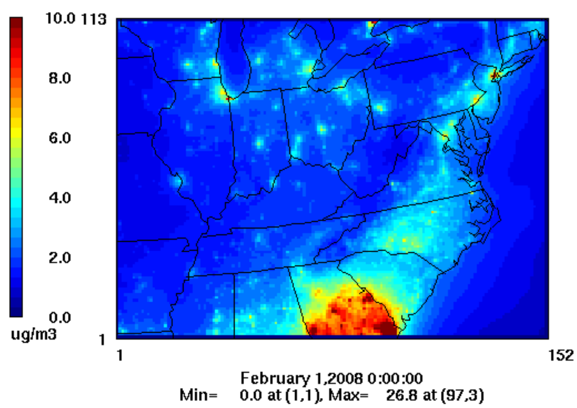
(c) Ammonium



(d) Soil



(e) Organic carbon



(f) Elemental carbon

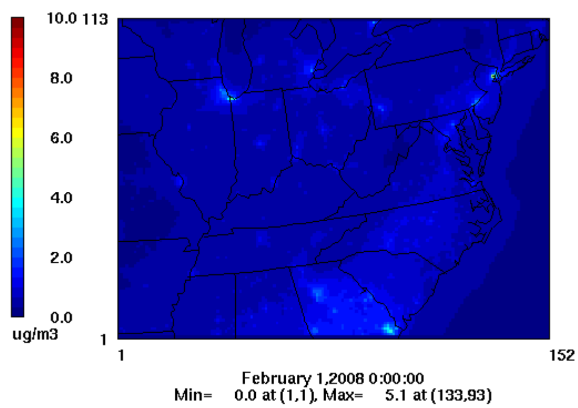


Fig. S8.1. Monthly mean concentrations of $PM_{2.5}$ components in the 12 km domain in February, 2008.

