NO_x emissions from diesel passenger cars worsen with age

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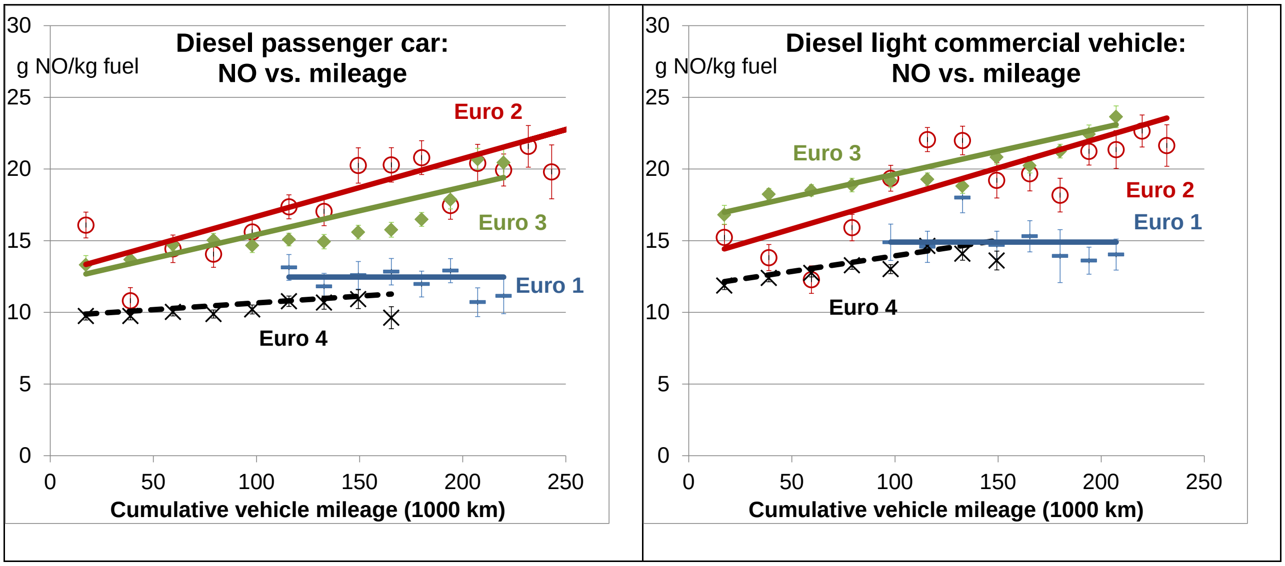
Abstract

Commonly, the NO_x emissions rates of diesel vehicles have been assumed to remain stable over the vehicle's lifetime. However, there have been hardly any representative long-term emission measurements. Here we present real-driving emissions of diesel cars and light commercial vehicles sampled on-road over fifteen years in Zurich/Switzerland. Results suggest deterioration of NO_x unit emissions for Euro 2 and Euro 3 diesel technologies, while Euro 1 and Euro 4 technologies seem to be stable. We can exclude a significant influence of high-emitting vehicles. NO_x emissions from all cars and light commercial vehicles in European emission inventories increase by 5% to 10% accounting for the observed deterioration, depending on the country and its share of diesel cars. We suggest monitoring the stability of emission controls particularly for high-mileage light commercial as well as heavy-duty vehicles.



