Speed Dependence of NO_2/NOx Emission Ratio?

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1. Introduction

Although levels of emitted nitrogen oxides (NOx) have fallen sharply, roadside measurements indicate that ambient concentrations of nitrogen dioxide (NO_2) appear to be stabilizing or even to be rising.

This rise in the ratio of NO_2 to NOx parallels the implementation of oxidation catalytic converters in diesel vehicles. These converters oxidize carbon monoxide and hydrocarbons originating from imperfect combustion in the engine, but may also convert NO to NO_2 in certain temperature conditions. In addition, more and more diesel cars are equipped with original equipment manufacturer (OEM) particle filter (PF) systems that employ NO_2 to oxidize trapped soot at lower temperatures. This NO_2 is intentionally generated from engine-out NO in the catalytic converter preceding the trap, but not controlled. Excess NO_2 may thus escape from the system as tailpipe emissions. Consequently, precise measurement of vehicle emissions of NO and NO_2 were performed (Alvarez et al., 2008).

Since NO_2 formation in diesel engines depends on temperature conditions, the NO_2/NOx ratio could depend on engine power and therefore on vehicles speed. Emission values from a measuring campaign with 20 diesel vehicles (Alvarez et al., 2008) with real-world highway cycles were reinvestigated with respect to speed dependency of the NO_2/NOx ratio. The results are compared with the corresponding guidelines if the handbook of emission factors (HBEFA 3.1, 2010).

On different highway sections in Austria, two different speed limits (100 km/h and 130 km/h) are applied depending on air pollution level. Different NO_2/NOx emission ratios of passenger cars and light duty vans due to different speeds should be detected in roadside ambient NOx and NO_2 measurements; this is presented in the last section of this paper.

2. NO_2/NOx emission ratios for diesel vehicles in real-world highway conditions

Emission ratios of NO_2/NOx of current car fleets are presented for real-world highway driving situations, concretely the BAB cycle (Bundesautobahn Deutschland) and the Common Artemis Driving Cycle (CADC) (André, 2004). The 20 vehicles are grouped according to their combustion principle and certification category: 5 diesel Euro-3 (PD3) and 9 Euro-4 (PD4). Diesel cars with OEM PFs are discussed as a separate group (6 PD4 PF), since NO_2 is intentionally formed in the precatalytic converter of some of these systems to oxidize particles in the trap.

The speed range for the different highway conditions was from 100 km/h to 140 km/h. For each speed level, the average of total NOx and NO_2 emission was calculated for all vehicles in the same group (PD3, PD4 and PD4 PF) together with the respective mean NO_2/NOx ratio. At a speed of 140 km/h, the NOx emission factors are 2-3 times higher than at 100 km/h. For our purposes the emission ratio of NO_2/NOx is of interest.

There is no evidence for a speed dependency of the NO_2/NOx emission ratio for the vehicle sample PD3, see Figure 1. For vehicle sample PD4, the NO_2/NOx ratio of the emission factor decreases by about -0.002/ (km/h) and for sample PD4 PF by about -0.004/ (km/h). Thus, the NO2/NOx emission ratio of Euro-4 diesel vehicles features a speed dependency.

