

Road traffic's contribution to air quality in European cities



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Traffic in Paris seen from Arc de Triomphe, picture from Wikipedia

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Summary

The contribution of road traffic emissions to air pollution in European urban areas has been analysed in this study focusing on NO₂ and PM, including non-exhaust particles, black carbon and secondary particles. Furthermore, measures currently applied to minimise the traffic contribution to urban air pollution in Europe have been reviewed.

Traffic contribution to air pollution in European urban areas

The emissions from road traffic are still one of the main sources of air pollution relevant for human exposure, especially in urban areas. 41 % of the European (EU 27) urban population lives in areas where the EU air quality 24-hour limit value (LV) for particulate matter (PM₁₀) was exceeded in 2010, while 7% lives in areas where the annual EU LV for NO₂ was exceeded in the same year. Exceedances of NO₂ and PM₁₀ limit values are widespread over Europe, occurring most frequently at traffic (roadside) stations. Exceedance of the NO₂ annual LV was reported for 44 % of all European traffic stations in 2010, while 33% of the traffic stations reported exceedance of the PM₁₀ 24-hour LV.

The increasing number of diesel cars has led to an increase in the fraction of direct NO₂ emissions in NO_x, partly counteracting the effects of generally reduced NO_x emissions from road traffic. Diesel vehicles are known to be significant emitters of both NO₂ and PM₁₀ and the share of diesel in the overall fuel consumption for the EU-27 has increased from 50 % in 2000 to 63 % in 2010. In addition, vehicle activity in urban areas has increased by 27 % in all EU Member States over the last decade (2000-2010). These developments have led to an increase in air pollution levels in some European cities, as registered for NO₂ by some traffic stations, or slowed down improvements brought about by mitigation measures.

There is a general consensus in the reviewed literature that road traffic is the main source of NO₂ and NO_x in urban areas, but the actual quantification of the traffic emissions contribution to ambient concentrations is seldom done. The authors of the reviewed articles show a concern for the observed NO₂ concentration levels, the increased fraction of direct NO₂ emissions from diesel vehicles, and the difference between the emission standards and real world driving emissions, especially for diesel vehicles. The literature from recent years has focused on the increasing NO₂/NO_x-ratio trends in ambient air and the contributions from secondary and primary NO₂.

In Member States' Time Extension Notifications (TENs) requests to the Commission for compliance with the set concentration limit values at a later date, the average of the reported contribution from urban and local traffic emissions to ambient NO₂ concentrations was 64% at traffic sites. The local traffic contributions ranged from 10 to 80% and from 3 to 56% for the urban traffic contribution. The regional contribution might of course also have road traffic as origin but this was not further investigated by the Member States.

Quantification of the traffic contribution to PM levels in urban areas is complex, both due to the many sources to primary emissions of PM and due to the secondary formation of PM, from precursor gases that are also emitted by other sources.

While emissions from road transport are mostly exhaust emissions arising from fuel combustion, non-exhaust releases contribute substantially to primary PM particulate matter resulting from direct wear emissions (tires, breaks, road, etc) together with suspension of dust load, already deposited on the surface, by tyres and vehicle induced turbulence. The relative importance of the non-exhaust emissions has increased in the last two decades due to regulation of exhaust emissions, while vehicle activity has at the same time increased. Emissions of non-exhaust PM are not yet regulated. Estimating emissions with the COPERT model the relative share from non-exhaust emissions has increased from 2 % to 21 % for PM_{2.5} and from 21 % to 33 % for PM₁₀ from 2000 to 2010. The estimate is possibly largely underestimated at some locations as road wear emissions and re-suspension were not included in the emission model. The percentage of non-exhaust in total primary PM road traffic emissions is expected to increase further. Distinguishing between the contributions of non-exhaust and the exhaust emissions to PM concentration levels is problematic, but essential for the development of appropriate mitigation plans. As a first step, it is a priority to develop and apply emission models that estimate all road traffic non-exhaust PM emissions.

In the reviewed articles which quantify source contributions to PM, road traffic is found to be a relatively large source in most cities. There are, nevertheless, large variations from city to city, as other important PM sources, e.g. desert blown dust or wood burning for domestic heating, vary considerably geographical and temporally.

In the reviewed articles the range of the traffic contribution to urban PM concentrations is from 9-53% for PM₁₀ and 9-66% for PM_{2.5}. The number of studies published is unfortunately too low and limited in spatial and sometimes temporal coverage to derive a useful statistical analysis representative for urban areas across Europe. On the other hand, the TENs indicate an average estimate of 34 % of urban and local traffic contribution to the measured PM₁₀ concentrations at traffic sites, and 15% to urban background sites. The reported local traffic contributions to PM₁₀ concentrations measured at traffic stations range between 6 and 54%. The reported overall regional total contribution to measured PM₁₀ at traffic sites ranged from 15 to 75%, with an average of 51%. The contributions reported in the articles seem to be somewhat larger than what is reported in the TENs, which report relatively high regional contributions. This discrepancy is probably due to the differences in methods and the uncertainty related to the quantification (or not) of the non-exhaust emissions, as well as the the differences in number of sites.

Table 1: Summary table of traffic contributions as reported in the TENs

| Component | Station type | Local traffic % range (average) | Urban traffic % range (average) | Local and Urban % range (average) |
|------------------|---------------------|--|--|--|
| PM10 | Traffic | 6-54 (21) | 3-39 (13) | 13-61 (34) |
| PM10 | Urban background | 0-10 (4) | 6-15 (11) | 6-22 (15) |
| NO2 | Traffic | 10-80 (47) | 3-56 (17) | 34-91 (64) |
| NO2 | Urban background | NA | NA | NA |

Table 2: Summary table of traffic contribution based on the literature review

| | Traffic sites | | Urban background | |
|--------------|---------------|---------|------------------|---------|
| | Range % | Average | Range % | Average |
| PM10 | 70-19 | 39 | 53-9 | 27 |
| PM2,5 | 65-22 | 43 | 66-9 | 34 |
| PMc | 89-18 | 47 | 70-7 | 39 |

When quantifying the road traffic contribution to ambient PM, the focus is often on the primary and local PM emissions. However, traffic emissions are also leading to the formation of secondary particles and long range transported particles contributing to ambient concentration levels, but this is often not included in the estimate. Traffic's contribution to the European regional background of PM₁₀ concentrations was estimated (through the use of modelling) to be about 15-25 % and around 30 % of the PM_{2.5} concentrations. The current literature review shows that the road dust contribution to the annual mean PM concentrations varies between 8% and 34% for the PM₁₀ fraction, with generally lower contribution to the PM_{2.5} fraction and larger to the coarse fraction (PM₁₀-PM_{2.5}). Large contributions from road dust, up to 74 % of PM₁₀, were found mainly during spring months in Stockholm, but it was much lower the rest of the year. Spring outbreak of road dust is due to drying of the roads after the snowmelt, releasing particles from sanding/salting of roads and particles caused by the use of studded tyres, which enhanced pavement abrasion during winter.

Quantification of the traffic gaseous emissions (NO_x, NH₃ and VOCs – volatile organic compounds) to the formation of secondary organic aerosols (SOA) or secondary inorganic aerosols (SIA) was not covered in this review, but it is evident that secondary particles contribute significantly to the PM levels. Focus in better understanding the formation of SOA and SIA in the urban environment is hence crucial for understanding how to reduce PM concentrations in most of Europe.

Road traffic is an important source of black carbon (BC) in urban areas, but its contribution to ambient concentrations is not quantified in the reviewed literature. BC concentration levels vary proportionally to those of traffic-related gaseous pollutants (CO, NO and NO₂) and increased levels are found close to traffic at kerbside stations. Elemental carbon (EC) emission estimated by COPERT show a reduction over the last decade from road traffic in European urban areas.

Mitigation measures

As road traffic is a major source of the air pollution to which urban populations are exposed, it is necessary to both reduce traffic emissions and find ways to minimise population exposure. European authorities seem to focus on measures aiming at the reduction of traffic emissions, rather than structural measures like urban (re)planning aiming at the reduction of the population exposure or reducing transportation needs.

A decrease in the vehicle park diesel share would clearly be beneficial to NO₂ concentrations, both due to the higher fraction of NO₂ in NO_x diesel emissions and the considerably higher NO_x emissions from diesel vehicles. Diesel engines are also the main contributors to primary BC emissions, whereas petrol has higher NH₃ emissions than diesel. The use of urea to reduce NO₂ emissions of the upcoming EURO 6 diesel vehicles will probably lead to an increase of NH₃ emissions. When

introducing a measure or new technology to decrease emissions of a certain pollutant, it is crucial to evaluate potentially negative effects in emissions of other pollutants.

Initiating a faster shift towards hybrid, electric cars, and smaller petrol cars should be among the prioritised measures, especially in urban areas. Taxation regimes have shown to be efficient in changing petrol and diesel vehicle shares, so a similar effect should be possible for less polluting vehicles. Taxation should take into account the emissions of air pollutants along with those of greenhouse gases (GHGs). In addition, other traffic measures to optimise the traffic flow and driving behaviour, e.g. by speed moderations, seem to be inevitable for most cities in order to comply with the LVs in the short term and before the shift towards less polluting vehicles has the necessary effect.

The need for modal change from passenger cars and motorbikes to low emission public transport, walking and biking is extremely important, especially taking into account the urban growth and predicted need for transportation. Considering emissions from heavy duty vehicles and busses, new technology and fuels might give improved reductions in the future, but an integrated urban planning will be important to ensure good transport solutions and reduce the need for transport work in cities. The goal should be that the total km driven of all vehicles decreases.

Measures to minimise non-exhaust PM emissions are also important to reduce PM concentrations. Studies looking at street washing or sweeping so far did not show a large effect and dust suppressant might be useful but might have negative consequences. Less use of studded tyres would improve the air quality in several Nordic cities and winter maintenance and traction control measures (salting and sanding) should be optimised to reduce the emissions. Speed limit reduction is likely to give effect also for the non-exhaust emissions, but as it depends on the local situation, and factors like car fleet, level of congestion and driving behaviour, a general recommendation cannot be given.

The evaluation of the effect of measures is difficult as trends in pollution levels can only be proven after several years, due to e.g. variations in meteorology and other factors. The results from models estimating the effect of a mitigation measure are highly dependent on the estimated emissions, which often tend to exclude non-exhaust and parameters like flow/congestion or driving pattern. The current review does not allow making recommendations on the most effective measures, due to the relatively few studies published with a quantification of the effect. Furthermore, the effectiveness of measures varies largely, depending for example on how and where measures are applied. However, action needs to be taken promptly as LVs are currently exceeded and the impacts have a high social cost.

Experience and knowledge on effective measures need to be shared among stakeholders, municipalities and countries. More interactions among decision makers are hence highly recommended. Understanding and quantifying the actual emissions on the road and their contributions to concentrations, together with further focus on understanding the processes behind non-exhaust emissions, and the secondary particles in urban environments, would lead to more knowledge based recommendations for mitigation of air pollution from road traffic.

1 Introduction

The emissions from transport and in particular from vehicles are still one of the main sources of air pollution relevant for human exposure. Emissions from traffic are made up of particulate and gaseous pollutants. In the particle phase, the main emissions are black carbon (BC) or soot-like aerosols, sulphate- and ultrafine particles. These particles may be primary (e.g., BC) and secondary in origin (e.g. secondary organic aerosols, SOA, or secondary inorganic aerosols, SIA). In the gas-phase, traffic emissions are dominated by nitrogen oxides (NO_x), carbon monoxide (CO), sulfur dioxide (SO₂), ammonia (NH₃) and volatile organic compounds (VOCs). The latter are directly emitted into the urban atmosphere, and they may become highly reactive species (Robinson et al., 2007). Traffic is also responsible for non-exhaust emissions related to brake, tyre and road wear as well as suspension of previously deposited particles. Finally, one additional source related to road transport, influencing air quality, is evaporation of VOCs from urban deposits for instance tanking stations (Huang et al., 2011; Bahreini et al., 2012).

1.1 Status of Air Quality in European Cities

The European air quality assessment (EEA, 2012a) showed that 41 % of the European urban population lives in areas where the EU air quality 24-hour LV for particulate matter (PM₁₀) was exceeded in 2010, while 7% lives in areas where the annual EU LV for NO₂ was exceeded in the same year. EEA (2012a) also shows that the annual mean NO₂ and PM₁₀ concentrations at urban background (non-traffic) sites have decreased slightly at most locations. However at traffic sites, close to the source (i.e. road traffic) the annual mean concentrations are generally higher than at urban background sites and show a less consistent downward trend. Conversely, some European cities have experienced an increase in road-side concentrations, particularly so for NO₂ (Guerreiro et al., 2010).

In 2010, as in previous years, nearly all European countries recorded exceedances of the annual NO₂ limit values (LV) at one or more stations, with the highest concentrations and number of exceedances recorded by traffic stations. Exceedance of the annual LV was reported at 44 % of traffic stations, with a maximum observed concentration of 2.6 times the LV. Most member states had also difficulties to comply with EU limit values for PM₁₀, for which the attainment year was 2005. In 2010, the PM₁₀ 24-hour LV was exceeded at 33% of traffic sites. and at 29% of urban background sites. The highest concentrations measured in the EU were exceptionally high, above 2000 µg/m³ in Iceland, due to the eruption of the volcano Eyjafjallajökull. (EEA, 2012a). In Figure 1 this domination of exceedances at traffic stations is illustrated by distance-to-target graphs (EEA, 2012a). Due to these exceedances a large percentage of the population is still exposed to concentrations levels which are expected to have adverse effects on human health. As shown in Figure 2 from 2001 to 2010 18%-41% of the urban population in the EU27 was exposed to PM₁₀ levels above the limit value and no particular trend was seen (EEA, 2012a). The large variation is explained by meteorology (changing atmospheric and dispersion conditions from year to year).

