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Effects of light duty gasoline vehicle emission standards in the United States on ozone and particulate matter

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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 very small benefit compared to Tier 1 to 2 change.

► Benefit of eliminating LDVs is smaller than the benefit from Tier 1 to 2 transition.

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ABSTRACT

More stringent motor vehicle emission standards are being considered in the United States to attain national air quality standards for ozone and PM2.5. 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 PM2.5 (up to 16 ppb (14%) reductions in daily maximum 8-h ozone, up to 10 ppb (11%) reductions in the monthly mean of daily maximum 8-h ozone, up to 4.5 μ g m⁻³ (9%) reductions in maximum 24-h PM_{2.5} and up to 2.1 μ g m⁻³ (10%) 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 (0.3%) reductions in daily maximum 8-h ozone, up to 0.2 ppb (0.2%) reductions in the monthly mean of daily maximum 8-h ozone, up to 0.1 μ g m⁻³ (0.5%) reductions in maximum 24-h PM_{2.5} and up to 0.1 μ g m⁻³ (0.5%) 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 (8%) reductions in daily maximum 8-h ozone, up to 4 ppb (6%) reductions in the monthly mean of daily maximum 8-h ozone, up to 2.8 μ g m⁻³ (7%) reductions in maximum 24-h PM_{2.5} and up to 1.8 μ g m⁻³ (8%) 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.

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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)

* Corresponding author. Tel.: +1 415 899 0700. E-mail address: krish@environcorp.com (K. Vijayaraghavan). 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 (O₃) and fine particulate matter (PM_{2.5}). With the potential lowering of the National Ambient Air Quality Standards (NAAQS) for 8-h O₃ and PM_{2.5}, States would likely seek additional means to reach or stay in O₃ and PM attainment including possibly adopting more severe LDV emission standards.



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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 O₃ and/or PM₂₅ concentrations and the impact of vehicle fuel and emissions controls on these concentrations (e.g., EPA. 1999: Matthes 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 O₃ 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 with a focus on Atlanta, Detroit, Philadelphia and St. Louis. 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) and parts of Canada and Mexico 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)

2022 was chosen as the future year for modeling because the proposed LEV III standard was originally scheduled to phase in completely by 2022 (this was subsequently revised to 2028 as discussed below). All simulations were conducted for a winter month (February) and summer month (July) to represent two time periods with typically high PM_{2.5} and ozone concentrations.

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.



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

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 2015–2028. 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. 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).

Canadian on-road emissions for the 2008 and 2022 scenarios within the 36 km grid were derived from the 2005 NEI and 2020 NEI, respectively (see below). Mexican on-road emissions within the 36 km grid for all scenarios were derived from the 2005 NEI and based on 2000 emissions.

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 within the 36 km grid were prepared from and set equal to emissions in the 2005 NEI (EPA, 2011c) and the 2020 NEI, respectively. Anthropogenic area and point emissions for Mexico within the 36 km grid 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. Emissions outside the 36 km grid are treated through the boundary conditions (see below).

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, SO₂, NH₃ 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; 2008).

2.5. Other model inputs

Boundary concentrations of O₃, 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° for both latitude and longitude and 28 vertical layers were mapped onto the CAMx domain and speciated for the CB05 chemical mechanism. The boundary conditions for the 36 km domain were kept constant across all scenarios. Boundary conditions for the 12 km domain are calculated within CAMx from the 36 km grid calculations in each scenario.

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 onroad 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 SO₂ (<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 NH₃ emissions increase more than two-fold from winter to summer. This is due, in part, to higher dairy NH₃ 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%-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 $\sim 20\%$ of total anthropogenic emissions in 2008

and, hence, are important to studying the potential contribution of LDVs to ambient O₃ and PM_{2.5}. Due to their slow reactivity, CO emissions have a much more limited effect on O₃ 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 NH₃ and SO₂ 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, MOVES default age distributions were used for St. Louis while local data on vehicle age distributions were used for other three urban areas; this likely introduced uncertainty in our estimates for St. Louis. For example, we determined that using local age distributions for Atlanta, Detroit and Philadelphia resulted in modeled VOC emissions that were approximately 10% lower than if we had used MOVES default age distributions (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-h 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-h 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-h 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. Figs. 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 SO₂ 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 µg 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.



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

Table 1			
		-	

Average-day emissions from	n gasoline-fueled LDVs in the	e continental US in model scenarios.
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	Winter						Summer					
	NOx	VOC	PM _{2.5}	CO	$\rm NH_3$	SO ₂	NOx	VOC	PM _{2.5}	CO	$\rm NH_3$	SO ₂
2008												
LDV emissions (Mg day ⁻¹)	8380	7039	242	92,610	272	79	9661	6384	123	68,064	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 $^{-1}$)	10,422	6127	250	101,288	457	48	12,047	5319	131	73,217	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 $^{-1}$)	2609	2269	188	69,576	166	41	2897	2247	108	40,745	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 $^{-1}$)	2505	2134	173	65,152	166	41	2781	2098	103	38,120	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%

The 2008 CAMx base case predictions of 1-h and 8-h 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-h) 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 Figs. 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

Table 2

Average-day emissions from	ı gasoline-fueled L	LDVs in four urb	an areas in 2008.
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Urban area	Pollutant	February 2008		July 2008		
		Emissions (Mg day ⁻¹)	% of all on-road	Emissions (Mg day ⁻¹)	% 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%	

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. Changes in LDV emissions between the 2008 scenario and the 2022 Tier 1 scenario are due to several factors including an approximately 20% increase in nationwide VMT from 2008 to 2022, changes in fleet composition and lower gasoline sulfur in 2022 compared to 2008 (MOVES default nationwide average gasoline levels in 2008 and 2022 are 42 and 21 ppm, respectively). 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.