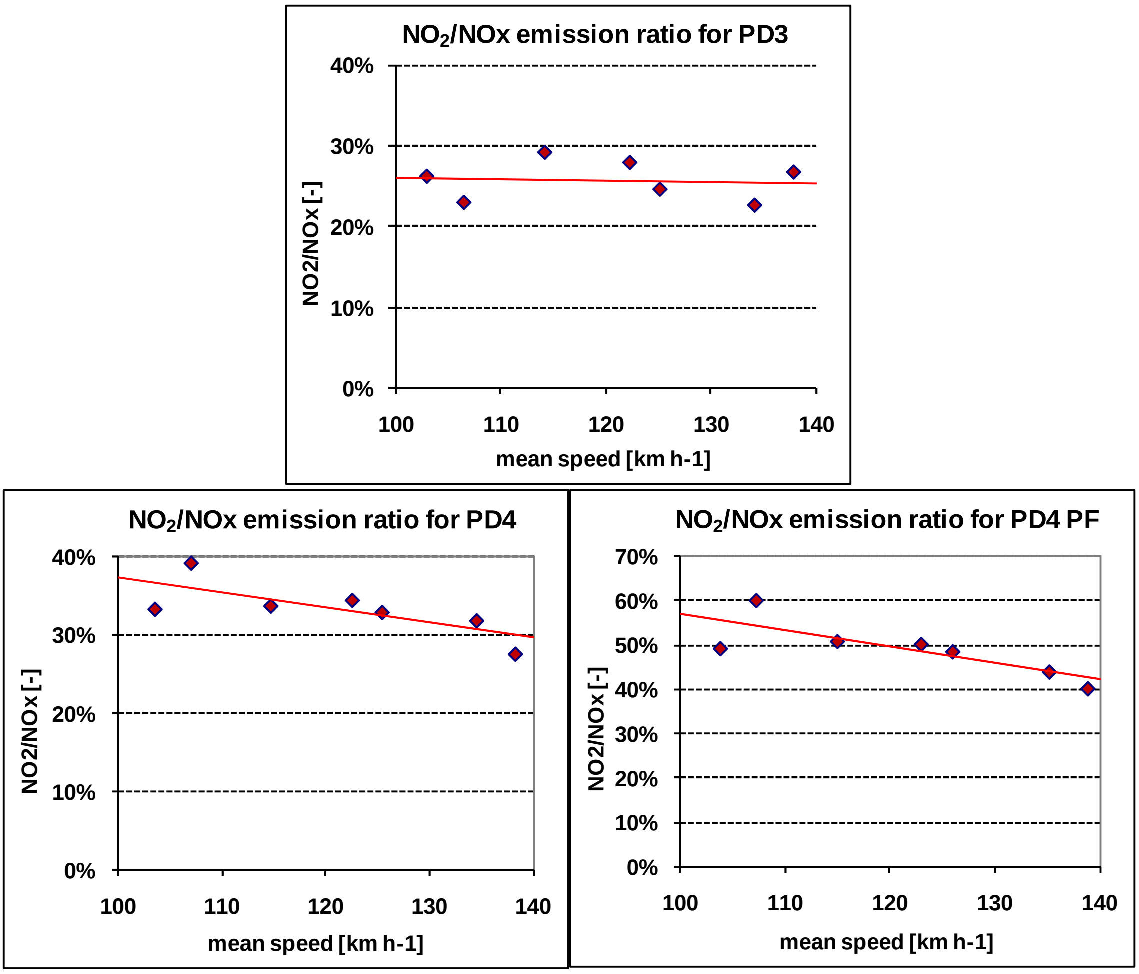

Figure 1: Mean NO_2/NOx emission ratios for diesel vehicle samples Euro-3 (PD3), Euro-4 (PD4) and Euro-4 with particle filter (PD4 PF) in dependency on mean vehicle speed.

3. NOx and NO_2 emissions of passenger cars on Austrian rural highways according to HBEFA 3.1

Emission factors of NOx and NO_2 for an average fleet of passenger cars on e.g. rural Austrian highways are derivable from the handbook of emission factors (HBEFA 3.1, 2010) for different mean speeds. They show a strong dependence on vehicles speed in good agreement with the emission above-mentioned measurements. Although the absolute emission factors are expected to decrease strongly in the next years, the relative speed dependence shall remain stable (fig. 2). In accordance to HBEFA 3.1 however, the NO_2/NOx emission ratio should not be dependent on vehicles speed (fig. 3).

The NO_2/NOx emission ratios for passenger cars presented in fig. 3 coming from HBEFA 3.1 are rather smaller than those presented in fig. 1. The HBEFA 3.1 gives average values for all Euro classes and also gasoline vehicles corresponding to the mean fleet, in that example of Austrian highways, whereas the diesel Euro-4 vehicles (fig. 1) show comparatively high NO_2/NOx ratios.