Introduction

Vehicle exhaust emissions tend to increase with the vehicle's age due to deterioration of the engine controls, the catalyst and potentially the particle filter. To contain the adverse effects on health and the environment, legislation requires a specific durability for a minimum mileage or lifetime. For light-duty as well as heavy-duty diesel vehicles however it is assumed that their NO_x emissions do not deteriorate: The emission regulations in the European Union require zero deterioration of NO_x (or NO_x + NMHC) unit emissions over 80'000 km for Euro 1, 2 and 3 light-duty diesel vehicles (model years 1992 to 2004), over 100'000 km for Euro 4 (model years 2005 to 2009) and over 160'000 km for Euro 5 (model years 2010ff). The maximum allowed NO_x deterioration for Euro IV heavy-duty vehicles is 5% over 200'000 or 500'000 km[1]. Likewise, Europe's standard vehicle emission models all assume zero deterioration of NOx exhaust emissions from light duty diesel vehicles[2,3]. As diesel vehicles are one of the biggest sources of NOx emissions this assumption is important for air quality[4] However, the experimental basis for the zero deterioration assumption has been surprisingly limited: Europe's databases have relied on only 3 chassis dynamometer tests for Euro 1 diesel cars, all with mileages above 110'000 km, on 28 chassis tests for Euro 2 diesel cars, but only eight of which with mileages above 80'000 km, and 15 chassis tests from diesel Euro 3 cars, of which only one car had a mileage larger than 50'000 km[3]. Moreover, there have been virtually no experimental deterioration data for light commercial vehicles (LCV); it was simply assumed that they behaved analogous to diesel passenger cars. Given that high on-road NOx emissions from the strongly increased numbers of diesel cars have become a major problem for air quality in Europe[5], there is an urgent need for a statistically reliable basis for the zero deterioration assumption. Moreover, the legislative laboratory test cycle fails to reflect real-driving emissions - hence real-world data are needed, covering higher mileages, and including more modern car technologies as well as light commercial vehicles.

In this paper we discuss the long-term emission behavior of diesel cars as well as LCVs. We analysed a sample of 62'000 records of real-driving emissions from fifteen years of consecutive annual measurements. This unique dataset from Zurich/Switzerland covers vehicles up to 21 years old. This allows answering the question of the long-term stability of the diesel emission control system. We focus on the NOx emission rates due to their importance for air quality.

Experimental and data treatment

We analyze about 44'000 records of hot on-road emissions from diesel passenger car and 18'000 records from light commercial vehicles collected over fifteen years of successive annual measurements in Switzerland (see Table S1 in SI for a summary). The majority of the measured vehicles are up to 15 years old, which corresponds to a cumulative mileage of about 240'000 kilometers[6]. Our records are representative for cars certified to Euro norms 1 to 4. These norms became mandatory in Europe in years 1992, 1996, 2000, and 2005 respectively for diesel passenger cars, and 1994, 1998, 2000/2001 (depending on the weight class), and 2005/2006 for diesel light commercial vehicles. No deterioration analysis is done here for Euro 5 diesel cars and LCVs because most of those vehicles are under 5 years old which is not long enough for time series analysis. Exhaust emissions were measured in real-driving by remote sensing. This in-situ technique provides incremental concentrations of a specific pollutant over incremental CO_2 emissions for each single vehicle passing. Assuming complete combustion this is equivalent to an emission factor per unit of diesel consumed[7]. To the extent that the fuel economy of the cars and LCVs remains stable over their lifetime (e.g.[3]) the relative deterioration rates derived below can be directly used for emission factors in the more common unit gram pollutant per distance travelled.

The information on the vehicle technology is retrieved via the recorded license plate from the vehicle registry. The details of measurement site have been described in previous articles[8,9]. The information for the equipment and calibrations are discussed in an earlier study[10]. It is important to note that all cars and LCVs have travelled about 1.5 km on an uphill road (9.2° gradient) before arriving at the measurement site. Therefore we can safely assume that vehicles are preconditioned hot and that cold-start effects are absent in the data. When calculating the engine load, the 9.2° road gradient is translated to an extra acceleration of about 1 m/s^2; thus our emission measurements per engine load are comparable with similar data from flat road driving. The measured cars and LCVs have a mean speed of 45 km/h and 44 km/h respectively. The 95th percentile of speed and acceleration are about 58 km/h and 2.5 m/s^2. The range of driving conditions of our measurement data is wider than the legislative chassis dynamometer test cycle, i.e. New European Driving Cycle, and we argue that the results are more representative to the real-world driving emissions[6]. The instrument was an Envirotest RS 3000 for ten years (2000-2010), then a Environtest RS 4600 for 2011 and 2012 and finally a different Environtest RS 4600 in 2013 and 2014. It is capable to measure remotely NO, CO, HC and opacity as well as CO_2. Unfortunately that generation of measurement devices has no NO_2 channel; therefore we will discuss carefully what to conclude on total NO_x (i.e. the sum of NO and NO_2) emission rates. The instrument drift was recorded through hourly calibration measurement with a reference gas; the data have been corrected for this drift (thanks to R. Gentala from Envirotest). The measurement uncertainty is given as ±15% for NO (likewise for HC, ±10% for CO[i]). Vehicle age is taken as the difference in months of the date of measurement and the date of first registration. Age in turn is translated to cumulative mileage using the latest Swiss mileage survey[6].