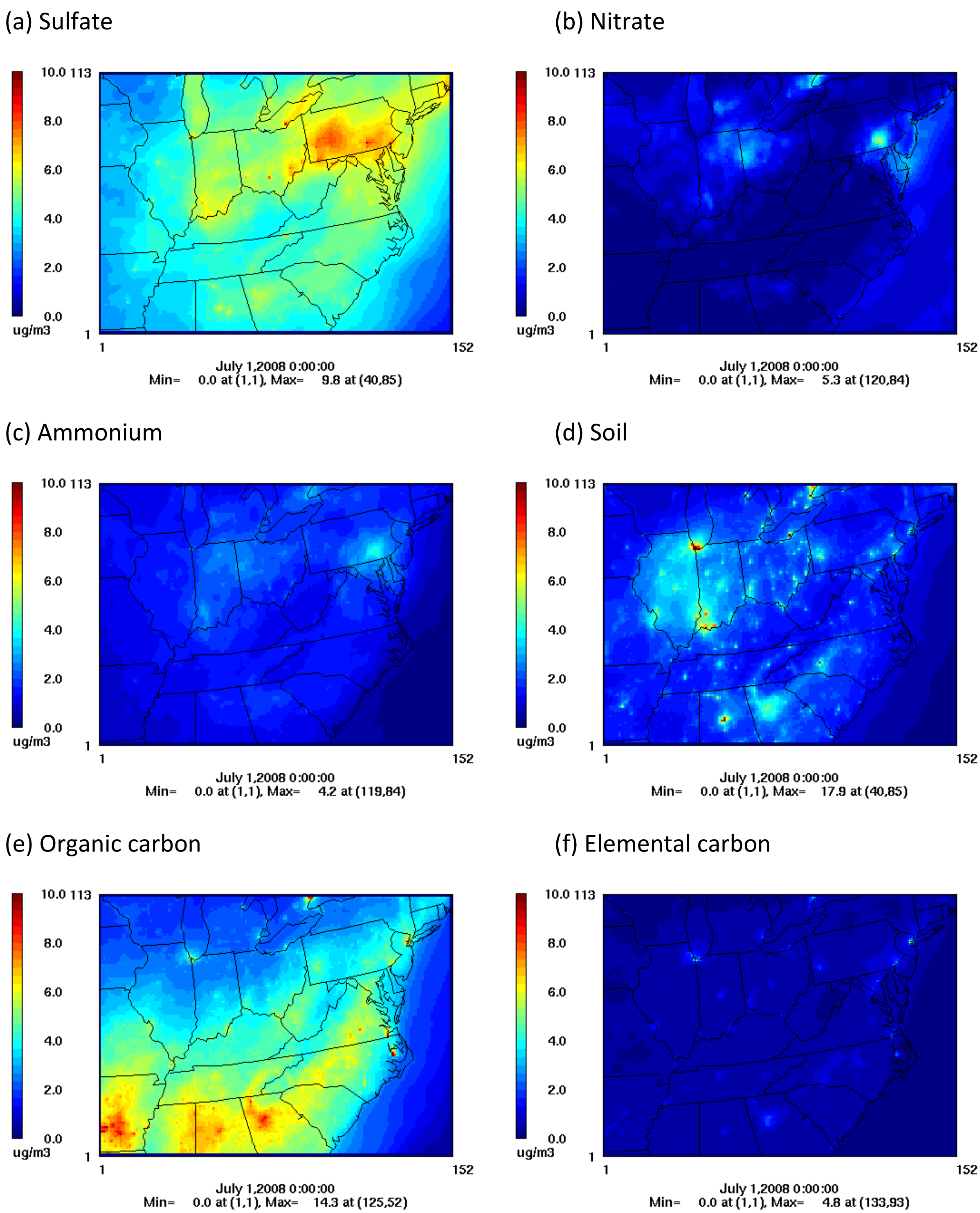


Fig. S8.2. Monthly mean concentrations of $PM_{2.5}$ components in the 12 km domain in July, 2008.

References for Supplementary Data

Baker, K., Scheff, P., 2007. Photochemical model performance for $PM_{2.5}$ sulfate, nitrate, ammonium, and precursor species SO_2, HNO_3, and NH_3 at background monitor locations in the central and eastern United States. Atmos. Environ. 41, 6185–6195.

Biswas, J., Hogrefe, C., Rao, S.T., Hao, W., Sistla, G., 2001. Evaluating the performance of regional-scale photochemical modeling systems: part III—precursor predictions. Atmos. Environ. 35, 6129–6149.

Boylan, J. W., Russell, A. G., 2006. PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. Atmos. Environ. 40, 4946-4959.

CASTNET, 2011. Clean Air Status and Trends Network, http://epa.gov/castnet/, accessed August 2011.

EPA. 1991. Guidance for Regulatory Application of the Urban Airshed Model (UAM). Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina. July. http://www.epa.gov/ttn/scram/guidance/guide/uamreg.pdf, accessed August 2011.

EPA, 2002. User Guide: Air Quality System. Report prepared by the U.S. Environmental Protection Agency, Research Triangle Park, North Carolina. April. http://www.epa.gov/ttn/airs/aqs/softw/AQSUserGuide_v1.pdf, accessed August 2011.

EPA, 2007. Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze. U.S. Environmental Protection Agency, Office of Air Quality and Planning Standards, Research Triangle Park, North Carolina. (EPA-454/B-07-002). April.

EPA, 2011. 2008 National Emissions Inventory: Data and Documentation. Clearinghouse for Inventories and Emissions Factors. http://www.epa.gov/ttnchie1/net/2008inventory.html, accessed July 2011.

Gaydos, T. M., Pinder, R. W., Koo, B., Fahey, K. M., Pandis, S. N., 2007. Development and Application of a Three-dimensional Aerosol Chemical Transport Model, (PMCAMx). Atmos. Environ. 41, 2594–2611.

Hu, J., Ying, Q., Chen, J., Mahmud, A., Zhao, Z., Chen, S.-H., Kleeman, M. J., 2010. Particulate Air Quality Model Predictions using Prognostic vs. Diagnostic Meteorology in Central California, Atmos. Environ., 44, 215–226.

IMPROVE, 1995. IMPROVE Data Guide. University of California Davis, August, 1995. http://vista.cira.colostate.edu/improve/Publications/OtherDocs/IMPROVEDataGuide/IMPROVEDataGuide.htm, accessed August 2011.

Liang, J.Y., Martien, P.T., Soong, S., Tanrikulu, S., 2004. A photochemical model comparison study: CAMx and CMAQ performance in central California. In: Proceedings of the Thirteenth Conference on the Application of Air Pollution Meteorology with the Air and Waste Management Association, Vancouver, Canada. American Meteorological Society, Boston. ams.confex.com/ams/pdfpapers/80097.pdf, accessed November 2011.

RDA, 2011. Research Data Archive (RDA). TDL U.S. and Canada Surface Hourly Observations, ds472.0 dataset. Maintained by the National Center for Atmospheric Research (NCAR). http://dss.ucar.edu/datasets/ds472.0/, accessed June 2011.

Russell, A.G., 2008. EPA supersites program-related emissions-based particulate matter modeling: initial applications and advances. J. Air Waste Manage. Assoc. 58, 289-302.

Seigneur, C., Pun, B., Pai, P., Louis, J.F., Solomon, P., Emery, C., Morris, R., Zahniser, M., Worsnop, D., Koutrakis, P., White, W., Tombach, I., 2000. Guidance for the performance evaluation of three-dimensional air quality modeling systems for particulate matter and visibility. J. Air Waste Manag. Assoc. 50, 588-599.