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

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-h O_3 concentrations and differences in these monthly means between pairs of 2022 LDV scenarios. The same quantities are listed in Table S9.1 (in Supplementary data) for the four urban areas. Also shown in this table are the monthly maximum 8-h 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-h 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-h O_3 ranges from 57 ppb at Detroit to 78 ppb at Philadelphia and the highest 8-h 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-h 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-h O₃ maxima decreasing by 9 ppb (11%) from Tier 1 to Tier 2 and the monthly highest O₃ decreasing by 16 ppb (14%) from Tier 1 to Tier 2. When compared to Atlanta and Philadelphia, Detroit shows a small benefit (3-4 ppb) for the monthly mean of daily maximum 8-h O₃ and the monthly highest 8-h O₃. St. Louis shows a reduction of 5 ppb in the monthly mean but a smaller reduction (2 ppb) in the monthly highest 8-h O₃ despite large reductions in NOx (74%) and VOCs (58%) from the Tier 1 to Tier 2 scenarios, suggesting that the highest 8-h 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-h O₃ from the Tier 1 to Tier 2 scenarios. The increases in ozone in these urban areas despite reductions in LDV NOx emissions from the Tier 1 scenario suggest that NOx that was otherwise titrating ozone becomes unavailable due to the Tier 2 LDV emissions reductions.

The monthly mean of daily maximum 8-h O₃ 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-h O₃ is reduced by 0.1–0.3 ppb (0.1–0.3%) (see Table S9.1). 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



Fig. 4. Monthly mean concentrations of PM2.5 in the 36 km domain (left) and 12 km domain (right) in February (top) and July (bottom), 2008.

achievable by 2022. We expect some 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-h maximum ozone and 3–7 ppb (3–8%) in the highest 8-h ozone below 2022 Tier 2 levels in the four urban areas. The maximum reduction in the monthly mean of daily 8-h 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 S9.1 shows similar information for monthly mean PM_{2.5} and monthly maximum 24-h PM_{2.5} in the four urban areas. Table S9.2 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 μ 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 μ g m⁻³ from Tier 1 to Tier 2 levels in broad swaths across the eastern US and by 2 μ g m⁻³ (~10%) in large urban areas such as Chicago,

Washington D.C., Detroit, Philadelphia and New York. Among the four urban areas analyzed here, the Tier 1 wintertime mean PM_{2.5} concentration ranges from 14 μ g m⁻³ in Atlanta to 19 μ g m⁻³ in Philadelphia, and the maximum 24-h PM_{2.5} ranges from 22 μ g m⁻³ in Atlanta to 48 μ g m⁻³ in Philadelphia (see Table S9.1). 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-h PM_{2.5} reduced by 4.5 μ 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 S9.2). 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 μ g m⁻³ (6%) from Tier 1 levels and maximum 24-h PM_{2.5} reduced by 1.5 μ 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 μ g m⁻³ reduction in monthly mean) than winter (Table S9.2).

Switching from the Tier 2 to LEV III results in less than 0.1 μ 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 μ g m⁻³ (0.5%) reduction in monthly maximum 24-h PM_{2.5} in the four urban



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

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Emissions from gasoline-fueled LDVs in four urban areas in 2022 LDV emissions scenarios.^a

Month	Area	Pollutant	LDV Tier 1 emissions (Mg day ⁻¹)	LDV Tier 2 emissions (Mg day ⁻¹)	LDV LEV III emissions (Mg day ⁻¹)	%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	0.8	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.

areas (see Table S9.1). 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 μ g m⁻³ (3–11%) reductions in the monthly mean PM_{2.5} and 0.3–2.9 μ g m⁻³ (2–7%) in the monthly maximum 24-h 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 μ 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.



Fig. 6. Monthly mean and differences in monthly mean of daily maximum 8-h ozone concentrations in July in 2022 scenarios: Tier 1 (top left), Tier 2 (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: Tier 1 (top left), Tier 2 (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).

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



Fig. 8. Monthly mean and differences in monthly mean of hourly PM_{2.5} concentrations in July in 2022 scenarios: Tier 1 (top left), Tier 2 (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).

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 incomplete 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. It would also be useful to compare the relative contributions of other sources to ozone and PM compared to LDVs.

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Appendix A. Supplementary data

Additional emissions data, model results and model performance evaluation can be found in the online version, at http://dx. doi.org/10.1016/j.atmosenv.2012.05.049.

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