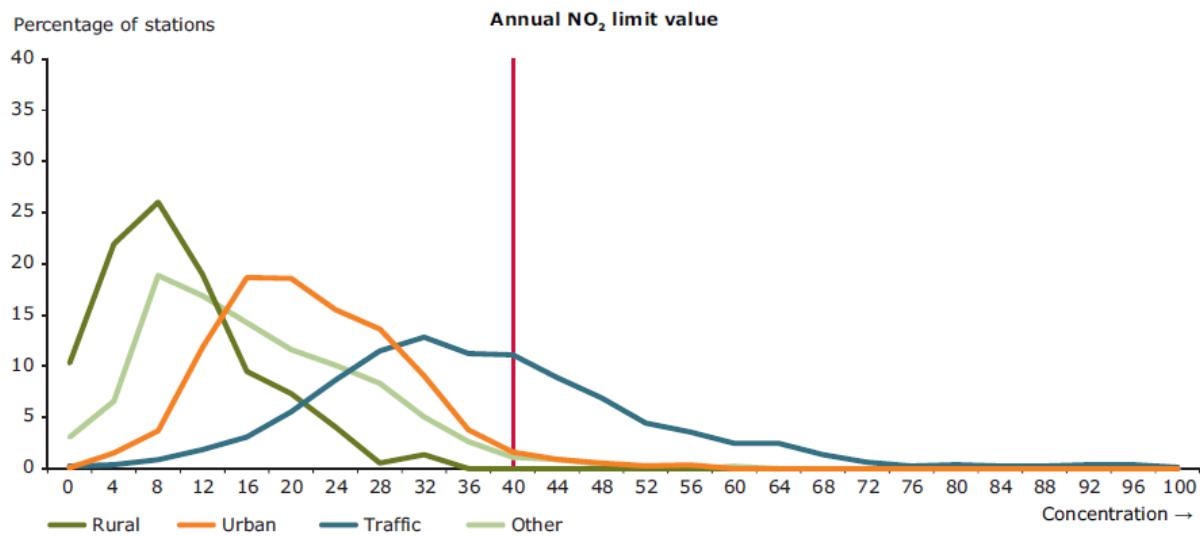
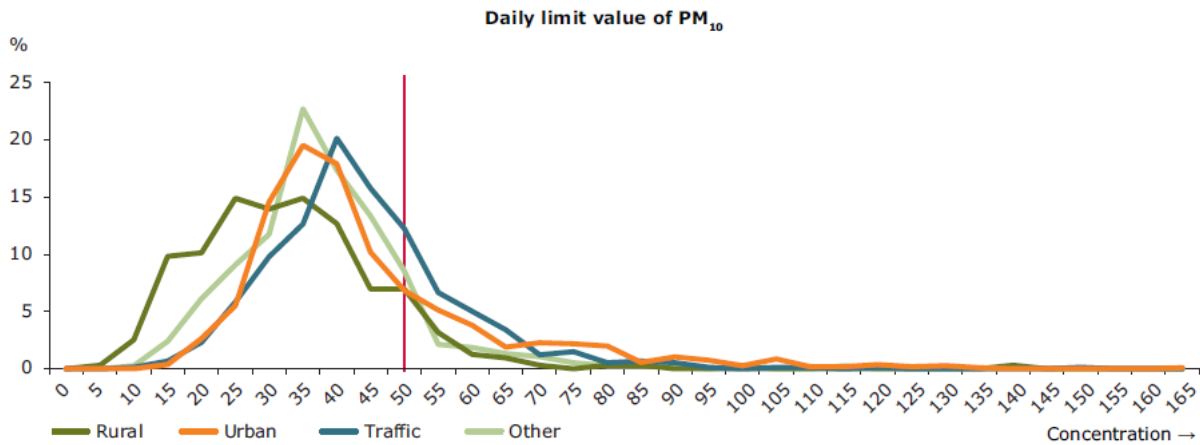


Figure 1 Distance-to-target graphs for limit values for PM₁₀ and NO₂ in 2010. Fraction (%) of stations with various concentrations µg/m³ (EEA, 2012a). The vertical red line corresponds to the daily (PM₁₀) and annual (NO₂) LV.

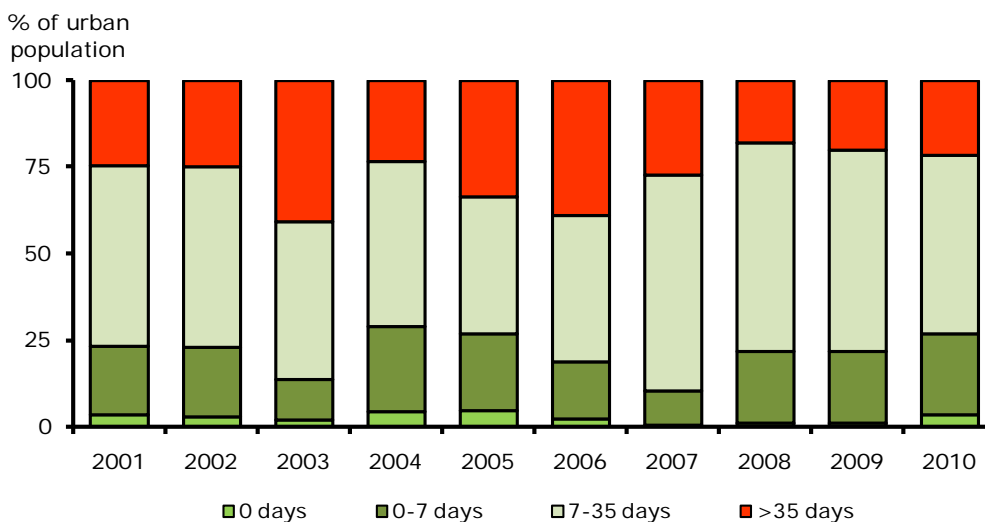
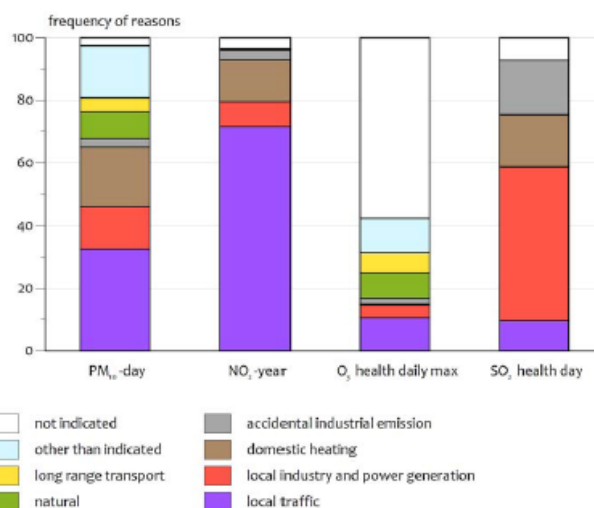


Figure 2 Percentage of European urban population (EU27) resident in areas where exposed to PM₁₀ levels exceeding the daily limit value, 2001-2010 (EEA, 2012a).



b)

Figure 3 Reported reasons for exceedances of limit and target values of PM₁₀, NO₂, O₃ and SO₂ for the year 2008 (Jimmink et al., 2010).

These findings demonstrate that PM₁₀ and NO₂ concentrations must be reduced substantially in larger areas of Europe, including the areas influenced by traffic, in order to meet the limit values. Based on the air quality questionnaires submitted by EU Member States to the Commission, was road traffic the main source of PM₁₀ and NO₂ in urban areas, and this situation has not changed in the last decade (Jimmink et al., 2010). Traffic was the single most reported reason for exceedances of the PM₁₀ and NO₂ limit values in 2008 (Figure 3).

The percentage of the EU urban population exposed to SO₂ concentrations above the EU 24-hour limit value was reduced from 11% in 1997 to 0% in 2010. Population exposure to CO ambient concentrations above the EU limit value is limited to a few restricted areas. Regarding VOCs, exceedances of the benzene limit value are limited to a few locations in Europe, primarily situated close to traffic. The average benzene concentration measured at traffic stations in 2010 declined to less than half of the 2001 level (EEA, 2012a). Therefore, it may be concluded that SO₂, CO and benzene concentrations emitted by traffic no longer pose a major threat to urban air quality.

1.2 Components in focus; non-exhaust particles, black carbon and ammonia

Even though most works on the impact of road traffic on urban aerosols have traditionally focused on exhaust emissions (Colville et al., 2001; Ruellan et al., 2001; Harrison et al., 2008), recent scientific and policy-oriented interest is growing on the non-exhaust emissions (Bukowiecki et al., 2010; Beuck et al., 2011; Amato et al., 2011). This is due to the relative increase of importance of the non-exhaust emissions. Over the past years huge efforts were made in regulating motor exhaust emissions (EUROx legislation, x = 1 to 6 for light-duty vehicles, or x= I-VI for heavy-duty vehicles), and in elaborating targeted air quality plans. The relevance of road dust emissions is stressed by: i) the large contribution in mass to PM causing exceedances of air quality limit values; ii) the lack of mitigating measures; iii) the increased health risk induced by inorganic (heavy metals) and organic (PAHs) toxics embedded in road dust. In California, a correlation between atmospheric concentrations of heavy metals (Fe, Cu, Zn, and Ni) and the mortality rate due to

ischemic heart diseases was recently found (Cahill et al., 2011). In Stockholm, Meister et al. (2012) estimated a 1.68% increase in daily mortality per $10 \mu\text{g m}^{-3}$ increase in $\text{PM}_{2.5-10}$ concentrations, which include road dust and other coarse-size particles (Pérez et al., 2008). Exposure to road dust particles (below 2.5 microns) was associated with a 7% increment of cardiovascular mortality in Barcelona (estimated excess risk of mortality per $10 \mu\text{g/m}^3$, Ostro et al., 2011). In Sweden particles from road wear caused by studded tires were found to be at least as inflammatory as particles from diesel exhaust (Gustafsson et al., 2008).

Black Carbon (BC) is an operational defined term of carbonaceous material having intense black or dark colour and which absorbs visible light efficiently (Cachier, 1995, WHO, 2012). By applying a correction factor, BC and elemental carbon (EC) may be considered equivalent (and reported as equivalent black carbon, EBC). The different terminology of carbonaceous particles are often interchanging, see Andreae and Gelencser (2006) and WHO (2012) and references there in for a further discussion. BC is a major constituent of fine particulate matter in the atmosphere and responsible for short-wave absorption of solar radiation. In 2007, the Intergovernmental Panel on Climate Change (IPCC) reported that BC contributes $0.2-0.4 \text{ Wm}^{-2}$ to radiative forcing (Foster et al., 2007). However, Ramanathan and Carmichael (2008) reported that radiative forcing by BC contributes 0.9 Wm^{-2} , suggesting that BC may be the second strongest contributor to current global warming, after carbon dioxide. BC is a unique primary tracer for combustion emissions as it has no non-combustion sources, and is stable once released into the atmosphere. The main sources are combustion engines (mainly diesel), residential burning, power stations, agricultural burning and forest fires.

BC has adverse effects on human health. It has been suggested that the main effect is that BC particles carry other toxic components co-released during the combustion or which attach to their surface during atmospheric dispersion (WHO, 2012). BC is useful for examining associations between primary combustion constituents of PM and health outcomes, especially from traffic (Löndahl et al., 2010; Janssen et al., 2011, WHO, 2012).

Ammonia (NH_3) is a reactive species involved in many processes leading to formation of secondary particles and can contribute substantially to the PM concentrations (Reche et al., 2012). As sulphur emissions have been reduced, NH_3 and NO_x have become the main air pollutants responsible for eutrophication and acidification of ecosystems. Further reduction of these compounds is hence needed and also stricter regulation of NH_3 emissions is necessary. The main source of NH_3 emissions is agricultural activity. However, in urban areas traffic has been shown to be a considerable source, mainly due to the use of three-way catalysts in petrol cars. The adoption of selective catalytic reduction (SCR) by the addition of urea or ammonia to diesel exhaust to meet nitrogen oxide emission standards could also result in elevated NH_3 emissions from traffic. Nonetheless, some studies conclude that new vehicles emit lower quantities of ammonia (Bugard et al., 2006; Kean et al., 2009).

1.3 Goal of this Technical Paper

Hak et al. (2010) looked into the contribution of traffic to air quality in Europe targeting 144 cities using a city survey and literature review. The present ETC/ACM Technical Paper is a follow-up on air quality and transport looking at both the emerging and

current problems – in particular traffic as a source of NO₂, primary PM and precursors to PM with emphasis on real world emissions and increased fraction of direct NO₂ emissions in NO_x; contribution of non-exhaust emissions of PM, BC and NH₃ emitted by traffic to concentrations in ambient air. The analysis is based on a systematic literature review of scientific papers from the last 5 years, supplemented by relevant grey literature (reports, communications, etc.) in the investigated area. Information from EU Member States' recent TENS are compiled and included in the analysis.

The assessment is aimed at answering the following questions:

- How much does road transport contribute to air pollution problems with PM, NO₂, BC and NH₃?
- How can the problems be solved by policies and at which scale?

2 Road Traffic Emissions

Even though mitigation strategies and significant reductions in emissions have been in focus for several years many components have concentrations above the limit values, as presented in the introduction. This has partially been linked to road traffic as the dominant contributor to primary emissions in urban environments, hinting at the possibility that the emissions inventories are postulating a continuing downward trend in emissions whereas emissions may not be declining in cities. Harrison et al. (2008a) discusses these issues of PM₁₀ levels not decreasing in cities and raise the question of possibly errors in the emission inventories and that the emissions might not have gone down as much as reported. Winiwarter et al. (2009) have reviewed information on PM emission inventories in Europe and discuss some of the challenges. Inventories still do not always satisfy the needs (e.g. to show emission trends and effect of measures), as there are often data gaps, missing sources and high uncertainties for the applied emission factors. Winiwarter et al. (2009) concluded nevertheless that road transport is one of the best covered source sectors. Smit et al., (2010) did however a review of available traffic emissions models and concluded that several models are not or only partly validated, leading to substantial uncertainties. Beevers et al. (2012) estimate different emission trends using emission factors from different sources and conclude that NO_x emissions in London could be as much as 31 % higher than the current official reported emissions.

Many factors influence the emissions and even if exhaust emissions from each vehicle have been reduced for most components one has seen an increase in overall traffic activity countering this reduction. The car fleet has also changed with respect to technology, fuel type, vehicle size and power. More passenger cars and vehicles on the road could mean more congestion in the cities, also increasing the emissions. Measurement of additional vehicle hours or time delay for a specific road stretch is used as a parameter for congestion (Prosam, 2011, Dijkema et al, 2008). Statistical data to estimate the possible trend in congestion for Europe is not easily available. NO₂ is not legislated under the EURO standard. Conflict of interest might therefore arise as regulations for emissions are for NO_x while the air quality limit values for ambient concentrations are for NO₂.

In this chapter the emissions are highlighted by discussing some of the factors mentioned above. Estimated trends for the emissions using the COPERT model with input from the EC4MACS project are shown in section 2.4

2.1 Emission regulations and real world exhaust emissions

The EURO emission regulations have severely decreased the allowed emissions of several pollutants since the EURO 1 standards. For PM the allowed emissions for diesel passenger cars have gone from 180 mg/km for the EURO 1 to 5 mg/km for the EURO 5 and 6. For NO_x the gap between the diesel and petrol car emissions are closing in for the upcoming EURO 6 regulations.(0.08 g/km for diesel cars and 0.06 for petrol). The worry is that real world emissions might not show the same decrease.

2.1.1 Euro type approval and different test cycles

In EU all vehicles need to pass their type approval according to the EURO standard. For light vehicles this is now done by the use of the New European Driving Cycle (NEDC) giving emissions as g/km. For heavy vehicles the approval is based on an

engine dynamometer testing and the emissions are given as units of the engine work (g/kWh). In EU this engine testing is following the European Transient Cycle (ETC) and European Stationary Cycle (ESC). For the upcoming EURO VI regulations for heavy duty vehicles a new cycle, the World Harmonized Transient Cycle (WHTC) with both cold and hot starts, will be applied.

