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PERSPECTIVE OPEN

Vehicle criteria pollutant (PM, NO_x, CO, HCs) emissions: how low should we go?

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Over the past 30–40 years, vehicle tailpipe emissions of particulate matter (PM), nitrogen oxides (NO_x), carbon monoxide (CO), and hydrocarbons (HCs) have decreased significantly. Advanced emission after-treatment technologies have been developed for gasoline and diesel vehicles to meet increasingly stringent regulations, yielding absolute emission reductions from the fleet despite increased vehicle travel. As a result of mobile and stationary source emission controls, air quality has generally improved substantially in cities across the US and Europe. Emission regulations (such as Tier 3 in the US, LEV III in California, and Euro 6 rules in the EU) will lead to even lower vehicle emissions and further improvements in air quality. We review historical vehicle emission and air quality trends, discuss the future outlook for air quality, and note that modern internal combustion engine vehicles typically have lower exhaust emissions than battery electric vehicle upstream emissions. As vehicle manufacturers and city officials grapple with questions about future mobility in cities, we raise the question "how low should we go?" for future vehicle criteria emissions. The answer to this question will have profound implications for automotive and fuel companies and for the future economic and environmental health of urban areas.

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INTRODUCTION

Vehicle emissions affect local and regional air quality. The criteria pollutant emissions generated from fuel combustion by internal combustion engines (ICE) include nitrogen oxides (NO and NO_2, together called NO_x), hydrocarbons (HC)—also known as volatile organic compounds (VOCs) or non-methane hydrocarbons (NMHC), carbon monoxide (CO), and particulate matter of size <10 microns (PM_{10}) and <2.5 microns ($PM_{2.5}$) including black carbon (BC). Governments worldwide regulate vehicle emissions of criteria pollutants.[1] The US and EU began regulating emissions in the 1970s. Currently in the US the federal Tier 3 and California LEV III light-duty vehicle (LDV) standards phase-in from 2015 to 2025. In the EU, the current Euro 6 LDV standard went into effect in 2015, while real driving emissions (RDE) standards phase-in beginning in 2017. Heavy duty vehicle (HDV) emission standards include the Euro VI standards (we adopt the convention of using Roman numerals and Arabic numerals for heavy- and light-duty standards, respectively), effective in 2013, and the current US HD standards, fully phased-in as of 2010. Other regions' vehicle emission standards are usually modified versions of the Euro or US standards (Euro 3-Euro 6 and Tier 1-Tier 3).[1]

The successive LDV emission standards have lowered the regulated emission intensity (g/mile or g/km) of $NMHC + NO_x$ emissions by 97% in the US (Tier 1–3 in 2025) and by 80–85% in the EU (Euro 1–6),[1] and of total hydrocarbons $(THC) + NO_x$ in China by 84% (China 1–5).[2] Likewise, HDV emission standards for $NMHC + NO_x$ have decreased by 95% or more since 1988 in the US.[1] To meet these standards, advanced vehicle emission after-treatment systems such as three-way catalytic converters, lean NO_x traps, selective catalytic reduction (SCR), and diesel particulate filters (DPFs) have been developed and implemented.[3–7]

This paper focuses on criteria pollutants. Emissions from ICE vehicles also include carbon dioxide (CO_2), a greenhouse gas that is the main combustion product of carbon-based fuels. Vehicle CO_2 emissions are regulated around the world and are reduced by improving the fuel efficiency of the vehicles.

As a result of regulations and technology improvements, emissions from vehicles and stationary sources have decreased and air quality has improved significantly in most large cities in the US and Europe. The following sections review historical vehicle emission and air quality trends, examine the outlook for air quality, and discuss the future of vehicle regulations.

VEHICLE EMISSION TRENDS

Vehicle emission intensities have decreased by factors of 2–100 over the past several decades to meet regulatory standards.[1,8] The regulatory test cycle emissions are generally achieved in US on-road operation. On-road measurements show substantial declines in real-world emissions of HC, NO_x, and CO at four U.S. urban locations since the late 1990s.[9–11] In contrast, in the EU the New European Drive Cycle (NEDC) and test procedure did not fully reflect on-road emissions, and the decrease in NO_x emissions from diesel vehicles in the real world has been less than expected based on emission standards as tested under laboratory conditions. Diesel LDV on-road NO_x emission rates in the EU have not improved relative to the 1990s and the measured NO_2 share of NO_x has increased.[12] However, gasoline vehicle on-road NO_x emission rates have decreased by a factor of 8–10 since pre-Euro 1 emission controls.[12] In 2017, the severity of Euro 6 regulations increased substantially by including an RDE component to bring on-road diesel NO_x close to the laboratory standard.[13,14] The