In order to avoid irregular emission behavior at deceleration or high instantaneous acceleration, we restrict the analysis to records with speeds between 27 and 57.5 km/h for cars and 23 and 58 km/h for LCVs, both are 5th percentile and 95th percentile of the data. In addition, the accelerations are restricted to between 0 and 2.5 m/s^2 for both cars and LCVs. In addition, we require 100 records for each mean value for statistically validity. After these filters, we have a total of 23,000 records for cars and 10,500 records for LCVs, disregarding about 10’000 records from Euro 5 vehicles (see SI Tables S1 and S2 for more details).

We calculate NO emission rates per unit of diesel consumption as a function of vehicle age or mileage, respectively, for diesel cars and LCVs for each Euro class separately. Then we test three major regression functions (linear, exponential and logarithmic forms) and chose the best fit based on a combination of statistical model fitting quality criteria, i.e. R^2 and p-value. Generally, a linear function is sufficient (Figure 3). The error bars represent 95th confidence interval for each mean value.

Driving and vehicle characteristics

Vehicles’ emissions can also be affected by instantaneous driving conditions and vehicle characteristics. To filter out these possible impacts, we evaluate the variation of vehicle specific power (VSP[11]) and of the ratio of rated engine power over mass across different age brackets for both cars and LCVs. The data shows uniformly distributed mean VSP of about 18 to 19±9 kW/t over age brackets within each technology layer (Figure 1 for cars, and Figure S1 in SI for LCVs). In order to eliminate a possible influence of engine load on the emission rate we calibrate all records as if measured at the mean VSP:

Emission records at higher VSP are downscaled, and records from lower VSP are scaled upwards. The scaling factor is taken from previous analysis of the same data showing a nearly linear increase of the emission rate with engine load for every Euro class[10].

The ratio of rated engine power to vehicle mass is used as an indicator for the vehicle characteristics. For diesel passenger cars, we observe an increase for more modern cars due to bigger engine power in modern cars (see Figure 1). But within each vehicle technology group, the mean values vary only by less than ±2 kW/t for Euro 1, Euro 3 and Euro 4 cars and ±3 kW/t for Euro 2. Similarly, we find engine power over mass for LCV vary less than ±2 kW/t within each vehicle technology group but modern vehicles tend to have higher engine power over mass values (see Figure S1 in SI). Given these small variations, we claim that vehicle characteristics are similar across age groups within each vehicle technology.

We therefore conclude that neither driving conditions nor vehicle characteristics can explain the changes in emissions from vehicles we measured over the fifteen years. Instead, we claim that the changes can be attributed to instability of the emission control system.