Critics against the type approval are focusing on the representativeness of the current test cycles for light vehicles compared to real world driving (Weiss et al, 2011) and for heavy duty also the test of the engine might not be well representing the emissions with chassis and real load conditions. Manufacturers may have optimised the emissions to the approval test cycle possibly leading to an even larger difference in the emissions from the approval test cycle and “real world” driving. To register real world driving and afterwards applying it for test purposes is possible. For instance in the ARTEMIS project this was done, building up different driving cycles representative for Europe with urban, rural and motorway driving which can be applied for dynamometer testing. Mobile systems for measuring “on the road” emissions are advancing, opening up for new possibilities to verify and improve the emission estimates. Emission legislation for “In-Service conformity” is introduced for EURO V and EURO VI with the use of Portable Emissions Measurement Systems (PEMS) and are pulling in the right direction for regulating efficiently the heavy duty emissions (Bonnell et al., 2011, EC 2011). PEMS do however not yet include a validated method for PM emission checks (Vermeulen et al., 2012).

2.1.2 Effect of driving cycle and real world emissions

It has been shown that the emissions from “real world” driving with “on the road” measurement using PEMS or use of chassis dynamometer test cycles like the ARTEMIS cycles yield higher emissions than what one would expect following the legislations of the EURO standards. The emissions are higher during urban driving which is characterised by lower speeds and being much more aggressive with more stop and go driving than simulated by the NEDC cycle.

Depending on driving cycles the NO_x emission factors in urban driving for EURO 3 and 4 diesel passenger cars was found to be up to 0.8 g/km which is much higher than the limit value of 0.5 g/km and 0.25 g/km respectively (Alvarez et al., 2008). The emissions from petrol cars have been shown to be low independent of driving cycle and under the limit values (Alvarez et al., 2008, Weiss et al., 2011). Test of petrol EURO 3 and EURO 4 passenger cars did show variations in the NH₃ and NO emissions depending on driving cycle (Heeb et al, 2008), but on average EURO 4 had lower emissions for both components. Consequently, replacing older petrol cars with newer diesel cars would then lead to an increase in the emissions of NO_x manifold.

VTT, Technical research centre in Finland tested the effect of low ambient temperatures on emissions comparing driving in -7°C with that in 23°C. The results were that cold temperatures could increase the NO_x emissions from diesel passenger cars even more in the first kilometres driven adding to the normal cold start emissions (Hagman et al., 2011). This could be an important extra contribution wintertime in colder urban areas of Europe. For the Nordic cities air pollution is typically a larger problem during cold winter periods with unfavourable dispersion conditions.

Large variation in real world emissions is also found for heavy duty vehicles dependent on age of vehicle, size of load compared to the capacity of the engine, driving pattern etc. (COST 346). Shah et al. (2006), used a Mobile Emission Laboratory to study “on the road” emissions from heavy duty vehicles and trucks and concluded that the vehicle operating mode is highly important for the emissions and especially so for NO_x. A Chinese study, showing results of on the road measurements of emissions from trucks conform to the EURO standards, concluded that hydrocarbon, CO and PM_{2.5} emissions have been reduced significantly. The authors of the study did however not see any reduction in NO_x from EURO I to EURO III but did see a reduction for EURO IV trucks (Huo et al., 2012). Durbin et al, (2008) found that newer trucks did show lower NO_x emissions (2003 and 2005 models) but confirmed a trade-off between NO_x and PM emissions. Testing at VTT of city busses using the so called “Braunschweig cycle” conclude that a reduction of NO_x was seen until the EURO III but that Euro IV and V did not reduce NO_x further and even increasing the NO₂ emissions due to increasing NO₂ fraction when particle filters were installed (Hagman et al, 2011). TNO testing of EURO V trucks which were mostly equipped with Selective Catalytic Reduction (SCR) and “Ad Blue” technology for exhaust after treatment showed that some vehicles have low emissions under most driving conditions, but that most trucks are having much greater emissions during “real world”-urban driving with low to medium vehicle speed. They also found that unloaded trucks could have greater absolute NO_x emissions than a loaded truck (Verbeek et al., 2010). A few vehicles on EURO VI technology were also tested giving encouraging results as they showed lower emissions than the EURO V (Vermeulen et al., 2012).

2.1.3 An increase in the NO₂ fraction of NO_x in diesel vehicles

Road side measurements of ambient air have indicated that NO₂ to NO ratio has increased due to the direct emissions (Hueglin et al., 2006, Carslaw et al., 2011). Emission measurements of diesel vehicles show a clear effect of the newest EURO models having larger NO₂ fraction. The explanation is on the one hand the catalytic converters which are used to treat CO and hydrocarbons might also convert NO to NO₂. Secondly, oxidizing particle filters use NO₂ to trap soot releasing also more NO₂ with the exhaust, and as a top layer the “real world” driving cycles seem to increase the NO₂ fraction even more (Alvarez et al, 2008). Petrol cars on the contrary have negligible NO₂ fractions (Heeb et al, 2008). The NO₂ mass fraction of total NO_x, is considered to be in the range of 11-70 % depending on the technology for diesel cars. Typical mass fractions are 30 % for Euro 3 and 55 % for Euro 4 diesel cars, whereas it is only up to 11 % for pre-Euro 3 diesel vehicles (Grice et al., 2009). The NO₂ fraction for heavy duty vehicles is also low for some newer vehicles (10 -14%). The NO₂ fraction seems to be larger for buses compared to other heavy duty vehicles (Grice et al., 2009, Hagman et al., 2011), which might be due to use of the Braunschweig cycle for testing and/or other factors related to the engine like that buses more often have particle filters installed.

2.2 Changes in traffic and vehicle fleet

2.2.1 Increase of number of diesel cars.

Several countries in Europe have vehicle taxations based on CO₂ with the motivation of reducing CO₂ emissions. According to European Automobile Manufacturer's Association (ACEA) 19 EU Member States had this kind of taxation in 2012 (ACEA,

2012). Taxations together with lower diesel fuel prices and better fuel efficiency have made it favourable to buy and/or own diesel cars and have led to an increasing share of diesel cars (dieselisation of the car fleet).

The fraction of diesel passenger cars among the new-registered cars has on average increased from 2001 to 2010. Sharp increase has been seen for instance in Norway (13,3% to 73.9%), Sweden (5% to 49,1%) and Ireland (12% to 62.3%) in the same time period. Some countries however show a decrease as for example the Netherlands from 22% in 2001 to 17.1% in 2010 or Austria that has gone from 71% in 2003 to 49.6% in 2010. A more stable situation but with quite high diesel fractions can be seen for instance in France, Spain and Belgium with an average of 68%, 65% and 72% respectively. One country being clearly different is Greece with very low fractions of 1% to 4% (OFV, 2011) In Greater Athens area circulation of diesel passenger cars has until 2012 been prohibited.

In 2008 64% of the total passenger car fleet of all EU member states were petrol cars and only 34% were diesel (ACEA, 2010). The share of diesel in the overall fuel consumption for the EU-27 has increased from 50 % in 2000 to 63 % in 2010.

2.2.2 Increased number of cars and mileage driven

Based on vehicle stock and activity data delivered by the European project EC4MACS (www.ec4macs.eu), the vehicle activity in urban areas (vehicle kilometres driven under urban conditions) has increased by 27 % in all EU Member States over the last decade (2000-2010), whereas the size of the vehicle population has grown by 15 %.

Increase in number of vehicles was reported for e.g. Prague in Branis (2008) with about doubling the amount of registered vehicles in the city of Prague from 1989 to 2003. For Greater Athens Area a sharp increase of mileage from 1990 to 2009 was mainly seen for passenger cars (about a doubling) and 2-wheelers (about 3.5 times more), with a weaker increase for the heavy and light duty vehicles and busses (Progiou and Ziomas, 2011). In the Paris region the number of passenger cars registered has gone up and down from the 1980 until 2008 but with no overall increase. However, for light commercial vehicles there is about a doubling and the number of motorbikes tripled (Roustan et al, 2011). For Paris an eye catching change is the amount of diesel passenger cars going from around 20% to 70 % between year 1990 and year 2008 see also section 2.2.1

2.3 Traffic induced non-exhaust emissions

Non-exhaust traffic induced particle emissions consist of material that is coming from many different sources like wear of the road surface, tires, breaks, tire studs, and even clutch and other car parts together with other material either deposited from the ambient air or from vehicle loads or tires transporting material from unpaved roads, parking lots and construction sites. Mass can also be intentionally spread onto the road surface like salt and sand for traction control. Material on the shoulder of the road is also contributing to the emissions.

Particle mass size distributions and/or composition of collected road dust sediments has been analyzed and found to contain a large variety of particles originating from soil dust, construction dust, exhaust particles, brakes, tires, and home heating

particles. However the quantity and composition vary largely from street to street, depending on location and local conditions (Chen et al., 2012, and Amato et al., 2011).

The processes leading to the emissions are many and most of them are not well understood or at least not studied in great detail. The processes can be classified in direct wear emissions (tire, road and breaks etc) together with (re-)suspension of dust load, already deposited on the surface, by the tyres and turbulence induced by the vehicles. The strength of the different wear and emission processes are depending on many factors like driving behaviour, speed, vehicle type, tire type, temperature, road wetness, road pavement, grain size and if the pavement is new or old (Mathissen, et al., 2012, Gehrig et al., 2010, Hussein et al., 2008, Gustafsson et al., 2008, Omstedt et al., 2005).

The road surface conditions and wetness will determine the amount of the wear that will be retained and also to what extent the re-suspension will be damped. Road surface water can remove some of the material on the surface by drainage, and splash and spray processes. In the Netherlands light rain fall however was considered not to be a very efficient removal process (Keuken et al., 2010). Other possible removal processes are road maintenance activities like snow ploughing or road cleaning by brushing, vacuuming or washing, see section on mitigation for a discussion on street cleaning.

The road surface wetness is crucial for the non exhaust emissions which hence do not necessarily correlate well with traffic numbers or NO_x concentrations (Norman and Johansson , 2006, Ketzel et al., 2007 and Bukowiecki et al., 2010). This depends mainly on the local meteorological conditions and in a stable dry place one could expect that the emissions are more often correlated with traffic numbers.

Since the emissions are strongly depending on meteorological conditions and site specific parameters, like car fleet and road pavement, emission factors obtained by road side measurements will be site specific and mostly not applicable elsewhere (Ketzel et al., 2007, Pay et al., 2011, Kauhaniemi et al., 2011), making it a challenge to estimate these emissions.

Often non-exhaust emissions are considered to be only in the coarse fraction (PM₁₀-PM_{2.5}) and this fraction is used to estimate the contribution of non- exhaust particles to PM₁₀ levels (Thorpe et al., 2007) but one should not ignore that some of the particles are in the fine particle range – PM_{2.5} or smaller. (Dahl et al., 2006, Mathissen et al., 2011). In a study reported by Kupiainen et al. (2005) as much as 15 % of the PM₁₀ from mechanical abrasion was due to particles in the fine and submicron fractions. Some of the material in the road sediment is also larger than PM₁₀. It is likely that the amount of PM₁₀ or PM_{2.5} could be enhanced by an added sandpaper effect and crushing of such larger particles (Kupiainen et al., 2003).

2.4 Trends in road traffic emissions in the EU27

With the aim to estimate the contribution of road traffic to air quality in European cities, urban emissions of NO_x, NO₂, PM (exhaust and part of the non-exhaust, PM_{2.5}, PM₁₀) and elemental carbon (EC) were calculated for all EU Member States for the years 2000 to 2010. To this aim, vehicle stock and activity data delivered by the EC4MACS project were used as input to the COPERT model.

EC4MACS (www.ec4macs.eu) is a LIFE+ project which provides the modelling framework for integrated assessment of air emission policies in Europe. The project has developed detailed projections of activity, energy consumption and air emissions for all European Member States, based on the PRIMES 2010 baseline scenario. COPERT (www.emisia.com/copert/) is a software tool used world-wide to calculate air pollutant and greenhouse gas emissions from road transport. The COPERT 4 methodology is part of the EMEP/EEA air pollutant Emission Inventory Guidebook for the calculation of air pollutant emissions and is consistent with the 2006 IPCC Guidelines for the calculation of greenhouse gas emissions.

Despite the increased vehicle activity, see section 2.2.2, emissions of the main regulated pollutants from road transport, i.e. NO_x and PM, have been reduced over the last decade. This is the result of advanced after-treatment technology in response to increasingly tight EU emission standards.

Urban NO_x emissions from traffic, calculated with the COPERT model as explained above, has been reduced by 16 % between 2000 and 2010, mainly due to the introduction of Euro 4 and Euro 5 emission standards for passenger cars (both petrol and diesel) in early 2005 and late 2009 respectively. Despite this decrease in total NO_x emissions, NO₂ emissions from road traffic have increased considerably by 85 % over the same period. This is due to the combined effect of fleet dieselisation and the introduction of new technologies, see sections 2.2.1 and 2.1.3.

Total PM_{2.5} and PM₁₀ emissions from road traffic have also decreased by 24 % and 18 % respectively between 2000 and 2010, as calculated with COPERT. These reductions are mainly attributed to the even higher reductions, by 31 %, in exhaust emissions due to the introduction Euro 4 and Euro 5 emission standards, as explained previously. On the other hand, non-exhaust PM emissions have increased by about 30 % due to the increased vehicle activity (urban vehicle kilometres). As a result, the fraction of non-exhaust over total PM emissions has increased considerably from 12 % to 21 % for PM_{2.5} and from 21 % to 33 % for PM₁₀ over the same period. It should be noted that non-exhaust emissions cover primary particles only, i.e. those particles emitted directly as a result of road vehicle tyre and brake wear (road surface wear is not included), and not those resulting from the suspension of previously deposited material. The contribution of non-exhaust emissions is hence underestimated. Urban emissions of elemental carbon have also been reduced by 14 %.

Compared to the other pollutants discussed above, NH₃ road traffic emissions have recorded the highest reductions in 2010, on the order of 41 % of year 2000 levels, as calculated with COPERT. This is due to the combined effect of fleet dieselisation, introduction of new technologies and improved fuel quality. Although NH₃ emissions from petrol cars increased in the 1990s due to the implementation of the catalytic converter in the early Euro vehicles (mainly Euro 1 and 2), they decreased thereafter (for post Euro 2 vehicles). A further reduction in emissions was achieved due to the penetration of low and zero sulphur petrol. On the other hand, emissions of NH₃ from diesel vehicles are lower compared to those of petrol vehicles. This might change in the future when introducing urea based technologies for diesel cars (see section 4.1.4). The above trends are graphically illustrated in Figure 4.