HIGHLIGHTS

- Simulations of the incremental benefits of successive US LDV emissions standards
- Tier 1, Tier 2, hypothetical nationwide LEV III standard and zero-out LDV scenario
- Calculated ozone and PM reductions assuming each standard is prevailing in 2022
- Tier 2 to LEV III switch offers negligible benefit compared to Tier 1 to 2 change
- Benefit of eliminating LDVs is smaller than the benefit from Tier 1 to 2 transition

SUPPLEMENTAL DATA FOR CRC ONLY

1. On-road motor vehicle emissions

Table C1 lists the on-road SCCs used in MOVES modeling

Table C1. List of on-road SCCs

SCC VehicleType	Description
2201001	Light Duty Gasoline Vehicles (LDGV)
2201020	Light Duty Gasoline Trucks 1 & 2
2201040	Light Duty Gasoline Trucks 3 and 4
2201070	Heavy Duty Gasoline Vehicles 2B thru 8B and Gasoline Buses
2201080	Motorcycles (MC)
2230001	Light Duty Diesel Vehicles (LDDV)
2230060	Light Duty Diesel Trucks 1 thru 4 (LDDT)
2230071	Heavy Duty Diesel Vehicles (HDDV) Class 2B
2230072	Heavy Duty Diesel Vehicles (HDDV) Class 3, 4, and 5
2230073	Heavy Duty Diesel Vehicles (HDDV) Class 6 and 7
2230074	Heavy Duty Diesel Vehicles (HDDV) Class 8A and 8B
2230075	Heavy Duty Diesel Buses (School and Transit)

PM emissions were speciated to CAMx model species, namely primary organic aerosol (POA), primary elemental carbon (PEC), primary nitrate (PNO3), primary sulfate (PSO4), primary others (FPRM), and coarse PM (CPRM; $PM_{2.5-10}$). The MOVES output provides total $PM_{2.5}$ and three components of $PM_{2.5}$: two pre-speciated components of $PM_{2.5}$ which are: 1) *PEC*, and 2) *PSO4*, and a non-speciated component termed "*PM25OM*", which is defined as the difference between total $PM_{2.5}$ and PEC. Steps to fully speciate the MOVES2010 partially-speciated EXHAUST $PM_{2.5}$ to create the model species needed for CAMx follow Baek and DenBleyker (2010) and are briefly described below.

Step 1. Assign PEC

$$PEC_{CAMx} = PEC_{MOVES} \qquad (1)$$

Step 2. Calculate PEC_72, mass of Primary elemental carbon when MOVES is run at 72 °F or higher temperature; calculated by backing out the temperature adjustment factor, PEC_Tadj

For gasoline vehicles, MOVES applies a temperature adjustment factor that accounts for the impact of cold temperatures on PM25OM and PM25EC with decreasing temperature at temperatures below 72 °F. At 72 °F or higher, there is no dependency of any component of $PM_{2.5}$ on temperature. There is also no dependency of any component of $PM_{2.5}$ on temperature for diesel vehicles. At temperatures lower than 72 °F, the temperature dependence is different for start emissions (including crankcase starts) versus running emissions (including crankcase running). , the unadjusted PEC is needed to compute the components of $PM_{2.5}$ that are not impacted by temperature. The unadjusted PEC is denoted as PEC_72.

PEC_72 = PEC / PEC_Tadj (2)

For start emissions, PEC_Tadj = 28.039*exp(-0.0463*T) (3.a)

For running emissions, PEC_Tadj = 9.871*exp(-0.0318*T) (3.b)

For simplicity, the mean daily temperature for each county for winter and summer months was used in equation (3.a) and (3.b).

Step 3. Assign PSO4

$PSO4_{CAMx} = PSO4_{MOVES}$ (4)

Step 4. Calculate PNO3 and metal

The primary nitrate is computed based on the ratio of nitrate to elemental carbon in the speciation profile, i.e., F_{NO3} / F_{EC} and metals component from the ratio of metals to elemental carbon, F_{METAL} / F_{EC} using equations (5) and (6), respectively.

$PNO3 = PEC\_72 \times F_{NO3} / F_{EC}$ (5)

$METAL = PEC\_72 \times F_{METAL} / F_{EC}$ (6)

Table 2 shows the values for the above fractions and the profiles from which they are to be derived.