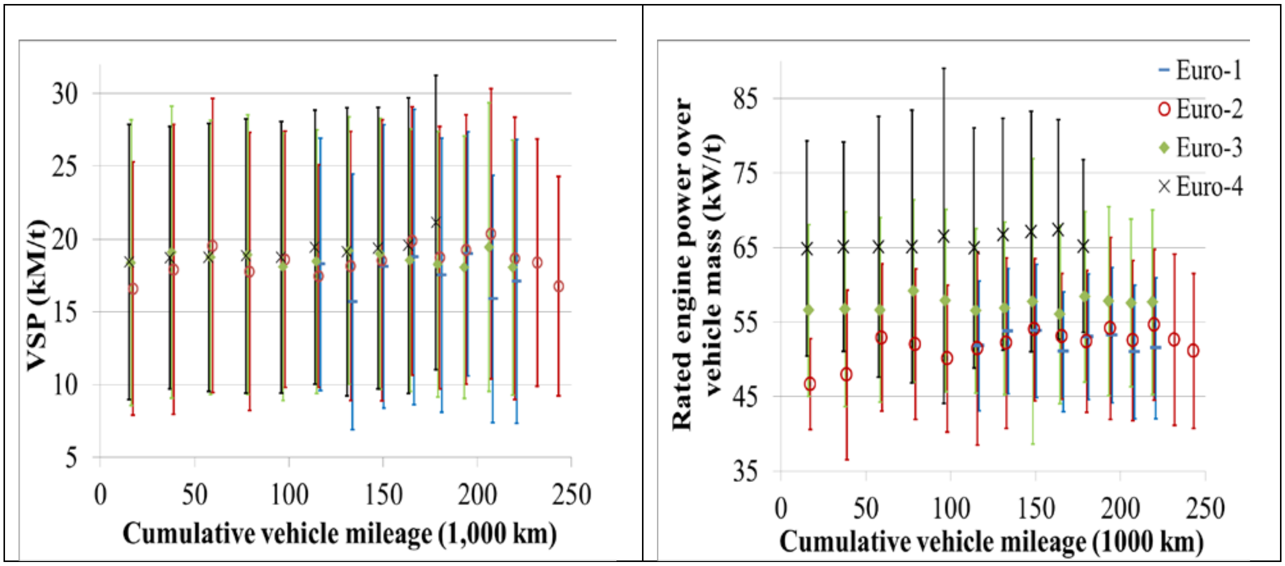


Figure 1. Mean VSP (left) and mean ratio of rated engine power (right) to vehicle mass as a function of cumulative mileage for each vehicle technology for diesel passenger cars (PC-D) from remote sensing in Zurich/CH. The error bars indicate one standard deviation of the mean.

Increasing NO emissions from diesel light-duty vehicles

In real-driving the emission rates do actually not decrease despite progressively tighter exhaust emission standards, as already noted for Europe's diesel cars earlier[9,12,13]. The oldest (Euro 1) and the latest cars and LCVs (Euro 4) actually have the lower emission rates; real-driving emission of Euro 2 and Euro 3 cars and LCVs are almost twice as high per unit fuel consumed as the Euro 1 counterpart's emissions.

We clearly find increasing unit NO emissions with increasing age and vehicle mileage for Euro 2, 3, and 4 passenger cars and LCVs (Figure 2). The linear regression is either the best fitting function or comparatively well fitting (see SI for R^2 and *p*-value). Notably for Euro 3 cars deterioration increases beyond 150'000 km but an exponential regression function does not fit markedly better. For simplicity we adopt a linear deterioration. Observations for Euro 4 cars extend up to 160'000 km. Over the mandatory 80'000 km durability interval, NO unit emissions of Euro 2, 3, and 4 diesel cars increase by 26%, 22% and 8%, respectively. Thus emissions (theoretically) double after 350'000 kilometers, when the car is older than 30 years. Compared to the exhaust emission control of gasoline cars this increase is two- to three-times lower[10], i.e. the emission performance is indeed much more robust. NO emissions from Euro 1 diesel cars seem to be very stable, at least in the observed range from 130,000 km to 220,000 km. These cars did not employ any dedicated exhaust emission control system. Therefore stable tailpipe emission rates indicate that the engine controls seem to be quite durable and working stably over this wide mileage range. This links up with earlier findings of stable emissions over a lower mileage range[2, 3]. We will discuss below how to interpret the increase also with respect to total NOx emissions.