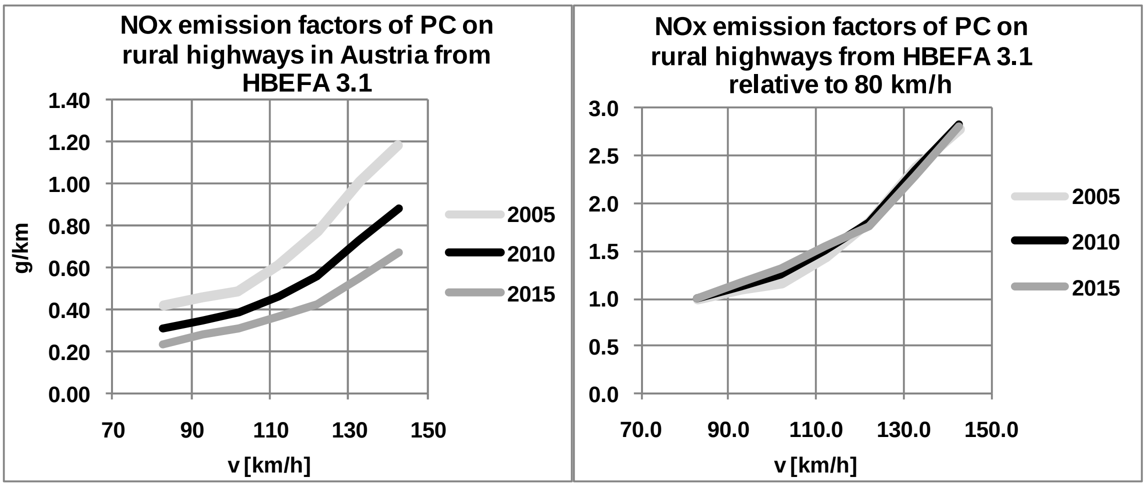

Figure 2: NOx emission factors of passenger cars (PC) on rural Austrian highways as derived from HBEFA 3.1, for 2005, 2010 and 2015; absolute values (left-hand) and relative to emission factors at 80 km/h (right-hand).

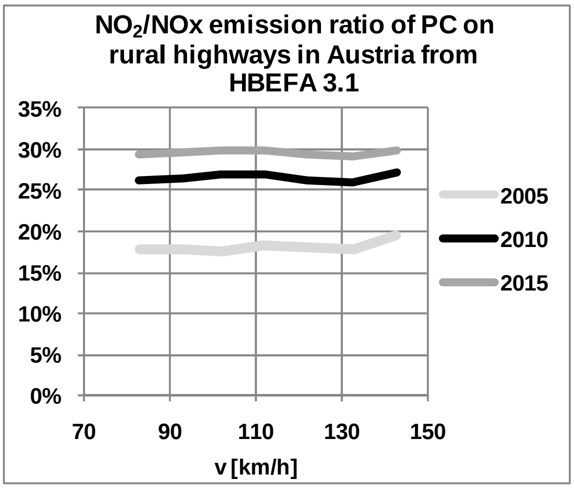

Figure 3: NO_2/NOx emission ratios of passenger cars (PC) on rural Austrian highways as derived from HBEFA 3.1, for 2005, 2010 and 2015.

4. Effect on mean vehicles speed on NO_2/NOx ratio of roadside ambient air pollution

The data base of 20 diesel vehicles is rather small for a generalized statement concerning speed dependence of the NO_2/NOx emission ratio. In addition, roadside ambient air pollution measurements of NOx and NO_2 at Austrian highways with varying speed limits have been investigated regarding speed effects on NO_2/NOx ratios.

On several highway sections in Austria, temporary speed limits alternate between 100 km/h and 130 km/h to improve air quality (Thudium et al., 2009). If NO_2/NOx emission ratios depend on vehicles' speed, a shift of the NO_2/NOx-ratio of measured air concentrations between these two speed phases should be derivable from roadside measurements.

A comparison between the two speed limit phases of 130 km/h and 100 km/h demands building of a really comparable database, since the NO_2/NOx ratio in ambient air depends on different parameters: the portion of light vehicles at total NOx emissions of the highway, the atmospheric mixing conditions, the amount of NOx concentration and the day time.

For explanation, roadside measurements at Kundl in Tyrol, Austria, performed in 2007 before installing variable speed limits are shown in figure 4 (Thudium, 2007): vertical transports in the atmosphere are very essential for diluting air pollutants, especially in valleys. In thermal inversions, these transports are reduced, leading to enhanced NOx concentrations. NO_2

concentrations are only slightly enhanced because of limited availability of ozone (fig. 4). The portion of NO_2 within NOx concentration depends on the amount of NOx for chemical reasons, what can be seen from the monthly averages of NOx and NO_2/NOx (fig. 4).