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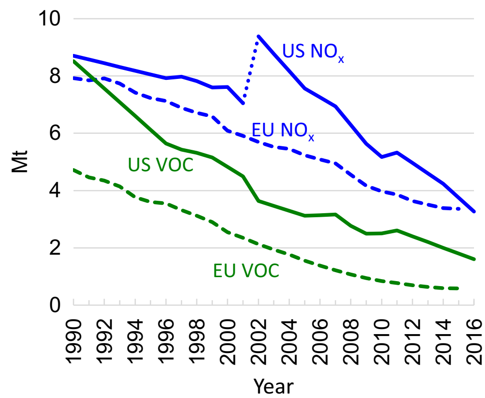

Fig. 1 Highway vehicle (LDV, HDV, commercial vehicle and motorcycle) NO_x (blue) and VOC (green) emissions (Mt) for 1990–2016 for U.S. (solid lines) and EU (dashed lines).[18,19] (The increase in 2002 U.S. NO_x is due to an EPA modeling methodology change.)

future China 6 vehicle emission regulations also include RDE testing modeled after the EU.[1]

As the vehicle emission intensity has declined, vehicle population and travel demand has increased. In the US, LDV distance traveled grew 180% between 1970 and 2015, and 50% between 1990 and 2015.[15] The number of registered passenger cars in the EU grew about 40% from 1995 to 2015 and passenger car travel (passenger km) increased 21% over the same time.[16] Since 1985 the number of vehicles in China has increased by a factor of 20.[17]

Nonetheless, total highway vehicle (LDV, HDV, commercial vehicle and motorcycle) NO_x and VOC emissions in the US and EU have declined 60–80% since 1990 as shown in Fig. 1.[18,19] Edwards et al.[20] modeled real-world LDV and HDV emissions for the years 2000–2030 following the implementation of Tier 3 and Euro 6 RDE regulations. Future emissions of most pollutants in most regions are projected to decline significantly following the introduction of these regulations, even without electric vehicles and with increased travel overall. The exception is in China where the travel demand increases by a factor of 10, partially offsetting the vehicle emission intensity improvements. Wu et al.'s modeling[2] shows that while the China passenger LDV stock more than doubles from 2015 to 2030, total vehicle emissions decline as China 6 regulations take effect and fleet electrification increases.

AIR QUALITY TRENDS

Air quality has improved steadily as vehicle and stationary source emissions have decreased. In the US, ambient concentrations of ozone (O_3), NO_x, PM, CO and other pollutants are declining.[21] US annual O_3 concentrations (4th highest 8-h average) have decreased 31% from 1980 to 2016 as shown in Fig. 2a,[22] and $PM_{2.5}$ (annual average) has decreased 42% since 2000. California peak O_3 concentrations have decreased by approximately a factor of 5 since the 1960s.[23]

In Europe, O_3 precursor emissions (NO_x and VOCs) are declining.[24] Figure 2b shows that EU region-wide average O_3 concentrations have been stable since 2000,[25] somewhat lower than the standard of 120 $\mu g/m^3$ (0.06 ppm).[26] NO_2 and $PM_{2.5}$ annual mean urban concentrations also have decreased since 2000.[27]

Studies in China analyzing data from a nationwide O_3 monitoring system implemented in 2012 show an increase in observed ozone between 2013 and 2015,[28,29] although decreasing emissions of NO_x, which can remove O_3 through chemical reactions, may have contributed to the ozone increase. Annual average $PM_{2.5}$ concentrations decreased 33–45% between 2013 and 2017 in the Beijing–Tianjin–Hebei area but still exceed the national standard.[30] In 2014–2015, 190 cities in China averaged annual $PM_{2.5}$ concentrations of $57 \pm 18\ \mu g/m^3$,[31] greater than the national standard of 35 $\mu g/m^3$, but source apportionment models indicate transportation accounts for <10% of $PM_{2.5}$ annual average concentrations in China.[32,33]