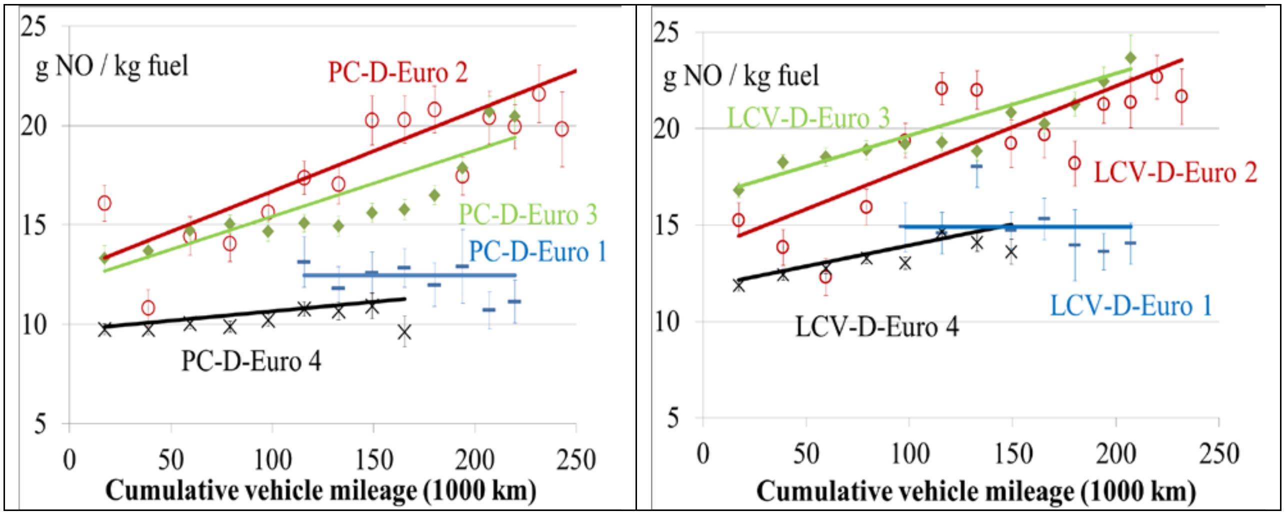


Figure 2. Increase of unit NO emissions of diesel passenger cars (left figure) and LCVs (right figure) as a function of vehicle mileage for Euro 1 to Euro 4 technologies. Each dot represents the mean value over at least 100 individual measurements; the error bars represent the 95$^{\%}$ confidence interval for the means.

Similarly for diesel LCVs, NO emissions increase linearly over the mandatory 80'000 km durability interval by 25%, 15% and 16% for Euro 2, 3, and 4 technologies, respectively (Figure 2). There are not enough measurement data for Euro 1 LCV for a statistical regression but emission rates appear certainly more stable. We observe the pattern of higher NO unit emissions for Euro 2 and Euro 3 LCV which is also found in diesel passenger cars. Euro 3 LCV's unit NO emissions are the highest and Euro 4 LCV's values are generally among the lowest level.

We also analysed unit HC, CO emissions as well as opacity as a function of vehicle mileage for Euro 1 to Euro 4 but no clear trends are observed. They are provided in SI for readers' reference (Figure S2-Figure S4).

No evidence for high emitters

One possible explanation of the gradual increase of unit emissions could be an increasing share of high emitters. Ideally, this question can be answered by repeatedly measuring the same group of vehicles over time. We do not have a substantial number of repeated measurements in our data sample, but we still can analyze the statistical behavior of the measured vehicle fleet. In Figure 3, we show the 50th percentile, 75th percentile, and 90th percentile of unit NO emissions as a function of mileage. The idea is that an increasing frequency of high emitting vehicles would lead to strongly increasing 90th percentile emissions as vehicles getting aged. However, for NO emissions, we observed that the change rates of different percentiles are quite similar. We acknowledge it might be wrong to associate 90th percentile emissions rates with individual cars that always emit, say ten times, higher than median emission level of the sample. But the similar change rates of the different percentiles demonstrate that there is no evidence for a higher frequency of high emission events as vehicle age. On the contrary, we can conclude a clear evidence for a gradual increase of unit emissions across the whole fleet.