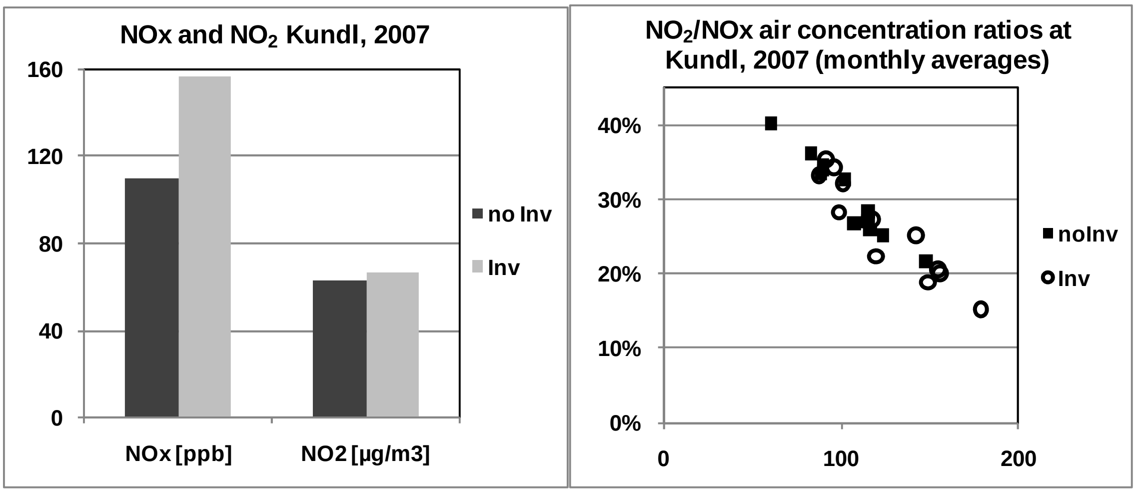

Figure 4: Mean NOx and NO_2 air concentrations at Kundl, roadside station in Tyrol, Austria, 2007 with and without thermal inversion, respectively (left-hand) and monthly averages of NO_2/NOx ratios (right-hand).

Typical NO_2/NOx ratios at a certain NOx level don't differ when there is inversion or not, but there are higher NOx values with inversion than without inversion, leading to lower NO_2/NOx ratios. The NO_2/NOx ratio shows a typical diurnal variation with a minimum in the morning and with lower values over the whole day in case of inversion (fig. 5).

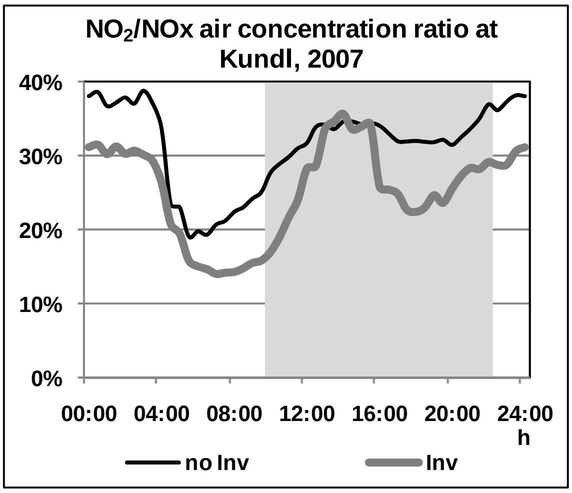

Figure 5: Diurnal variation of NO_2/NOx air concentration at Kundl, 2007, with and without thermal inversion, respectively.

In consequence, an evaluation of the influence of vehicles speed on NO_2/NOx ratio of roadside air concentration has to be investigated in speed limit intervals where other variables essentially determining the NO_2/NOx ratio are similar. So a division in inversion and 'no inversion' situations had to be made. Only workdays on day time between 10 and 22 h were considered, so that the frequencies of speed limit of 100 km/h and 130 km/h were about 50% each. Furthermore only periods were considered where the portion of light vehicles NOx emissions at total highway emission were within certain limits, so that the average of that portion was equal for both speed limits at about 40%. Thus, the influence of heavy duty trucks emitting only small portions of NO_2 (HBEFA 3.1) on ambient air NO_2/NOx ratio was kept constant.