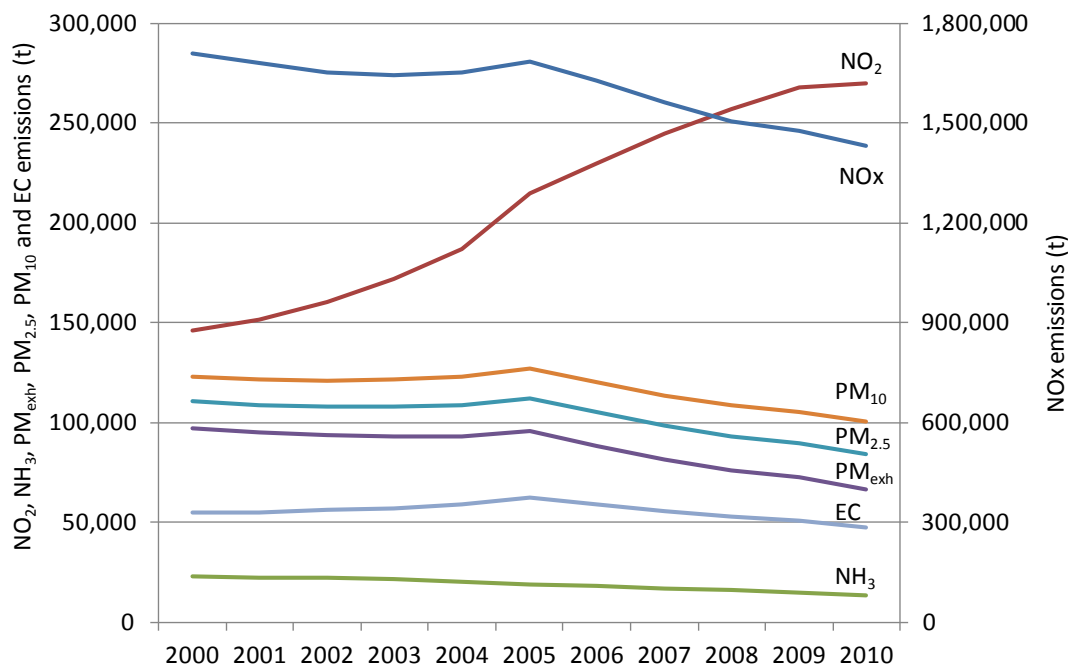


Figure 4 Evolution of road traffic urban emissions of NOx, NO₂, exhaust PM, PM_{2.5}, PM₁₀ and elemental carbon (EC) in the EU in the 2000-2010 period

2.5 Evaporative emissions of organic pollutants

Organic pollutants also escape into the air through fuel evaporation. Evaporative losses may account for a significant proportion of the total hydrocarbon pollution from road transport on hot days when ozone levels are highest. Evaporative emissions occur in several ways:

- Diurnal: evaporation increases as the temperature rises during the day, heating the fuel tank and venting vapours.
- Running losses: the hot engine and exhaust system can vaporise fuel when the car is running, both through the exhaust port but also through the fuel tank and other vehicle parts (e.g., the engine).
- Hot soak: the engine remains hot for a period of time after the car is turned off, and fuel evaporation continues when the car is parked.
- Refuelling: petrol vapours are always present in fuel tanks. These vapours are forced out when the tank is filled.

Automotive diesel is less volatile than petrol so it is mainly petrol cars that are considered to contribute to evaporative emissions. Evaporative control systems exist (carbon canisters) and in the ARTEMIS project it is reported that newer cars show in general less evaporation than older cars or cars without control systems or failure in their fuel systems (ARTEMIS WP 600). The contribution of evaporative emissions to total NMVOC (non-methane VOCs) emissions from road transport has hence decreased considerably since the introduction of carbon canisters.

3 Methods for estimating contribution from traffic to concentrations

A variety of methods for estimating the contribution from traffic are found. Some of the methods can be applied for different components and some are component specific. In this technical paper we focus on PM, BC, NH₃ and NO₂ and only the most applied methods in the reviewed literature will be listed.

3.1 Short description of methods

Time variation analysis: This method consists of analysing measurement data to see if the concentrations are co-variant with traffic numbers either hourly or daily variations, like weekday vs. weekends (Makra et al., 2010). The method gives some indication if traffic is or not an important source but does not exclude other sources that might have similar time variations.

Chemical species like NO₂ might have a time variation pattern that does not necessarily covariate with the traffic numbers. Also non-exhaust contribution might not covariate as there are a large number of interfering factors, especially related to the road surface wetness, influencing the emissions.

Using knowledge about local conditions and possible other sources would make the application of the method more reliable. The method can also be applied and combined with other methods like emission models (Barnpadimos et al., 2011).

Increment method: The method was proposed by Lenschow et al. (2001) and consists of taking road side measurements and subtracting the urban background concentrations. This then assumes that the reference urban background station is not influenced by the local source/ road, which often can be questioned (Thorpe et al., 2007).

Relationships among pollutants: Regression analysis and high positive correlations among pollutants known as resulting from road transport are used to show the significant role of traffic emissions. One method consist of subtracting the regression intercept (of PM and NO_x) from the mean concentration (Liu and Harrison, 2011). Such method is also likely to underestimate the non-exhaust contribution which is known to be less correlated with exhaust emissions. Analysing trends or time variations in fractions like NO₂/SO₂ (Branis, 2008) or combined components like the oxidant, NO₂+O₃, is used to explain the role of traffic emissions in the observed concentration values (Notario et al., 2011, Ha). Even if quantification is not done, it can give valuable insight into the origin of air quality problems (Dongarra et al., 2010).

Dispersion modelling can give quantitative answers to the contribution of specific sources to pollutant concentrations but is dependent on the quality of the emission data applied in the model together with the uncertainties in the dispersion modelling (Gariazzo et al., 2007 a and b).

Receptor modelling is the most widely used method for particle source apportionment. Under the umbrella of receptor models one can find a variety of

models and mathematical procedures like Principal Component Analysis (PCA), Chemical mass balance models (CMB) and Positive Matrix Factorization (PMF). Methods for source apportionment and receptor models are reviewed by Viana et al. (2008). The review article sums up several gaps of the source apportionment receptor modelling techniques and indicate possible future focus in such studies. A major problem in source apportionment analyses is ambiguity in source classification and misclassification of sources. Estimate of uncertainty is in general also not available in most studies. One key issue is also that the size of the data sets often is smaller than what is considered optimal. Finding good tracers to be used in receptor modelling for instance for brake wear tire wear and road dust seems difficult (Kreider et al, 2010, Bukowiecki et al., 2009, Thorpe and Harrison, 2008). There is no unique tracer for non-exhaust contributions.

4 Contribution of traffic to urban air quality

4.1 Findings from the literature review

4.1.1 NO_2

Transport is the dominant sector for NO_x emissions, accounting for over 40 % of the total NO_x emissions in 2010, followed by the energy sector, which contributed about one fifth of the total (EEA, 2012a). Road traffic is the main transport mode that will influence the transport emissions in most cities but contribution from harbours and waterways to the air quality in cities is also a partly non-investigated field (van der Zee et al., 2012, Marr et al., 2007). Harbour areas and local ship traffic might contribute with substantial fractions of the NO_2 concentrations (Lucialli et al., 2007).

From the reviewed literature the overall conclusion is that NO_2 is considered to be dominated by road traffic emissions but actual quantification is seldom done. The general consensus is that road traffic is the main source of NO_2 and NO_x in urban areas especially at traffic sites and time variations of the measured levels follow traffic patterns (Colville et al., 2001, Notario et al., 2011, Branis, 2008, Parra et al., 2009, Makra et al., 2010, Yannopoulos, 2007, Reche et al., 2011a). NO_2 has a large spatial variation, and concentrations fall rapidly with distance to the road (Cape et al., 2004, Parra et al., 2009, Vardoulakis et al., 2011). The authors of the reviewed articles show however a concern for the observed concentration levels and the increased fraction of direct NO_2 emissions and difference between the emissions' regulations and real world driving emissions as discussed in section 2. In the literature from recent years focus has been on the trends of increasing NO_2/NO_x ratio in ambient air and the contributions from secondary and primary NO_2 .

4.1.1.1 *Secondary and primary NO_2 and the NO_2/NO_x ratio*

Being a photochemical specie NO_2 takes part in complex chemical processes with a large number of reactions which include O_3 , VOCs and CO. For urban sites, however, NO_2 chemistry is often only treated as a $\text{NO}-\text{NO}_2-\text{O}_3$ equilibrium which is considered adequate for many situations. Total oxidants with relation to NO_x has been used to estimate the relative importance of the direct emissions compared to the contribution from chemical reactions related to the available background level of O_3 . However as many traffic stations do not measure O_3 , Carslaw and Beevers (2005) propose a method based on the background O_3 and meteorological parameters. At traffic sites the conversion of NO to NO_2 is mostly understood to be ozone limited as NO emissions are abundant.

Höpfner and Lambrecht (2005) showed that in Germany a clear increasing trend in the NO_2/NO_x ratio was observed at traffic stations whereas the rural background did not show any increase. Referring to a study of a German highway the increasing NO_2/NO_x ratio was due to increase in ozone levels until 1997 but then the direct emissions were the dominant cause for the further increase in NO_2 concentrations. In Switzerland at a roadside station an increase was reported from 14 % NO_2 of total NO_x in 1992 to 23% in 2004 which was explained by the changes in direct emissions of NO_2 (Hugelin et al., 2006). Air quality in London and other cities in the UK has been thoroughly studied also with respect to the trends in the NO_2/NO_x ratio. The observed

increase of this ratio is explained with an increase in direct emissions which resulted in ratios of 5-7 % in 1996, increasing to 21-22% in 2009 (Carslaw et al., 2011). Branis (2008) showed the increase of the NO₂/NO_x ratio at several stations in Prague and that the total NO₂ level remained more or less stable with maybe a slight increase in the summer levels. However the author does not explore the causes other than discussing the possible mixed contribution of increasing O₃ levels with possible changes in the NO₂ emissions.

Doing a trend analysis of measurement data from a regional and an urban background station in Rotterdam, Keuken et al. (2009) concluded that the observed increasing trend of the NO₂/NO_x ratio in ambient urban air was mainly due to the O₃-NO_x equilibrium and increasing ozone background concentrations. NO₂ concentrations in the urban background had a decrease in the late 80s but remained rather stable after that with a yearly average of 35 µg/m³. The overall increase of NO₂ direct emissions was found to be 3-4% from 1986 to 2005. Based on these findings the conclusion was that a reduction of emissions at “hotspots” by reducing local traffic would be a more effective abatement strategy than to cut the overall NO_x emissions. Mavroidis and Chaloulakou (2011), looked at trends of NO_x, NO₂, O₃ and the NO₂/NO_x ratio in the Athens Area. Both levels of NO₂ and NO_x had a decreasing trend but NO_x more than NO₂ such that the NO₂/NO_x ratio had increased somewhat in the period analysed. The increase was explained by increasing contribution from secondary photochemical production of NO₂ and not due to primary NO₂ contribution which was seen unchanged from 1998 to 2006. The Athens situation is explained by the fact that diesel passenger cars are not allowed and the use of particle filters and oxidation catalysts for heavy duty vehicles were not common. The effect of air transport, like sea breeze and canalisation of winds, on the variation of oxidant and NO₂ levels, explaining ozone’s contribution to the observed time variation of NO₂ is explored in Notario et al. (2011), concluding that the photochemical processes are dominating the air pollution in the Seville area in south of Spain.

Helsinki has also shown less reduction of NO₂ compared to the NO_x reductions in the time period from 1994-2007 (Anttila and Tuovinen, 2010). Anttila et al. (2011) explored the contribution of primary NO₂ emissions, the NO- NO₂-O₃ equilibrium, and the contribution of regional pollution to NO₂ concentrations. They found that the total primary NO₂ emissions have decreased, even if it is evident that the primary NO₂/NO_x ratio has increased. The NO₂ budget at a heavily traffic influenced site in Helsinki was made of 51% from the photochemical conversion of NO to NO₂, 31% from the direct NO₂ emissions and the rest from regional background, averaged over the years 1994-2004. At another traffic site in Helsinki the contribution from direct emissions was much higher in 2009 than in 2005 and the NO₂ direct contribution was in 2009 of 44% which then was similar to the photochemical contribution of 43%. The authors conclude that there was a clear relative increase in the direct emissions, due to increase in the fraction of diesel vehicles.

Data reported by EEA (2012a) show that at urban background locations NO₂ levels decreased over the last decade at 92 % of the stations registering a statistically significant trend. At traffic locations however 80 % of the stations show NO₂ concentrations with a statistically significant decreasing trend. These trends reflect both the increase in the share of NO₂ in the NO_x emissions from traffic and the shift in the photo stationary state in favour of NO₂ that results from a decrease in NO_x,

without an equivalent decrease in O₃ concentrations (Guerreiro et al., 2010). These are probably also the main reasons for the much lower average decrease in NO₂ concentrations measured over Europe (8 % decrease measured at stations closed to traffic) than the decrease in NO_x transport emission (EEA, 2012a).

4.1.2 PM

Particulate matter have many sources in the city, road traffic is likely to be a large one but also combustion sources from residential heating, power stations or other industries can be important sources. The local direct emissions will contribute together with secondary particles and long range transported particles which make the quantification complex.

PM from exhaust emissions from each vehicle has gone down, but at the same time the traffic volume has increased, also increasing the importance of non-exhaust emissions. From the emission estimate in section 2.4 the relative contribution from non-exhaust emissions has increased from 2 % to 21 % for PM_{2.5} and from 21 % to 33 % for PM₁₀ from 2000 to 2010. The estimate is possibly largely underestimated at some locations as road wear emissions and re-suspension were not included. The German Federal Environmental Agency expects that as much as 80 % of the traffic PM₁₀ emissions will come from non-exhaust emissions in 2020 (Mathissen et al, 2012). Distinguishing between the contributions of non-exhaust and the exhaust emissions to the concentration levels is problematic but essential since different processes are involved and different control measures/strategies are necessary. Also there is a lack of models that include the non-exhaust in a generic manner. Closure of this knowledge gap is a priority.

4.1.2.1 Total contribution from road traffic

In the reviewed articles, which quantify the contribution to PM, road traffic is found to be a relatively large source in most cities (see Table 3 to Table 4). There are some exceptions, for instance Freiburg where it is concluded that local traffic is a minor source to PM₁₀ at a traffic site (Makra et al, 2010). For Helsinki PM_{2.5} is considered to be dominated by long range transport whereas in Oporto traffic contributes with over 60 % to PM_{2.5}. A review article of Viana et al. (2008) from different sites in Europe found traffic exhaust to contribute up to 55% for PM₁₀ and up to 49% for PM_{2.5}. In this review, the contributions range from 13 to 70% for PM₁₀ and 19 to 66% for PM_{2.5}.