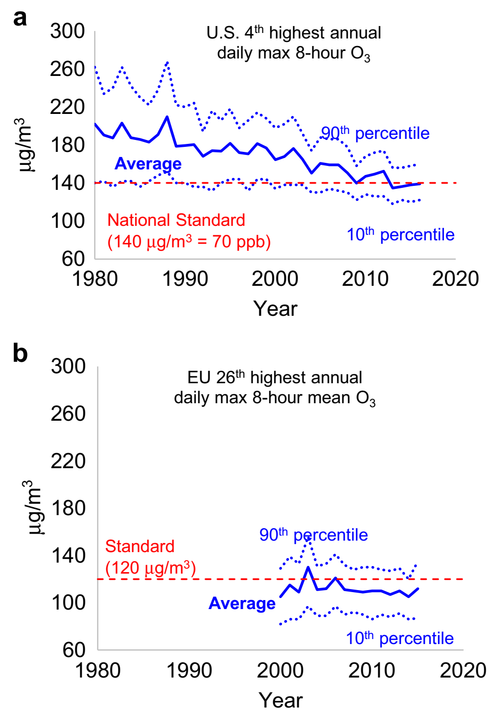

Fig. 2 U.S. (**a**) and EU (**b**) regional average ozone measured concentrations (N^{th} highest daily maximum 8-h average) (blue) compared to the ambient air quality standards (red).[22,24] (1 ppb O_3 $= 2\ \mu g/m^3$ O_3)

While country-wide average air quality today is much improved in the US and EU, local concentrations can still exceed the ambient air quality standards. In many areas of the US, especially in California, O_3 concentrations continue to exceed the air quality standard (0.07 ppm=140 $\mu g/m^3$), despite the large decreases in emissions and ambient concentrations of VOC and NO_x.[34,35] Likewise, many European monitoring stations exceed the O_3 standard (120 $\mu g/m^3$ on >25 days per year)[36] Furthermore, the EU annual air quality standard for NO_2 is still widely exceeded, particularly at roadside monitoring stations.[37]

ZERO-EMISSION VEHICLES

Because of local exceedances of air quality standards, vehicle emission programs beyond the recently implemented Tier 3 and Euro 6 RDE standards are being considered. Limits or bans on ICE vehicles in several major city centers are being implemented to further control emissions.[38] Countries are considering bans on the sale of gasoline and diesel vehicles beginning in 2025 or later.[39] Zero-emission vehicle (ZEV) mandates are in place in California, 11 other US states, and China. All these programs require ZEVs, which can be pure battery electric vehicles (BEV) or hydrogen fuel cell vehicles (FCV). Plug-in hybrid electric vehicles (PHEV) may count as transitional ZEVs.

ZEVs do not produce tailpipe emissions, thus reducing roadside emissions and improving local air quality. However, ZEVs are not zero-emission in a regional or global sense because electricity (or hydrogen) generation can produce upstream emissions. Tailpipe emissions from an ICE vehicle can be comparable to the upstream

Table 1. Comparison of US and EU vehicle emission standards with emissions from selected gasoline ICE vehicles and BEVs, and non-exhaust brake wear and tire wear emissions

	mg/km				
	$PM_{2.5}$	NO_x + HC	NO_x	SO_2	CO
Vehicle standards (test cycle)					
US Tier 3	2	53		0.6[a]	1057
Euro 6 (gasoline)	0.3[b]	170	60		500
Euro 6 (diesel)	0.3[b]		80		
US 2017 ICE					
Best-in-class (HEV) (test cycle)[42]	0.06[c]	2	0.3		31
2016 fleet average[d] (on-road)[10]		66	28		231
EU ICE					
Average Euro 6 gasoline DI ICE (RDE)[43,44]	0.2–0.4[b]		12–20		17–100
Typical 2017 BEV electricity emissions					
2014 US elec. grid[45–47]	7	71	70	123	
2016 US elec. grid[41]			37	41	
2030 US elec. grid[41]			30	32	
Brake and Tire Wear[50–52]					
Brake wear	2–6				
Tire wear	1–5 ($PM_{2.5}$)				
	4–13 (PM_{10})				

[a]Based on 5 mg S/kg fuel, fully converted to SO_2 during combustion, 8 L/100 km (29.4 miles per gallon)
[b]Based on particle number standard of 6×10^{11} #/km; 2×10^{12} #/km equals 1 mg/km. PM mass standard is 4.5 mg/km
[c]Total PM
[d]Chicago, IL area; assumed 22 miles per gallon gasoline [FHWA Highway Statistics 2016, Table VM-1]

electricity generation emissions. For example, a typical US 2017 BEV has a label electricity consumption of ~25 kWh/100 miles.[40] The US electric grid, on average, produces 0.224 g NO_x/kWh of generated electricity.[41] Assuming 7% grid loss, the BEV produces upstream NO_x emissions of 0.06 g/mile, somewhat less than the vehicle standard of 0.086 g NO_x + NMHC/mile. The best-in-class ICE vehicle, an HEV, emits 0.004 g NO_x + HC/mile,[42] ten times less than the BEV. Table 1 compares the emissions of criteria pollutants for the BEV in the US, best-in-class U.S. ICE, U.S. average on-road fleet,[10] typical Euro 6 gasoline direct injection (DI) ICE RDE models,[43,44] and the EU passenger car and the US LDV standards. Ranking by total NO_x emissions per km, new vehicle ICE emissions are less than BEV emissions, and both are below the new vehicle standards. The estimated emission intensity of the 2016 US on-road (ICE) fleet[10] shown in Table 1 is comparable to the new vehicle standards and less than BEV total emissions. BEV-related NO_x and SO_2 emissions in the US are expected to improve over time[41,45–47] as power generation regulations take effect, including the Cross-State Air Pollution Rule.