Finally the comparison of NO_2/NOx ratios between situations with speed limit of 100 km/h and 130 km/h, respectively, has to be performed per interval of NOx, as shown in fig. 6:

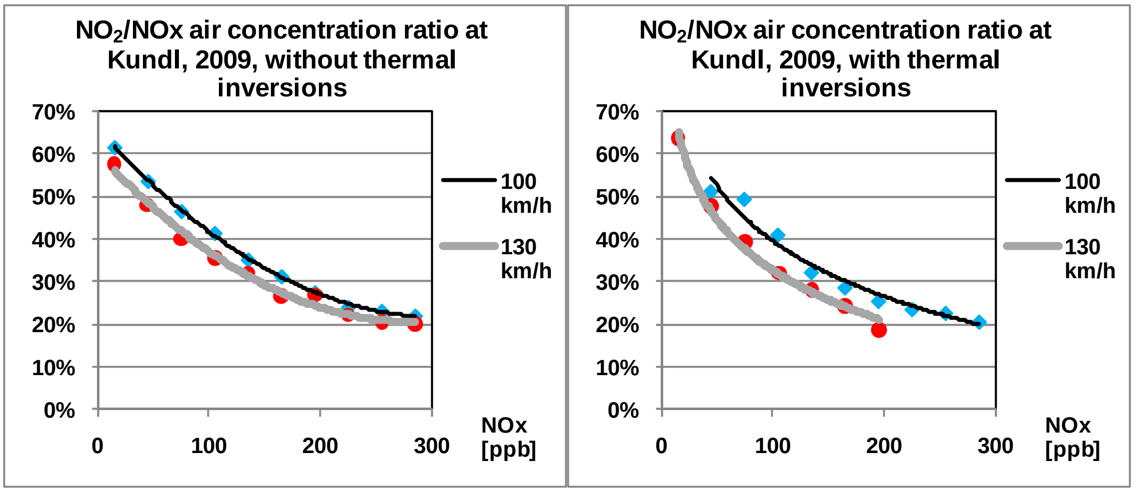

Figure 6: Roadside NO_2/NOx air concentration ratio at Kundl (Tyrol, Austria), 2009, for speed limits of 100 km/h and 130 km/h, respectively, in situations without (left-hand) and with (right-hand) thermal inversions.

The NO_2/NOx ratio in air concentration was larger with a speed limit of 100 km/h than with a speed limit of 130 km/h; the difference amounts to 3-4% in absence of thermal inversions. In the presence of thermal inversions, the difference of NO_2/NOx ratio in air concentration between the speed limits of 100 km/h and 130 km/h amounts to 4-8%, but there residual NO_2 from former times may also play a role.

With a speed limit of 130 km/h, it's not only the faster speeds of passenger cars and light duty vehicles which lead to smaller portions of NO_2 in the NOx emissions (the absolute amounts of emissions are always increasing with speed!). Then the style of driving becomes more aggressive, the speeds are more varying, both leading to lower NO_2/NOx emission ratios intentionally. In October 2006, with permanent speed limit of 130 km/h on this highway, the amount of passenger cars with more than 150 km/h was 15 times more frequent (0.7% of all passenger cars) than in November 2006 with a permanent speed limit of 100 km/h. Speeds with more than 160 km/h were in October 10 times more frequent than in November 2006. Such very fast vehicles oblige all the others to evade on the right-hand lane which is rather occupied by trucks.

5. Conclusions

Emission measurements of NOx and NO_2 with 20 diesel vehicles of certification category Euro-3 and 4 show a speed dependence of the NO_2/NOx ratio for Euro-4 vehicles, where higher vehicle speeds lead to lower ratios. Although the handbook of emission factors HBEFA 3.1 does not contain such dependence, roadside air concentration measurements with different speed limits indicate lower NO_2/NOx ratios at increased vehicle speeds. It has to be emphasized that with higher speeds the absolute emissions both of NOx and NO_2 increase essentially; only the ratio of the two compounds decreases.

References

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