For the total contributions it should be noted that the studies listed treat the non-exhaust differently, making it difficult to compare the results. Also the uncertainties of the results depend on the method applied. Many source apportionment studies with receptor modelling define a road traffic contribution, at the same time as they have a mineral /crustal factor that also contains road dust, but a further quantification is not given. In general it seems that the non-exhaust contribution is underestimated. Some studies do however look only at the different road traffic contributions or manage to distinguish the road dust.

It is expected that the sources for the particle concentrations also vary depending geographical location. Nordic cities, where use of wood combustion for domestic heating and where traction control and studded tires are used, give a different source composition than Mediterranean cities with different climate and where Saharan dust

as well as secondary organic aerosols are a more relevant concern. However, no overall conclusion can be drawn from the numbers in the reviewed literature. Factors related to the specific site seem to be more important.

Seasonal variations in the road traffic contributions were also observed. Some sites have higher levels in winter while others in summer. This is due to both the presence of other sources in winter compared to summer and the generally large differences in meteorology. The contribution from non-exhaust emissions is also seasonal in most cases. In many Nordic cities the spring dry up gives large emissions in places where studded tyres still are widely used, this is confirmed in Furusjö et al., (2007).

In quantifying road traffic contributions the focus is often on the local and direct contribution. Emissions from traffic are also leading to SOA, SIA and long range transported particles, but this is often not included in the road traffic contribution estimate. Differentiating traffic as a source of the secondary particles, long range particles and regional background would be useful, because that would show the total contribution of road traffic. The contribution to the regional background was estimated in Hak et al., (2010) by use of the EMEP model. The authors found that traffic contribute with about 15-25 % to the PM₁₀ regional background levels. For PM_{2.5} the contribution increased to 30 % also including traffic SOA. From a study in Switzerland also quite large fractions of 17-24 % road traffic contributions were found for the rural background (Gianini et al. 2012).

4.1.2.2 Non-exhaust contributions

One problem of quantifying road traffic contributions is to be able to separate the non-exhaust traffic induced emissions from other dust sources. Traffic induced non-exhaust emissions include road wear, tyre wear and brake wear particles, but also other kind of particles found in ambient air, see section 2.3 for more detailed information. Elemental composition for tracing these emissions is difficult to define or quantify, because there is an overlap with other soil and crustal sources (both natural and anthropogenic). Some models do quantify brake wear and possibly tyre wear but the road wear and suspension are mostly not quantified or not included. There is also a mix of terminology of non-exhaust, mineral dust, soil, road dust and re-suspension contributions making it difficult to understand to what extent and/or what part of the non-exhaust has been included in the studies.

Higher levels of suspended PM₁₀ mineral matter are found in the urban areas of Southern Europe as compared to Central Europe (Amato et al., 2010b, Putaud et al., 2010 and 2004; Perez et al., 2008; Rodriguez et al., 2007; Ariola et al., 2006; Marelli et al., 2006; Querol et al., 2004, 2001 and 1998). In a comparative study between European sites, Querol et al. (2004), highlighted that in Central Europe, the mineral contribution increases from 3-5 µg/m³ at urban background sites to 4-7 µg/m³ at kerbside sites. In Spain the contribution found to be induced by local traffic suspension was much higher: ranging from 10 to 16 µg/m³. In Sweden the mineral aerosol accounts for 7-9 µg/m³ in urban background but increases dramatically to 17-36 µg/m³ at the traffic sites. Querol et al. (2004) summarized hence that the local road dust emissions account for 9-24 µg/m³ in Sweden, 16 µg/m³ in Spain and 1-5 µg/m³ for the rest of countries studied: England, Switzerland, UK, Germany and Austria. Furusjö et al. (2007) found that re-suspension accounted for 4.6 µg/m³ at an

urban traffic station and $2.1 \mu\text{g}/\text{m}^3$ at a highway location in Sweden.

In our review of articles listed in Table 3 and Table 4 the road dust contribution varies between 8% and 34% for the PM_{10} fraction with generally lower contribution to the $\text{PM}_{2.5}$ fraction and larger to the coarse fraction PMc ($\text{PM}_{10}-\text{PM}_{2.5}$). At the urban background of Barcelona, Amato et al. (2010b) revealed that road dust emissions were responsible in average for 16% of PM_{10} concentrations. In Athens, Karanasiou et al.(2009) resolved road dust, motor exhaust and soil factors estimating the road dust contribution to be up to 34% of PM_{10} of the urban background, for $\text{PM}_{2.5}$ and PMc the percentage was found to be 27% and 53% respectively. For Birmingham the contribution from road dust was 13 % for PM_{10} , 5% for $\text{PM}_{2.5}$ and 27% of PMc (Jones et al., 2008). In Milan the road dust was contributing with 8% of PM_{10} in winter and 28% in summer. Less road dust contribution is reported in winter also in Spain, Greece and UK, due to meteorological conditions with wetter winter climate (Kassomenos et al., 2012). For traffic stations two studies give specific numbers for the non-exhaust and only related to the road dust. Those studies conclude that road dust contributes with 29 % of the PM_{10} at a traffic site in Madrid (Karanasiou et al., 2011) and in Birmingham road dust make up 17 % of the PM_{10} and 40 % of the coarse fraction (PMc) during weekdays (Jones et al., 2008).

Large contributions from road dust, up to 74 % of PM_{10} , were found mainly during spring months in Stockholm, but the contributions were much lower the rest of the year. In the same study break wear was found to contribute with up to 20 % ($1.5 -20 \mu\text{g}/\text{m}^3$) much enhanced in the urban street compared with the motorway site (Furusjö et al., 2007). The Nordic spring outbreak of road dust is due to drying of the roads after the snowmelt together with the sanding and salting of roads during the winter period and the use of studded tyres (Norman and Johansson, 2006; Tervahattu et al., 2006; Areskoug et al., 2004). Studded tyres generate large quantities of PM due to enhanced pavement abrasion (Kupiainen et al., 2005 and 2003).

Table 3 Road traffic's contribution to PM levels at urban traffic sites. PMc is referring to the coarse fraction equal to PM₁₀-PM_{2.5}

| Country /City | Component | Concentration level in ug/m3 | Total Contribution(%) | non-exhaust included? | Year of data | Reference |
|--------------------|-----------|-------------------------------|---|--|-------------------------|--|
| FINLAND | | | | | | |
| Helsinki | PM2.5 | 8-12 | 23-30% | Partly, an ekstra 5-12 % "crustal factor" .LRT with 50-75% in episodes | 1996 og 1997 1999-2007 | Vallius et al(2003) Niemi et al(2009) |
| FRANCE | | | | | | |
| Toulouse | PM10 | | 34 % | Partly,A crustal factor contribute with 25 % some of that road dust. | 1991-2005 | Calvo et al(2008) |
| GERMANY | | | | | | |
| Dresden | PM10 | 41.8 average of selected days | 21 % | Yes, but only local contribution | 2006-2007 | Gnauk et al(2011) |
| Freiburg | PM10 | 12 | Time variations are not traffic dependent | | 1997-2001 | Makra et al(2010) |
| HUNGARY | | | | | | |
| Szeged | PM10 | 45 | Traffic as main source | Yes | 1997-2001 | Makra et al(2010) |
| ITALY | | | | | | |
| Genoa(UB) | PM10 | 38.8 | 34 % | partly, soil source of 33 % , some of that is road dust. | Dec. 2004-Jul. 2005 | Mazzei et al.(2008) |
| Palermo | PM10 | 32.9-46.2 | 32% and 56% in summer 53%- 70% in winter | Yes | Nov 06-Feb08 | Dongorà et al.(2010) |
| = | PMc | 8.3-12 | 60-89% in winter 42%-18% in summer | Yes | Nov 06-Feb08 | = |
| = | PM2.5 | 23.2-34.2 | 50%-63 %in winter 29%-65% in summer | Yes | Nov 06-Feb08 | = |
| NETHERLAND | | | | | | |
| Rotterdam | PM2.5 | 17.5 | 21% | Yes | aug2007-sep.2008 | Mooibroek et al.(2011) |
| POLAND | | | | | | |
| Zabrze | PM10 | 252 | 19 % | Yes, local contribution | Episode in Jan. 2006 | Pastuszka et al.(2010) |
| = | PM2.5 | 228.4 | 22 % | = | = | = |
| PORTUGAL | | | | | | |
| Oporto | PM2.5 | 22.6 summer - 25.9 winter | 56% in summer 66% in winter | Yes | 2003 filters, 2002-2007 | Oliveira et al(2010) |
| = | PMc | 19.3 summer 17.1 winter | 55 % in summer 42% in winter | Yes | 2003 filters, 2002-2007 | = |
| SPAIN | | | | | | |
| Madrid | PM10 | 44 | 60 % | Yes about 29% road dust | 2009 | Karanasiou etal (2011) |
| SWEEDEN | | | | | | |
| Stockholm | PM10 | 37.4 | 51 % | Yes | Mar.2003-Feb.2004 | Furusjö et al (2007) |
| SWITZERLAND | | | | | | |
| Bern | PM10 | 29.4 | 30 % | Yes | Aug.2008-Jul.2009 | Gianini et al.(2012) |
| Zürich | PM10 | | 25 % | Yes , 40 % exhaust 60 % non-exhaust | periods of 2007 | Bukowiecki et al(2010) |
| UK | | | | | | |
| London | PM10 | 32-37 | 49.4% | Yes, partly | 2003-2008 | Liu and Harrison(2011) |
| = | PM2.5 | 19-22 | 50.2% | Yes, partly | = | = |
| = | PMC | 13-18 | 48.3% | Yes, partly | = | = |
| London | PM10 | 34 | 42 - 65 % | Yes | 2002 | Jones et al(2008) |
| = | PM2.5 | 21 | 47 - 59 % | Yes | 2002 | = |
| Birmingham | PM10 | 24.8 | | 17 % , during weekdays | 2005, 2006 | Jones et al(2008) |
| = | PM2.5 | 14.1 | | 4% , during weekdays | = | = |
| = | PMc | 10.3 | | 40%,during weekdays | = | = |

Table 4 Road traffic's contribution to PM levels at urban background sites

| Country /City | Component | Concentration level , ug/m3 | Total Contribution(%) | non-exhaust included? | Year of data | Reference |
|--------------------|-----------|---|--------------------------------|---|----------------------------|---------------------------|
| GERMANY | | | | | | |
| Mülheim-Styrum | PM10 | 25.8 | | 8% road dust | Apr.2007- Mar.2008 | Beuck et al (2011) |
| GREECE | | | | | | |
| Athens | PM10 | 54 | 53 % | Yes including 34% | 2002 | Karanasiou et al. (2009) |
| Athens | PM2.5 | 41 | 54 % | Yes 27% | = | = |
| Athens | PMc | 18 | 61 % | yes 53% | = | = |
| ITALY | | | | | | |
| Genoa | PM10 | 25 | 21 % | partly, soil source of 18 % , some of that is road dust. | Dec. 2004- Jul. 2005 | Mazzei et al.(2008) |
| Lecce | PM10 | 26.3 | 16.5% | NO, large crustal source 49.5% some of that road dust | jan 2007 - jan.2008 | Contini et al (2010) |
| Milan | PM10 | 63 (41.5 in summer 87.4 in winter) | 27 % | partly, 14.5% soil dust | 2001 | Marcazzan et al, 2003 |
| Milan | PM10 | considered high lower in summer than winter | 24% in winter 42% in summer | Yes, include 8% and 28% road dust | 2006 with 2 campaigns | Bernardoni et al.(2011) |
| NETHERLAND | | | | | | |
| Schiedam | PM2.5 | 12.5 -17.5 | 9 % | Yes | aug2007- sep.2008 | Mooibroek et al.(2011) |
| PORTUGAL | | | | | | |
| Oporto | PM2.5 | 18 summer 20.5 winter | 44% summer 60% winter | Yes | 2002-2007 | Oliveira et al(2010) |
| = | PMc | 11 winter 12.5 summer | 33 % summer 12% winter | Yes | 2003 filters, 2002-2007 | = |
| SPAIN | | | | | | |
| Barcelona | PM10 | 40 | 46 % | Yes , 16 % of total PM10 | 2003-2007 | Amato et al(2009) |
| = | PM2.5 | 28 | 51 % | Yes, 8% | = | = |
| Elche | PM10 | 34.5 | 13 % | partly, but local crustal source 21% | Des.2004 to Nov.2005 | Nicolás et al(2008) |
| SWITZERLAND | | | | | | |
| Zürich | PM10 | 21 | 18 % | Yes | Aug.2008- Jul.2009 | Gianini et al.(2012) |
| Zürich | PMc | 5.9 | 70% (57%-86%) in weekdays | Yes only, PMc 35 % of total PM10 | 1998-2009 | Barmpadimos et al(2011) |
| UK | | | | | | |
| London | PM10 | | 24.5% | Yes, partly | | Liu and Harrison(2011) |
| = | PM2.5 | | 33.5% | Yes, partly | | = |
| = | PMc | | 11.6% | Yes, partly | | = |
| London | PM10 | 19 | 9-25 % | | 2002 | Jones et al(2008) |
| = | PM2.5 | 14 | 12 % | | 2002 | = |
| Birmingham | PM2.5 | 11.6 | 18.7 % summer 29% winter | No | May 2007- Apr. 2008 | Yin et al(2010) |
| Birmingham | PM10 | 21.1 | | 13% road dust | 2005, 2006 | Jones et al(2008) |
| = | PM2.5 | 13.3 | | 5 % road dust | 2005, 2006 | = |
| = | PMc | 7.3 | | 27 % road dust | 2005, 2006 | = |

4.1.2.3 Secondary aerosols

Secondary organic aerosols

Many gaseous organic compounds undergo oxidation in the gas phase to yield products, generally oxygenated, that have sufficiently low vapour pressures and that will partition between the gas and aerosol phases. Such compounds are often referred to as semi- or non- volatile, and reside in the aerosol phase as SOA. Thus, in its common usage, SOA refers to the organic component of particulate matter that transfers to the aerosol phase from the gas phase as products of gas-phase oxidation of parent organic species (Kanakidou et al., 2005).

Detection of SOA compounds in ambient air is challenging due to their great variety and presence at very low concentrations (Edney et al., 2003). In the last years, significant improvements have been made in the characterisation of organic aerosols and the understanding of their sources and impacts on the environment. Real-time aerosol mass spectrometers produce organic aerosols mass spectra with high time resolution (Canagaratna et al., 2007).