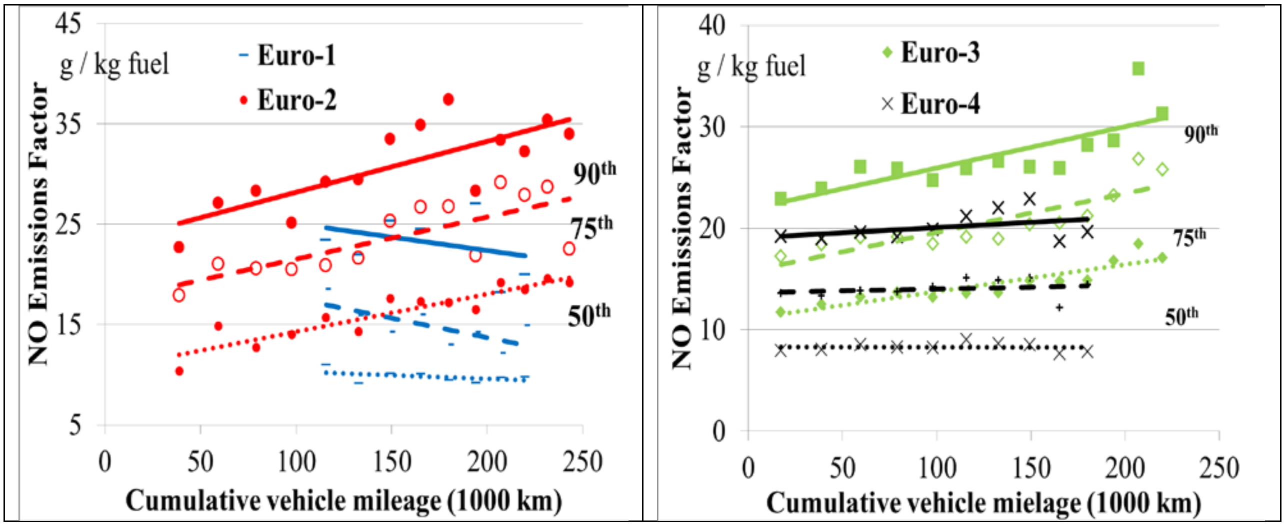


Figure 3. NO unit emissions from diesel passenger cars as a function of vehicle mileage (left charts: Euro 1 and Euro 2; right charts: Euro 3 and Euro 4). Plotted are the 50th, 75th, 90th percentiles. If the changing patterns of unit emissions are similar across the percentile classes, then deterioration seems to affect the whole fleet in general. If the emissions from the 90th percentile however increase strongly in large mileage categories, this might indicate an increasing frequency of cars with very deteriorated or non-functioning emission controls, that we might call high emitters.

Discussion of NOx deterioration rate

Apart from clustering by age and correcting for different engine loads we did not further manipulate the data. Hence the clear increase in NO tailpipe emission is are a feature of the observed fleet. In the following we discuss what this means for the deterioration of NO_x emissions. NO_x is the sum of the reactive nitrogen oxides NO and NO_2, commonly expressed in NO_2 mass units as follows: $NO_x = 46/30 * NO + NO_2 \equiv NO^e + NO_2$. The share p of primary NO_2 in total NO_x in the exhaust has been determined e.g. in chassis dynamometer tests (Grice et al. 2009b). Using this share p we can express

(1) $NO_x = NO^e + NO_2 = NO^e + p * NO_x = NO^e / (1-p)$

The deteriorated emission rate of NO at mileage t can be expressed – in our linear scheme – as