As vehicle exhaust emissions have decreased, non-exhaust emissions have become relatively more important.[48,49] In particular, PM emitted from tire and brake wear is now comparable to exhaust emissions, as shown in Table 1,[50–52] but BEVs and ICE vehicles can have different emission levels. Tire wear is a function of many factors: heavier BEVs are expected to give more tire wear PM emissions while brake wear PM emissions can be lower on electrified vehicles, which use regenerative braking. Timmers and Achten[53] estimate the total PM emissions (exhaust + non-exhaust, including road wear and resuspension) are similar for ICE vehicles and BEVs. For windshield washer fluid VOC emissions, we assume ICE vehicles and BEVs emit similar amounts because this function is independent of powertrain and fuel.

FUTURE AIR QUALITY

Air quality models provide insights into the outcomes of future vehicle emission regulations and programs. Modeling for the US shows that when the Tier 3 standards are fully phased-in to the on-road fleet (2030), ozone will be reduced in most areas, but exceedances of the standard can occur.[54] The same study also shows diminishing returns of successive vehicle emission regulations. Peak ozone declined by 1–13% from 2008 to 2018 (Tier 1–2) but is only expected to decrease by 1–4% from 2018 to 2030 (Tier 2–3).

European assessments also project substantial air quality improvements. Street canyon modeling in Germany shows significant reduction in local NO_2 concentrations as a result of fleet turnover with replacement by Euro 6 RDE-compliant vehicles. By 2030 only 1% of roadside air monitors exceed the NO_2 standard compared with 49% in 2015.[55] Recognizing that the NO_2/NO_x ratio of diesel vehicle exhaust is smaller than previously assumed, Grange et al. conclude NO_2 may improve faster at roadside monitors than anticipated in current emission inventories.[56]

The ongoing reductions for LDV emissions mean other sectors have increasingly greater leverage. Nopmongcol et al.[57] modeled the effect of electrification of LDVs, HDVs and off-road equipment in the US in 2030. When 8–17% of vehicle travel is electrified, emission reductions lead to modest decreases in ozone and $PM_{2.5}$ throughout the country. Electrification of off-road equipment (garden equipment, construction equipment) would provide more air quality improvement than on-road electrification. McDonald et al.[58] showed that consumer products (adhesives, personal care products, etc.) are becoming the single largest source of petrochemical VOC emissions in industrialized cities.

CONCLUSIONS

Vehicle emission reductions and improvement in air quality in the US and EU have been impressive over the last three decades, despite growth of the LDV fleet. As other regions implement the latest US and European emission regulations, similar results can be expected. Regulations and auto manufacturer plans include increased vehicle electrification, further reducing local emissions. With continued improvement in air quality following vehicle emission reductions, it is time to consider the long-term outlook for future vehicle regulations.

Data and observations indicate ICE vehicle emissions may be approaching a ZEV-equivalent level. Modeling shows that successively more stringent vehicle regulations provide diminishing air quality benefit. As the vehicle sector emits a smaller share of the total emissions, other emission-reduction strategies can be more cost-effective in improving air quality. For example, reducing emissions from non-vehicle sources (power generation, house heating, off-road equipment) will yield a greater impact on air quality. Future vehicle emission-reduction efforts might be more profitably targeted on reducing the effect of gross emitters, which represents 2–5% of the fleet but can produce up to half the emissions.[59] PHEVs may be used in electric mode where ICE bans are present, offering a solution for commercial, medium-, and heavy-duty vehicles whose duty cycles are not amenable to a fully electric platform. Going forward it will be important to have a more holistic view of emission sources and to assess the most cost-effective actions to achieve the desired air quality improvements.

With the continued reduction in fleet vehicle emissions and anticipated air quality improvement, the key question for industry, regulators, air quality experts, health impact researchers, city planners, and society at large is "How low should we go?"

Data availability

All data are publicly available and cited in the references.

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AUTHOR CONTRIBUTIONS

T.J.W. and W.C.R. developed the concept; S.L.W. conducted analyses and wrote the paper; J.E.A. and R.V. provided technical reviews; L.G. coordinated the project. All authors edited the manuscript.

ADDITIONAL INFORMATION

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