Urban sources of anthropogenic organic aerosols include mainly aerosols derived from fossil fuel combustion (Schauer et al., 2002; Docherty et al., 2008; Yin et al., 2010), but also from meat cooking, biomass burning, the use of biofuels, and other human activities that lead to the emission of both fossil and modern volatile organic carbon. Consequently, one must separate road traffic-derived SOA from carbonaceous compounds emitted by other urban and regional sources. Laboratory experiments have indicated that organic aerosols can be formed from photooxidation of precursors present in petrol fuel and diesel exhaust (Miracolo et al., 2010; Odum et al., 1997). The exhaust fumes from petrol vehicles have been demonstrated to contribute significantly to the production of SOA (Bahreini et al., 2012). They estimated that SOA from petrol exhaust emissions may reach 4 Tg/year, which is about 16% of recent global estimates of biogenic SOA (Andreae and Rosenfeld, 2008; Monks et al., 2009) and they concluded that a decrease in the emission of organic species from petrol engines may significantly reduce SOA concentrations on local and global scales.

The quantification of how SOA contributes to PM is a question of special interest among the scientific community. SOA accounts for the largest contribution to organic aerosols (Jimenez et al., 2009). In Europe, this SOA contribution accounts for about 30-80% of the total organic aerosols in the PM_{2.5} fraction in urban environments (Salma et al., 2004; Lanz et al., 2007; Lonati et al., 2007; Grivas et al., 2012). In Barcelona, Mohr et al. (2012) quantified 41% fossil carbon in the organic aerosols in the PM₁ fraction, which is lower than expected. This quantification was obtained by Positive Matrix Factorization (PMF) conducted on the organic aerosol data matrix measured by an aerosol mass spectrometer. These results were in agreement with those derived from the radiocarbon (¹⁴C) analysis for organic carbon and source apportionment (Minguillón et al., 2011).

Secondary inorganic aerosols

Reactive nitrogen compounds (RNCs) such as nitric oxide (NO), nitrogen dioxide (NO₂), and ammonia (NH₃) have a substantial impact on urban air quality. NO and NO₂ support the formation of ozone, and NH₃ is the major alkaline forming gas in the atmosphere. NH₃ reacts primarily with acidic species and neutralizes a substantial part of the acid produced by sulphur oxides (SO_x), nitrogen oxides (NO_x) and hydrochloric acid (HCl). These processes form secondary particulate matter including ammonium sulphate, ammonium nitrate and ammonium chloride, and as a result, ammonium (NH₄⁺)-bearing species are major components of atmospheric aerosols (Asman et al., 1998). The NH₄⁺ aerosols contribute significantly to aerosols mass (PM_{2.5}, PM₁₀) (Figure 5) and have implications for human health (Brunekreef and Holgate, 2002; Verma et al., 2009) and for the exceedance of PM₁₀ daily and annual limit values (Reche et al., 2011b). Ammonium nitrate and its gaseous counterparts ammonia and nitric acid also play a key role in acidifying and eutrophying deposition over Europe. See also the section on ammonia. The understanding of the formation, transport and fate of these components is crucial to assess their role in air quality of cities.

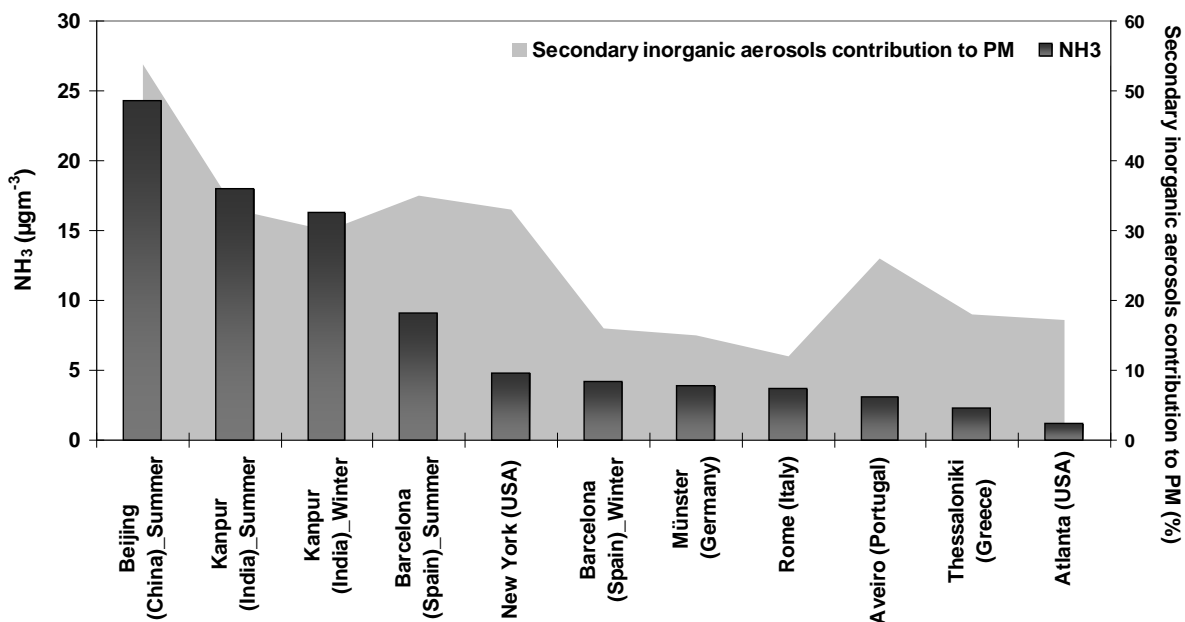


Figure 5 Percentage of secondary inorganic particles contribution to PM and NH₃ concentrations (µgm⁻³) in different cities (Reche et al., 2012 and references therein).

Several articles about source apportionment reviewed in our study show quite large contributions of secondary particles (Beuck et al,2011, Amato et al,2009 b , Oliveira et al, 2010, Bernardoni et al. 2011, Karanasiou et al., 2011, Liu and Harrison, 2011, Nicolás et al., 2008). Secondary inorganic aerosol (SIA) contributes with a significant part (Putaud et al., 2004).

In previous studies nitrate was found to be the dominant secondary particle component in western Europe in the form of ammonium nitrate (NH₄NO₃) (ten Brink et al., 1997; Schaap et al., 2002). Sodium nitrate was reported larger in northern and southern Europe (e.g. Pakkanen et al., 1999).

In Barcelona 70 % of the secondary nitrate and organics was associated with traffic (Ostro et al., 2011). Nitrate concentrations is found to vary according to season and in Revuelta et al. (2012) both London and Madrid showed nitrate reductions of around 20 % during summer.

In Switzerland an increase of nitrate was found (Gianini et al., 2012) between 1998/1999 to 2008/2009 whereas sulphate, EC and many trace elements decreased. Gianini et al. (2012) find the increase in nitrate remarkable but not fully understood. They discuss a possible relation to decreasing sulphate concentrations or other hypothesis like increased formation of nitric acid during night, and hence nitrate, due to increasing ozone levels. Increasing primary NO₂ emissions might also work in the same direction as they might contribute to increased ozone formation. They conclude however that further NO_x reduction is anyway necessary for lowering the nitrate concentrations in PM₁₀ in Switzerland. As mentioned, NO_x contributes to secondary particle formation in urban atmospheres, especially through interaction with NH₃. Reducing nitrate by reducing NO_x emissions was also the conclusion of Millstein et al. (2008) investigating fine particle nitrate response to weekly changes in emissions at four US urban sites.

Pay et al. (2012) used the CALIOPE-EU modelling system and EMEP measurements for studying indicators of SIA production. The conclusion was that SIA formation is most often SO₂ and HNO₃ gaseous precursors limited, especially over North Western Europe where agriculture is the largest source of ammonia emissions. Regulation of NO_x and SO₂ rather than NH₃ would hence be more efficient for reduction of SIA.

4.1.2.4 Ultrafine particles

Among the numerous components of vehicle-produced pollution, ultrafine particles (UFPs, diameter < 100 nm) are of interest as a potential health threat to populations living and working near major roadways. While the ultrafine fraction has a minimal contribution to current regulatory mass-based particulate matter measures (PM_{2.5} or PM₁₀), UFPs typically dominate the particle number size distribution. Given their small size, UFPs have been shown to efficiently penetrate the respiratory system and even transfer to extrapulmonary organs, including the central nervous system (Oberdörster et al., 2004). The majority of particulate matter emitted through vehicle tailpipe exhaust occurs in the PM₁ size range, with mass median diameter generally between 100 and 200 nm (Kleeman et al., 2000) and number median diameter around 20 nm (Kittelson et al., 2004). At a near-road location, UFP were observed to have a magnified response to roadway emissions in comparison to larger particle sizes (Molnar et al., 2002). Other studies have observed a strong spatial gradient associated with UFPs, exponentially decreasing with distance from major roadways (e.g., a horizontal gradient of 8% drop per 10 m distance (Hagler et al., 2009). These results suggest that road traffic is a major source of UFP and that UFP heavily influence air concentrations in the nearby vicinity of major roadways.

4.1.3 Black Carbon

Black Carbon (BC) is strongly associated with road traffic emissions, which have been found to govern the daily cycle of this parameter in several studies throughout Europe (Saha et al., 2009; Rodríguez et al., 2008; Pérez et al., 2010; Lyamani et al., 2011).

Airborne BC exhibits strong spatial variability, being present at high concentrations in the vicinity of traffic sources, with a lognormal dispersion within 200m of highways (Zhu et al., 2002). Based on data gathered from 5 studies covering 9 cities in 5 countries, the contribution of BC to PM10 levels account for 3-10 % in urban background environments and may reach 24 % at kerbsides, the contribution to PM2.5 is around 5-20 % at urban background sites (see Table 5 and references therein).

BC levels can increase about 0.7-1.3 $\mu\text{g}/\text{m}^3$ at traffic rush-hours at urban background sites during weekdays, while this increase is about 2.5-5 $\mu\text{g}/\text{m}^3$ at kerbsides; daily average values can be up to 0.6 $\mu\text{g}/\text{m}^3$ higher during weekdays than at weekends at urban background sites and up to 3 $\mu\text{g}/\text{m}^3$ at kerbsides (Reche et al., 2011b). A geographical dependence is not observed and thus this increase mainly depends on the traffic volume in each studied site.

Table 5: Black Carbon contribution to PM in different fractions and stations types

| Country /City | PM fraction | Contribution | Station type | Reference |
|------------------------|-------------|---------------|--------------|----------------------------|
| Austria | | | | |
| Vienna | PM10 | 9.80 % | Urban | Hitzenberger et al. (2001) |
| Finland | | | | |
| Helsinki | PM2.5 | 19 % | Urban | Pakkanen et al. (2000) |
| Spain | | | | |
| Barcelona | PM10 | 6 % | Urban | Reche et al. (2011) |
| Huelva | PM10 | 3 % | Urban | = |
| Santa Cruz de Tenerife | PM10 | 4 % | Urban | = |
| Santa Cruz de Tenerife | PM10 | 8 % | Urban | Rodriguez et al. (2008) |
| = | PM2.5 | 13 % | = | = |
| = | PM1 | 21 % | = | = |
| Switzerland | | | | |
| Bern | PM10 | 13 % | Kerbside | Reche et al. (2011) |
| Lugano | PM10 | 8 % | Urban | = |
| UK | | | | |
| London | PM10 | 10 % | Urban | Reche et al. (2011) |
| = | PM10 | 24 % | Kerbside | = |
| Birmingham | PM2.5 | 10 % | Urban | Harrison et al.(2008) |
| | PM2.5 | 20 % | Kerbside | = |

BC levels vary proportionally to those of traffic-related gaseous pollutants (CO, NO and NO₂), and due to this high correlation, one may suppose that monitoring the levels of these gaseous pollutants would be enough to extrapolate exposure to BC concentrations. However, the BC/CO, BC/NO₂ and BC/NO ratios vary widely depending on the specific characteristics of urban sites, as a function of the distance to traffic, the vehicle fleet composition and age, and the influence of other carbonaceous sources such as biomass burning (Reche et al., 2011b).

BC may be proposed as a vehicle exhaust emissions tracer to be potentially included in urban air quality control networks even if BC emissions are not a major contributor

to the exceedances of air quality limit values (EEA, 2012b). BC levels have been used as an indicator of air quality benefits of traffic restriction policies (Invernizzi et al., 2011).

4.1.4 Ammonia

Because of the numerous atmospheric interactions of ammonia (see section 4.1.2.3), the identification of its emission sources in urban environments is a complex task. Road traffic is recognized to be a NH_3 source from a local perspective in urban areas (Sutton et al., 2000; Battye et al., 2003). NH_3 has been identified to be formed during the reduction of different nitrogen oxides in the air of highway tunnels (Moeckli et al., 1996; Fraser and Cass, 1998; Kean et al., 2009 Emmenegger et al., 2004); at roadsides (Cape et al., 2004); and in urban ambient air (Perrino et al., 2002; Moya et al., 2004).

Although the quantification of NH_3 emissions from traffic in urban environments is a complex issue, studies have evidenced the link between road traffic volume and ambient NH_3 levels (Perrino et al., 2002; Gadsdon and Power, 2009; Reche et al., 2012). In a typical Mediterranean environment, a reduction of NH_3 levels by more than 55% with increasing distance (50 m) to the direct emissions from traffic was observed (Reche et al., 2012), levels varied from $6.8 \mu\text{g m}^{-3}$ immediately in the road to $3 \mu\text{g m}^{-3}$ at 50 meters away from it.

The implementation of three-way catalysts converters (TWC) in petrol vehicles has been a significant initiative for the abatement of pollutants like CO, hydrocarbons, and NO. A subsequent formation of NH_3 has been demonstrated from TWCs (Pierson and Brachaczek, 1983; Baum et al., 2001; Durbin et al., 2002; Huai et al., 2003; Heeb et al., 2006), but also the emissions of NH_3 from petrol cars have been reduced for newer car models (Heeb et al., 2008), see also section 2.4. Heeb et al. (2011) studied the effects of a urea-based selective catalytic reduction (SCR) to the emissions of a diesel engine, obtaining that the investigated SCR technology substantially lowered NO and NO_2 emissions, while NH_3 levels were comparable to those of TWC vehicles.

In spite of the contribution of NH_3 to the PM concentrations, to date, no significant regulatory effort has been made in Europe to control NH_3 emissions. In Spain, a new regulation recently implemented in January 2011 (R.D. 102/2011), established the requirement to measure ambient ammonia levels at five rural background stations covering the country and in one traffic site in cities with more than 500000 inhabitants. The Critical Level of NH_3 in ecosystems was set to $8 \mu\text{g m}^{-3}$ as an annual average, although recent studies reported important effects on vegetation when levels are above $3 \mu\text{g m}^{-3}$ (Cape et al., 2009). For reference, typical urban air NH_3 concentrations in Barcelona (Spain) range between 0.5 and $8 \mu\text{g m}^{-3}$ (Reche et al., 2012). The issues implicated in addressing the critical level were summarised in the proceedings of a workshop held in Edinburgh in December 2006 under the sponsorship of the United Nations Economic Commission for Europe (UNECE) (Cape et al., 2008; Sutton et al., 2009).