(2) $NO(t) = NO(0) * (1+DR_{NO}(t))$

Note that the NO deterioration rate expressed in NO_2 mass equivalents is identical, as the mass conversion factor cancels out. The deterioration rate of NO_x can now be expressed as a function of this NO deterioration rate and the share of primary NO_2 in the exhaust as follows:

(3) $$\boldsymbol{NO_x(t)} / \boldsymbol{NO_x(0)} = \left[\boldsymbol{NO^e(t)} / \boldsymbol{(1-p(t))} \right] \div \left[\boldsymbol{NO^e(0)} / \boldsymbol{(1-p(0))} \right] = [\boldsymbol{1 + DR_{NO}(t)}] * \boldsymbol{(1-p(0))} / \boldsymbol{(1-p(t))}$$

This formula helps understanding several points about the deterioration of NO_x exhaust emissions: The experimentally observed NO deterioration rate is an upper limit to the NO_x deterioration rate. Both are only equal if the share in primary NO_2 remains constant with mileage, i.e. if p(t) = p(0). When the share in primary NO_2 decreases, then the resulting NO_x deterioration will be lower than the observed NO deterioration, offsetting part of the NO increase. The lower limit to deterioration would be zero. In that case the factors in eq. (3) either do not change at all or an increase in NO emissions is offset by a decrease in primary NO_2 emissions. Applied to the different vehicle generations we can infer (Table 1):

a) Euro 1 cars (model years 1992 to 1996) were generally without exhaust after-treatment and had no more than 11% primary NO_2 in the exhaust (Grice et al. 2009a)[12, 16]. Thus the factor (1-p(0))/(1-p(t)) becomes only 0.94 even if the primary share p would drop from 11% to 5% over the vehicles lifetime mileage. The observed NO emission rate is rather stable. Therefore with high probability we can infer that also the NO_x emission rate will be stable confirming the old measurements that indicated zero NO_x emission deterioration[2,3].
b) The NO emissions of Euro 2 light duty diesel vehicles deteriorate by about 25% over the mandatory durability interval of 80’000 km. As argued above, if the primary NO_2 share p dropped by half over this period, the NO_x deterioration rate would still be at least 23%. Hence we argue that our measurements suggest a NO_x deterioration rate of 23% to 25% over 80’000 km for Euro 2 light duty diesel vehicles.
c) The NO emissions of Euro 3 diesel cars increase by 22% over the mandatory durability interval of 80’000 km. In order for NO_x emissions to remain constant the initial share of 30% primary NO_2 would need to be halved over this mileage. Assuming that p(80k) is three quarters of its initial value then the resulting NOx deterioration rate becomes 11%; which appears plausible. The NO deterioration rate for light commercial vehicles is 16%, and assuming that p(80k) is three quarters of its initial value then the resulting NOx deterioration rate becomes 5%.
d) The NO emissions of Euro 4 diesel cars increase only by 8% over 80’000 km. Total NOx emissions would remain constant if the share of primary NO2 decreased from an initial 40% to 35%. This appears within the range of reported p values anyway[12, 16]. We therefore tend to the assumption that there is hardly NO_x deterioration for Euro 4 diesel cars. The NO deterioration rate for light commercial vehicles is 15% respectively, and assuming that p(80k) is three quarters of its initial value then the resulting NOx deterioration rate becomes 0%.

Table 1 also gives a lower deterioration rate in case that the conversion efficiency of the diesel oxidation catalyst dropped to two thirds its initial level after 80’000 km. This shows that this assumption only makes a difference for the calculated NO_x deterioration rate of Euro 3 light duty diesel vehicles.