Table C2. Values and basis for fractions used to compute PNO3 and METAL

Vehicle Type	**SCC list**	*FEC (%)*	*FNO3 (%)*	*FMETAL (%)*
LDDV	All SCCs that begin with: 2230001, 2230002, 2230003, 2230004, 2230005, 2230006	57.48051203	0.23	0.6513
HDDV	All SCCs that begin with: 223007	77.1241	0.1141	0.2757
LDGV and HDGV	All SCCs that begin with 2201	20.80113619	0.1015	2.2256

Step 5. Calculate PNH4 based on stoichiometric calculations

$PNH4 = (PNO3/MW_{NO3} + 2 \times PSO4/MW_{SO4}) \times MW_{NH4}$ (7)

Step 6. Calculate POA and FPRM

POA = PM25OM – METAL – PNH4 – PNO3 (8)

FPRM = METAL + PNH4 (9)

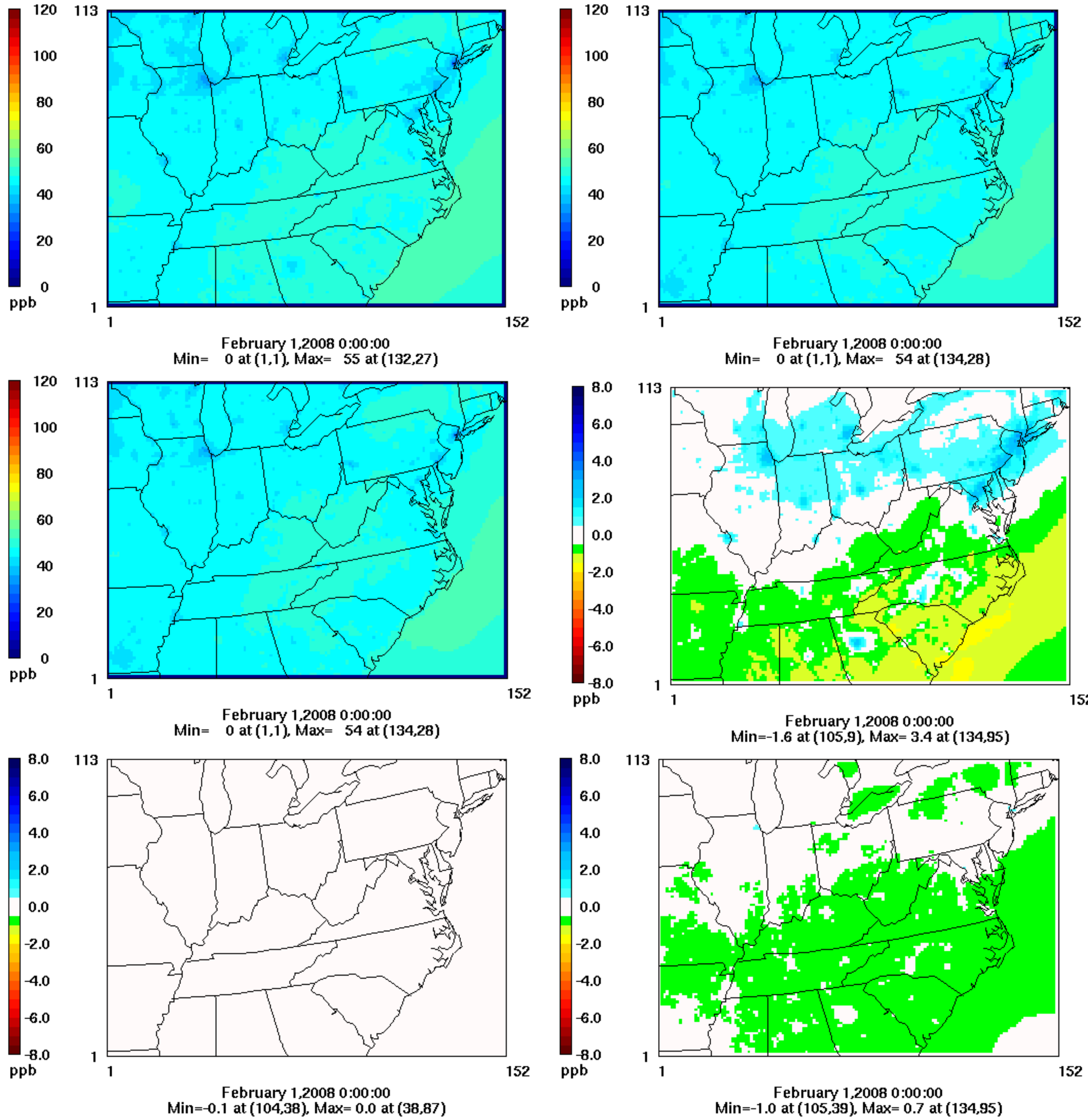


Fig. C1. Monthly mean and differences in monthly mean of daily maximum 8-hr ozone concentrations in February in 2022 scenarios: Tier1 (top left), Tier2 (top right), LEV III (center left), Tier 2 – Tier 1 (center right), LEV III – Tier 2 (bottom left), and zero-out LDV – Tier 2 (bottom right).