National policies and control strategies on NH_3 might be implemented to achieve a reduction of ammonia-derived secondary aerosol levels. However, it is difficult to estimate the effect on road traffic-related aerosols as other urban sources may have a

major impact on NH_3 emissions, such as sewage systems or waste disposals (Meng et al., 2011; Reche et al., 2012).

The role of NH_3 enhancing SOA formation by means of the interaction with gas-phase organic acids forming condensable salts has been reported in previous studies (Na et al., 2007).

4.2 Findings from the Time Extension Notifications

A number of Member States (MS) have experienced difficulty meeting the requirements of Directive 2008/50/EC on ambient air quality and cleaner air for Europe. A requirement in case of exceedance of a limit value (plus margin of tolerance, as applicable) is that MS submit information in an Air Quality Plan, on the quality of air and the actions to improve it. A significant proportion of the MS have reported exceedances of the limit values both for NO₂ and for particulate matter (PM₁₀). As a result they have introduced additional measures to address these problems and some MS have submitted Time Extension Notifications (TEN) to the European Commission (EC) according to Art. 22 of the above Directive, detailing what has been done and documenting the path to compliance at the end of the time extension period.

Hereafter we present a review of the source apportionment included in the TENs for compliance with the PM₁₀ and NO₂ limit value. The following sources of information have been used:

1. Original TENs submitted to EC (available on CIRCA):
http://ec.europa.eu/environment/air/quality/legislation/time_extensions.htm
2. Database compiled by Moosmann (2010), collecting the information from the TENs for PM₁₀:
http://circa.europa.eu/Members/irc/env/ambient/library?!=/management_database&vm=detailed&sb=Title

4.2.1 PM₁₀ assessments in TENs

Twenty European MS submitted TENs for compliance with the PM₁₀ limit value to the European Commission. The areas with PM₁₀ levels exceeding the daily or annual limit value are divided into zones that may include an entire city, an agglomeration or several separate cities. The number of countries confirms that a majority of European member states still have not attained the PM₁₀ limit values, which were defined already in Directive 1999/30/EC, although they have been mandatory since 1 January 2005.

The information to be provided in the TENs is specified in COM(2008)403 and in the Staff Working Paper accompanying that Communication. Member States must provide information on the origin of pollution contributing to the exceedance. A quantitative source apportionment for the exceedance situation in the reference year is required. The source apportionment must reflect regional, urban and local contributions within the MS. The urban and local contributions are further subdivided in order to identify any significant sources such as road traffic contributions. For PM₁₀, it is also important to indicate significant natural sources.

Table 6 lists the countries and the number of zones that have been considered for assessing the contribution of road traffic to PM₁₀. We have only considered those zones with sufficiently detailed source apportionments for a number of cities larger than 250 000 inhabitants. There are countries, however, where the forms have not been filled in with sufficient thoroughness or just very rough source apportionments were supplied, in the table they appear as they have 0 of zones for source apportionment. Some source

apportionments reported could not be used in this assessment due to incompatibility of the information. This same situation was also reported in Hak et al. (2010).

Table 6 Number of zones with sufficient information to assess traffic contribution to PM_{10} sorted by country. Information obtained from UBA database.

| Country | Thereof cities > 250000 inh. | Number of source apportionments |
|---------------------|------------------------------|---------------------------------|
| Austria (AT) | 3 | 8 |
| Belgium (BE) | 1 | 1 |
| Bulgaria (BG) | 0 | 0 |
| Cyprus (CY) | 0 | 0 |
| Czech Republic (CZ) | 3 | 6 |
| Germany (DE) | 9 | 20 |
| Denmark (DK) | 1 | 2 |
| Greece (EL) | 2 | 2 |
| Spain (ES) | 2 | 2 |
| France (FR) | 0 | 0 |
| Hungary (HU) | 1 | 8 |
| Italy (IT) | 7 | 10 |
| Latvia (LV) | 1 | 1 |
| Malta (MT) | 0 | 0 |
| Netherlands (NL) | 0 | 0 |
| Poland (PL) | 4 | 4 |
| Portugal (PT) | 2 | 2 |
| Romania (RO) | 0 | 0 |
| Slovakia (SK) | 0 | 0 |
| United Kingdom (UK) | 4 | 5 |

The response to the questions on source apportionment was not expressed in a standardized way in the TENs provided by the various countries. The most often applied responses were to fill in concentrations and contributions from the individual sources to concentrations, percentage contributions of sources that sum up to 100% within each sub-sector (i.e. regional, urban and local part), or percentage contributions of the sources that sum up to a total of 100%. The latter, being the most frequently used method, was also utilized here. Those forms which supply the information in other ways were normalized so that sources sum up to a total of 100%. Apportionments which consisted in concentration contributions were converted to percentage contributions. This methodology is the same used in the ETC/ACC Technical Paper by Hak et al. (2010) for PM_{10} assessments.

The TENs also provide information about the type of station (urban, suburban, traffic, industrial, background, etc.). In the tables below the information regarding source apportionment has been classified according to the type of station. For some of the cities the source apportionment was estimated for a set of stations that include several types.

Table 7 Urban and local traffic contribution to PM10 concentrations at Urban Traffic (UT) sites. Information obtained from source apportionments in TENs to the EC, compiled by UBA. The information was normalized so the sources sum up to a total of 100%. r: regional; u: urban; l: local

| Description of the zone | | Description of the exceedance | | | Source apportionment | | | | |
|-------------------------|---------------------------------------|-------------------------------|-----|-----------------|------------------------|----|----|--------------------------|----|
| | | | | | Total contribution (%) | | | Traffic contribution (%) | |
| Member State | City or municipality | Zone code | LV | No. Of stations | r | u | l | u | l |
| AT | Wien | AT09 | d | 1 UT | 75 | 13 | 13 | 6 | 6 |
| AT | Wien | AT09 | d | 1 UT | 60 | 10 | 30 | 5 | 22 |
| BE | Brussels | BEB10a | d | 1 UT | 52 | 21 | 27 | 16 | 13 |
| CZ | Praha | CZ010 | a | 3 UT | 44 | 11 | 45 | 1 | 23 |
| CZ | Brno | CZ0622 | a | 1 UT | 56 | 24 | 20 | 7 | 17 |
| CZ | Brno | CZ0622 | a | 1 UT | 56 | 24 | 20 | 17 | 15 |
| CZ | Brno-Tuřany; Brno | CZ0622 | d | 1 UT | 65 | 24 | 11 | 17 | 10 |
| DE | Bremen | DEZEIX003A | d | 1 UT | 53 | 6 | 42 | 2 | 42 |
| DE | Augsburg | DEZDXX0002A | d | 1 UT | 56 | 8 | 36 | 1 | 36 |
| DE | Augsburg | DEZDXX0002A | d | 1 UT | 54 | 22 | 24 | 1 | 24 |
| DE | München | DEZDXX0001A | a | 1 UT | 63 | 16 | 22 | 1 | 22 |
| DE | München | DEZDXX0001A | d | 2 UT | 44 | 11 | 44 | 0 | 44 |
| DE | München | DEZDXX0001A | d | 1 UT | 44 | 11 | 44 | 0 | 22 |
| DE | Dortmund | DEZJXX0008A | d | 1 UT | 64 | 7 | 29 | 3 | 29 |
| DE | Düsseldorf | DEZJXX0009A | d | 1 UT | 53 | 5 | 42 | 4 | 42 |
| DE | Düsseldorf | DEZJXX0009A | d | 1 UT | 57 | 6 | 37 | 4 | 37 |
| DE | Essen | DEZJXX0006A | d | 1 UT | 61 | 13 | 26 | 3 | 26 |
| DE | Essen | DEZJXX0006A | d | 1 UT | 68 | 13 | 19 | 6 | 19 |
| DE | Wuppertal | DEZJXX0002A | d | 1 UT | 66 | 3 | 31 | 1 | 31 |
| DE | Wuppertal | DEZJXX0002A | d | 1 UT | 71 | 3 | 25 | 2 | 25 |
| DE | Duisburg | DEZJXX0007A | d | 1 UT | 60 | 31 | 9 | 4 | 9 |
| DE | Duisburg | DEZJXX0007A | d | 1 UT | 64 | 10 | 26 | 6 | 0 |
| DE | Duisburg | DEZJXX0007A | d | 1 UT | 69 | 6 | 25 | 3 | 0 |
| DE | Leipzig | DEZNXX0001A | d | 1 UT | 52 | 22 | 26 | 6 | 26 |
| DK | Copenhagen | DK0001 | d | 1 UT | 75 | 10 | 15 | 10 | 15 |
| DK | Copenhagen | DK0001 | d;a | 1 UT | 59 | 7 | 34 | 7 | 34 |
| ES | Abanto / Barakaldo / Basauri / Bilbao | ES1602 | d | 1 UT | 44 | 34 | 22 | 21 | 15 |
| HU | Budapest | HU0001 | d;a | 1 UT | 42 | 21 | 37 | 15 | 25 |
| IT | Firenze | IT0905 | a | 1 UT | 35 | 35 | 30 | 18 | 20 |
| IT | Firenze | IT0905 | d | 1 UT | 20 | 55 | 35 | 22 | 14 |
| IT | Napoli//Caserta | IT0601 | d | 2 UT | 45 | 15 | 40 | 6 | 16 |
| IT | Catania (Sicilia) | IT10A7 | a;d | 1 UT | 36 | 50 | 14 | 32 | 10 |

| | | | | | | | | | |
|----|--|--------|---------|-----------------------|----|----|----|----|----|
| IT | Palermo | IT19A1 | a;d | 1 UT | 35 | 51 | 14 | 39 | 10 |
| IT | Messina | I19E07 | d | 1 UT | 38 | 49 | 13 | 37 | 8 |
| LV | Riga | LV0001 | d; a | 1 UT | 51 | 22 | 27 | 13 | 27 |
| PT | Lisboa; Cascais; Odivelas | PT3001 | d;a | 1 UT | 50 | 6 | 44 | 3 | 16 |
| UK | Glasgow Urban Area | UK0024 | d | 1 UT | 29 | 17 | 54 | 7 | 54 |
| UK | Greater London Urban Area | UK0001 | d;a | 1 UT | 39 | 27 | 34 | 13 | 34 |
| | | | | | | | | | |
| PT | Porto; Espinho; Matosinhos;V.Conde | PT1004 | d;a | 1 UT 1 ST | 34 | 27 | 39 | 2 | 20 |
| PL | Area of Krakow City: Kraków | | d;a | NA but prob. UT | 15 | 35 | 50 | 19 | 20 |
| UK | West Midlands Urban Area (Birmingham) | UK0002 | d | NA but prob. UT | 39 | 30 | 31 | 9 | 31 |

Table 8 Urban and local traffic contribution to PM10 concentrations at Urban and Suburban Background sites (UB and SB). Information obtained from source apportionments in TENs to the EC compiled by UBA. The information was normalized so the sources sum up to a total of 100%. r: regional; u: urban; l: local

| | | | | | Source apportionment | | | | |
|-------------------------|---------------------------|-------------------------------|-----|-----------------|------------------------|----|----|--------------------------|----|
| Description of the zone | | Description of the exceedance | | | Total contribution (%) | | | Traffic contribution (%) | |
| Member State | City or municipality | Zone code | LV | No. Of stations | r | u | l | u | l |
| HU | Budapest | HU0001 | d | 1 UB | 54 | 8 | 38 | 8 | 27 |
| HU | Budapest | HU0001 | a;d | 1 UB | 43 | 21 | 36 | 15 | 21 |
| IT | Firenze | IT0905 | d | 1 UB | 20 | 45 | 35 | 14 | 5 |
| PL | Łódź | PL.10.01.a.03 | a | 1 UB | 17 | 1 | 82 | 0 | 1 |
| PL | Zgierz | PL.10.01.a.03 | a | 1 UB | 24 | 1 | 75 | 0 | 2 |
| PL | Gdańsk, Gdynia and Sopot | PL.22.01.a.03 | a | 1 UB | 13 | 4 | 84 | 1 | 20 |
| UK | West Yorkshire Urban Area | UK0004 | d | 1 UB | 36 | 21 | 44 | 6 | 0 |
| | | | | | | | | | |
| AT | Steiermark without Graz | AT06 | d | 3 UB 2 SB | 55 | 35 | 10 | 10 | 7 |
| CZ | Brno-Tuřany; Brno | CZ0622 | d | 1 SB | 65 | 24 | 11 | 12 | 10 |
| HU | Budapest | HU0001 | d | 1 SB | 66 | 16 | 16 | 7 | 3 |
| HU | Budapest | HU0001 | d;a | 1 SB | 45 | 22 | 34 | 16 | 22 |

Table 9 Urban and local traffic contribution to PM10 concentrations at Urban and Suburban Industrial sites (UI and SI). Information obtained from source apportionments in TENs to the EC compiled by UBA. The information was normalized so the sources sum up to a total of 100%. r: regional; u: urban; l: local

| Description of the zone | | Description of the exceedance | | | Source apportionment | | | | |
|-------------------------|----------------------|-------------------------------|----|-----------------|------------------------|----|----|--------------------------|---|
| | | | | | Total contribution (%) | | | Traffic contribution (%) | |
| Member State | City or municipality | Zone code | LV | No. Of stations | r | u | l | u | l |
| AT | Wien | AT09 | d | 1 UI | 71 | 12 | 18 | 6 | 4 |
| DE | Duisburg | DEZJXX0007A | d | 1 SI | 83 | 11 | 6 | 5 | 0 |
| DE | Duisburg | DEZJXX0007A | d | 1 UI | 71 | 6 | 23 | 2 | 0 |

Table 10 Urban and local traffic contribution to PM10 concentrations at mixture of sites. Information obtained from source apportionments in TENs to the EC compiled by UBA. The information was normalized so the sources sum up to a total of 100%. r: regional; u: urban; l: local

| Description of the zone | | Description of the exceedance | | | Source apportionment | | | | |
|-------------------------|--|-------------------------------|-----|-----------------------|------------------------|----|----|--------------------------|----|
| | | | | | Total contribution (%) | | | Traffic contribution (%) | |
| Member State | City or municipality | Zone code | LV | No. Of stations | r | u | l | u | l |
| AT | AG Graz | AT60 | a | 1 UT 1 UB | 50 | 25 | 25 | 10 | 18 |
| AT | AG Graz | AT60 | d | 1 UT 1 UB 1SB 1 ST | 37 | 39 | 24 | 16 | 15 |
| CZ | Praha | CZ010 | d | 6 UT 4 SB | 48 | 7 | 45 | 1 | 14 |
| EL | Athens; Maroussi; Lykovrissi; Piraeus | EL0003 | d;a | 3 UT 2 UB | 23 | 27 | 50 | 2 | 27 |
| EL | Echedoros; Eleftherio Kordelio; Thessaloniki; Kalamaria | EL0004 | d;a | 2 UT 2 UI | 23 | 28 | 49 | 2 | 20 |

Regional, urban and local contributions (all sources) to total PM

In summary, at urban traffic sites the regional contribution ranges between 15 and 75%, with an average value of 51%; the total urban contribution ranges between 3 and 55%, with an average value of 20%; and the total local contribution ranges between 9 and 54%, with an average value of 30%. This assessment is based on a total of 38 source apportionments for sites classified as urban traffic stations plus one suburban traffic site (Porto), and two sites (Kraków and Birmingham) not classified but according to their distribution of sources probably corresponding to urban traffic sites.