		Cars			**LCV**		
	p(0)	**DR_{NO}**	**DR_{NOx} @ 75% * p(0)**	**DR_{NOx} @ 66% * p(0)**	**DR_{NO}**	**DR_{NOx} @ 75% * p(0)**	**DR_{NOx} @ 66% * p(0)**
Euro 1	11%	0%	*0%*	*0%*	0%	*0%*	*0%*
Euro 2	11%	26%	22%	21%	25%	21%	20%
Euro 3	30%	22%	10%	6%	16%	5%	1%
Euro 4	40%	8%	*0%*	*0%*	15%	*0%*	*0%*

Table 1: Summary of observed deterioration rates for NO emissions DR_{NO} after 80’000 km and inferred deterioration rate for NOx emissions DR_{NOx} assuming 75% or 66% of the initial NO_2 conversion efficiency. Primary NO2 emission shares p from (16).
Values in italics: Lower limits, i.e. zero deterioration.

The conversion characteristics of the diesel oxidation catalyst are very different for HC and CO. Hence results obtained here for NO/NO_X deterioration rates cannot be transferred to these other pollutants.

Consequences for emission inventories

In summary we suggest a deterioration of tailpipe NO_X emissions over 80’000 km of 0%, 22%, 10% and 0% for Euro 1, 2, 3, and 4 technologies, respectively, for diesel cars and the same for light commercial vehicles expect a 5% deterioration for Euro 3. We suggest revising standard emission factor models accordingly. This revision can be relevant for historic emission inventories with higher shares of Euro 2 and Euro 3 cars, but will be of limited consequence for current emission inventories. The total contribution depends on several factors: The share of diesel in the car fleet, which tends to increase in European countries; the share of Euro 2 and Euro 3 technologies in the diesel fleet, which decreases since 2005; the share of older, i.e. deteriorated cars, which increases with time; and the difference in the average emission factor between diesel and gasoline cars.

For example, most diesel cars measured in Zurich in 2005 were already Euro 3 cars younger than five years, hence with moderate deterioration only. Only 10% were older Euro 2 diesel cars, with correspondingly much higher deterioration. Accounting for the deterioration of these Euro 2 and 3 cars increases the average NOx emission rate of all diesel cars by 8% (cf. SI Table S4 for numbers). With a share of only 10% in the total car fleet, i.e. including gasoline cars, and roughly five times higher emission rates than gasoline cars[9], NO_x emissions should be some 3% higher when accounting for deterioration. If the share had been 30% (as in the year 2014 in Zurich) or 50% as in other European countries[17], the increase would have been 5% and 6% respectively. The strong rise of diesel cars in European countries began only after the year 2000[17], when the share of the more deteriorating Euro 2 cars was already diminishing. Therefore we estimate that the total NO_x emissions from the car fleet will not have increased by more than 10% in any year in any European country. The diesel share of LCVs however has been higher from the beginning and increased from 36% in 2001 over 57% in 2005 to 81% in 2014 at the measurement site. Hence the extra NO_x emissions due to deterioration are higher and estimated between 7%, 11% and 5% for these years in Zurich respectively. This also indicates the magnitude of the increase to be expected in other European countries.

Euro 4 diesel cars were found with negligible NO_x deterioration; however, the emission control depends on the technology employed, its design and complexity. Therefore it cannot be taken for granted that Euro 5 and later cars have stable emission behavior. For instance many of the latest generation of diesel light-duty vehicles (Euro 6) are equipped with SCR controls, for which durability problems have been reported[18] and whose functioning also depends on operating history determining its operating temperature as well as supply of a reduction agent. Hence, it is advised to monitor their long-term emission behavior. In addition the observed deterioration of emission controls for light-duty vehicles calls into question the assumed long-term stability of exhaust emission controls for heavy-duty vehicles. We suggest reviewing their real-driving emission behavior urgently.

Supporting Information

Supporting information contains details on the distribution of measured diesel passenger cars and LCVs. Unit CO, HC and Opacity emissions as a function of vehicle mileage for diesel passenger cars and LCVs of Euro 1 to Euro 4 are also shown in SI.

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