At urban background sites (including 4 suburban background sites), the regional contribution is about 40%, ranging from 13% to 66%, the total urban contribution is estimated to contribute in a 18% as average, ranging from 1 to 45%, and finally, the total local contribution has values between 10 and 84%, with an average of 42%. A total of 7 sites were classified as urban background stations, one was classified as urban background and suburban background, and three places were classified as suburban background. A total of 11 source apportionments were used for the assessment at urban background sites.

At industrial sites (two urban and one suburban), the regional contribution ranges between 71 and 83%, with an average value of 75%; the total urban contribution is about 10% and has values between 6 and 12%; and the total local contribution is about 15%, ranging between 6 and 23%.

A total of 6 source apportionments correspond to a set of different type of stations. For that set of municipalities, the total regional contribution is about 31%, ranging between 19 and 50%, the total urban contribution is about 27% and has values between 7 and 39%; and the total local contribution is 41% as average, and ranges between 24 and 50%.

Contribution of urban traffic

The road traffic within the urban area was assessed to contribute 10% to PM10 concentrations measured at urban traffic sites, as an average of 41 urban traffic sites in 30 municipalities. The urban traffic contribution at urban traffic sites ranges between 0.4 and 39%. The lowest contribution corresponds to the city of München (Germany), and the largest one to the city of Palermo (Italy). Contributions below 3% of the cities' own traffic to urban traffic sites may be considered suspicious (Hak et al., 2010). If the sites with low contributions are left out (12 sites in 7 cities with <3% from the urban traffic to concentrations at UT sites), the average for the 29 remaining sites becomes 13% with the range 3 to 39%.

The urban traffic contribution to urban background sites is about 8%, as an average of 11 urban background sites in 8 cities; and it ranges between 0 and 16%. The null urban traffic contribution corresponds to the city of Łódź (Poland), and the largest one to the city of Budapest (Hungary). Leaving out the sites with low contribution from the urban traffic (3 sites in 3 cities with <3%), the average contribution of urban traffic to UB stations becomes 11% with the range 6 to 15%.

Looking at the contribution from the road traffic within the urban area at the 29 UT sites and the 8 UB sites that had reported consistent data, the ratio between urban traffic contribution to PM10 concentrations at UT and UB sites is 1.2. The cities of Budapest (Hungary) and Firenze (Italy) have provided source apportionment for UT and UB stations. The ratio between urban traffic contribution at UT and UB at these two cities is 1.3 and 1.5, respectively. Moussiopoulos et al. (2004) reported an average ratio of 1.36

(sd=0.25) between PM10 concentrations at UT and UB sites in the same city (number of station pairs: 86).

At industrial sites, the average contribution of urban traffic is about 4% and it ranges from 2 to 6 %, the lowest value corresponds to the city of Duisburg (Germany) and the largest to the city of Wien (Austria).

Contribution of local traffic

The contribution of local traffic at urban traffic sites, meaning in the immediate proximity of receptor or monitoring sites accounts for 22% of the PM10 concentrations, as an average at 41 sites at 30 cities. The range is between 0 and 54%, the null local traffic contribution corresponds to the city of Duisburg (Germany) and the largest contribution to the city of Glasgow (United Kingdom). A contribution of 0% of local traffic in an urban traffic station is suspicious. Leaving out the two sites from Duisburg with null traffic contribution, the average local traffic contribution at urban sites become 21%, with range from 6 to 54%. The lowest contribution corresponds now to the city of Wien (Austria). The large range reflects that the local traffic contribution depends upon the traffic volume, street characteristics (if it facilitates or not the pollutants dispersion), as well as the distance between the site and the street.

The contribution of local traffic at urban background stations ranges between 0 and 27%, and the average contribution for the 11 sites is 11%. The city of Budapest gives a contribution above 20% of local traffic for 2 UB and 1 SB sites, and the conurbation of the cities of Gdańsk, Gdynia and Sopot (Poland) also has a contribution of local traffic of 20%. If those four sites are removed, the average contribution of local traffic at urban background sites is 4%, and the range is 0-10%. The lowest contribution of local traffic to PM10 concentrations corresponds to the city of Yorkshire (UK) and the largest to the municipality of Brno-Tuřany (Czech Republic). Leaving out the suspicious sites, the ratio between local traffic contribution at urban traffic stations and at urban background stations (including also suburban background stations) is 5.3.

The average contribution of local traffic at industrial sites is 1%. The contribution varies between null contribution at the two sites in Duisburg (Germany) and 4% at the site in Wien (Austria). For the mixed sites, the average contribution of local traffic to PM10 concentrations is 20%, with the range 14-27%.

Combined urban and local traffic contribution

Combining the urban and local traffic contribution to urban traffic sites for the 29 sites that had reported non suspicious data (with >3% of urban traffic), the averaged total contribution of traffic to PM₁₀ concentrations measured at urban traffic sites is 34% (13% urban and 21% local), in the range from 13% (Duisburg) and 61% (Glasgow). The contribution of traffic at urban background sites is 15% (10% urban and 5% local) averaged over 5 sites (with <20% of local traffic and >3% of urban traffic), and it ranges between 6% (Yorkshire) and 22% (Brno). This contribution from traffic to PM10

concentrations is in agreement with the one reported by Hak et al. (2010). The average contribution from urban and local traffic to urban traffic sites was found to be about 36%. In that report 21 cities were analyzed, but unlike in this report not all of the cities had more than 250000 inhabitants. 16 cities included in Hak et al. (2010) have not been included in this report.

4.2.2 *NO₂ assessments in TENS*

Sixteen countries reported exceedances of the hourly or annual NO₂ limit value at one or more zones. **Table 11** shows the number of zones reported by each country and the number of cities with more than 250000 inhabitants included in the TENS for NO₂ limit value. This information shows that most of the European countries recorded exceedances of the hourly or annual NO₂ limit value in one or more zones. According to article 22 in Directive 2008/50/EC where in a given zone or agglomeration conformity with the limit values for NO₂ cannot be achieved by the deadline of 1 January 2010, a MS may notify the EC if, according to their opinion, conditions are met in a given zone or agglomeration for being exempt from the limit values for NO₂. A quantitative source apportionment on the origin of pollution values contributing to the exceedance is essential. The source apportionment must reflect regional, urban and local contributions. The urban and local contributions are further subdivided, so that, for instance, road traffic contributions can be identified.

Table 11 lists the countries and the number of zones that have been considered to assess the contribution of traffic to NO₂ levels. The table includes those zones with sufficiently detailed source apportionments for a number of cities larger than 250000 inhabitants. There are countries, however, where the forms have not been filled in with sufficient thoroughness or just very rough source apportionments were supplied, in the table they appear as they have zero zones for source apportionment. Some source apportionments, as in the case of PM, could not be used for this assessment due to incompatibility of the information.

Table 11 Number of zones with sufficient information to assess traffic contribution sorted by country. Information obtained from NO₂ TENs.

| Country | No. zones reported | Thereof cities > 250000 inh. | Number of source apportionments |
|---------------------|--------------------|------------------------------|---------------------------------|
| Austria (AT) | 12 | 2 | 4 |
| Belgium (BE) | 2 | 1 | 1 |
| Bulgaria (BG) | 2 | 1 | 1 |
| Czech Republic (CZ) | 4 | 3 | 3 |
| Germany (DE) | 108 | 27 | 53 |
| Denmark (DK) | 3 | 2 | 2 |
| Spain (ES) | 6 | 1 | 1 |
| Finland (FI) | 1 | 1 | 1 |
| France (FR) | 3 | 0 | 0 |
| Hungary (HU) | 2 | 1 | 1 |
| Italy (IT) | 54 | 8 | 12 |
| Luxembourg (LU) | 1 | 1 | 1 |
| Latvia (LV) | 1 | 1 | 1 |
| Poland(PL) | 1 | 1 | 1 |
| Portugal (PT) | 5 | 1 | 1 |
| United Kingdom (UK) | 1 | 0 | 0 |

The TENs for NO₂ have the same form as for PM₁₀, providing information about the type of stations (urban, rural, industrial, etc.) and the contributions from the individual sources to concentrations. As for PM₁₀, the response to the questions on source apportionment was not expressed in a standardized way in the TENs from the countries. Percentage contributions of all sources sum up to a total of 100%. Apportionments which consisted of concentration contributions have been converted to percentage contributions.

A total of 83 source apportionments were evaluated, 76 of them correspond to stations classified as traffic stations, and one to a station not classified but that according to its source distribution can be classified as UT. However, three sites have not been considered for the analysis of source contribution because the data reported was considered suspicious. The city of Florence has not been taken into account in the statistics because the contribution of 0% of local sources at an urban traffic station reported was considered suspicious. The city of Bielefeld has not reported urban traffic contribution, and the city of Hannover has reported a very low (0.2%) contribution from urban traffic. Also 6 sources apportionments reported for a set of different types of stations (traffic, industrial and background) were analyzed.

Table 12 : Urban and local traffic contribution to NO2 concentrations at Urban Traffic sites. Information obtained from source apportionments in TENs. The information was normalized so the sources sum up to a total of 100%. r: regional; u: urban; l: local

| Description of the zone | | Description of the exceedance | | | Source apportionment | | | | |
|-------------------------|-------------------------------|-------------------------------|----|---------|------------------------|-------|-------|--------------------------|----|
| | | | | | Total contribution (%) | | | Traffic contribution (%) | |
| | Cities or Municipalities | Code | LV | Station | r | u | l | u | l |
| AT | Wien | AT09_NO2_1, | a | UT | 7 | 23 | 70 | 15 | 70 |
| AT | Wien | AT09_NO2_2, | a | UT | 14 | 43 | 43 | 28 | 43 |
| AT | Wien | AT09_NO2_3 | a | UT | 13 | 42 | 45 | 27 | 45 |
| AT | AG Graz | AT60_NO2_01 | | UT | 3 | 33 | 65 | 26 | 61 |
| CZ | Brno | CZ0642-1 | a | 2 UT | 21 | 10 | 69 | 8 | 69 |
| CZ | Ostrava | CZ080-1 | a | 1 UT | 22 | 33 | 55 | 11 | 55 |
| DE | Brandenburg a. d. H. (region) | DEBB_E_04 | a | UT | 18 | 10 | 71 | 7 | 71 |
| DE | Berlin | DEBE_E_02 | a | UT | 12 | 42 | 46 | 30 | 46 |
| DE | Mannheim | DEBW_E_20 | a | UT | 18 | 44 | 38 | 25 | 28 |
| DE | Karlsruhe | DEBW_E_22 | a | UT | 20 | 49 | 31 | 34 | 23 |
| DE | Stuttgart | DEBW_E_10 | a | UT | 44 | 27-65 | 14-64 | 17-39 | 18 |
| DE | Stuttgart | DEBW_E_10 | a | UT | 8 | 28 | 64 | 17 | 59 |
| DE | München | DEBY_E_01 | a | UT | 18 | 13 | 69 | 5 | 69 |
| DE | Augsburg | DEBY_E_03 | a | UT | 29 | 22 | 49 | 5 | 49 |
| DE | Nürnberg | DEBY_E_04 | a | UT | 26 | 26 | 47 | 11 | 47 |
| DE | Bremen | DEHB_E_01 | a | UT | 17 | 27 | 57 | 16 | 49 |
| DE | Frankfurt | DEHE_E_06 | a | UT | 23 | 46 | 31 | 31 | 31 |
| DE | Frankfurt | DEHE_E_17 | a | U (UT) | 30 | 51 | 19 | 33 | 19 |
| DE | Wiesbaden | DEHE_E_07 | a | UT | 21 | 39 | 42 | 41 | 42 |
| DE | Hamburg | DEHH_E_010 | a | UT | 8 | 14 | 79 | 5 | 79 |
| DE | Hannover | DENI_E_41 | a | UT | 13 | 16 | 71 | 11 | 43 |
| DE | Wuppertal | DENW_E_204 | a | UT | 25 | 16 | 59 | 7 | 59 |
| DE | Münster | DENW_E_192 | a | UT | 26 | 20 | 53 | 9 | 53 |
| DE | Bonn | DENW_E_162 | a | UT | 35 | 40 | 25 | 9 | 25 |
| DE | Bonn | DENW_E_162 | a | UT | 21 | 15 | 65 | 5 | 65 |
| DE | Köln | DENW_E_184 | a | UT | 17 | 13 | 70 | 5 | 70 |
| DE | Köln | DENW_E_184 | a | UT | 21 | 17 | 61 | 6 | 61 |
| DE | Köln | DENW_E_184 | a | UT | 21 | 21 | 58 | 6 | 58 |
| DE | Köln | DENW_E_184 | a | UT | 28 | 24 | 48 | 8 | 48 |
| DE | Köln | DENW_E_184 | a | UT | 27 | 25 | 48 | 7 | 48 |
| DE | Köln | DENW_E_184 | a | SUT | 51 | 35 | 14 | 14 | 53 |
| DE | Köln | DENW_E_184 | a | UT | 37 | 25 | 38 | 10 | 38 |
| DE | Köln | DENW_E_184 | a | UT | 20 | 16 | 64 | 5 | 64 |

| | | | | | | | | | |
|----|---|---------------|-----|------------|----|----|----|----|----|
| DE | Köln | DENW_E_184 | a | UT | 21 | 15 | 64 | 6 | 64 |
| DE | Köln | DENW_E_184 | a | UT | 35 | 25 | 40 | 10 | 40 |
| DE | Gelsenkirchen | DENW_E_175 | a | UT | 30 | 26 | 44 | 10 | 44 |
| DE | Essen | DENW_E_174 | a | UT | 24 | 23 | 53 | 7 | 53 |
| DE | Essen | DENW_E_174 | a | UT | 35 | 30 | 35 | 8 | 35 |
| DE | Essen | DENW_E_174 | a | UT | 28 | 35 | 37 | 17 | 37 |
| DE | Essen | DENW_E_174 | a | UT | 44 | 30 | 25 | 8 | 25 |
| DE | Essen | DENW_E_174 | a | UT | 27 | 59 | 15 | 46 | 15 |
| DE | Essen | DENW_E_174 | a | UT | 30 | 21 | 49 | 10 | 49 |
| DE | Essen | DENW_E_174 | a | UT | 24 | 23 | 53 | 7 | 53 |
| DE | Essen | DENW_E_174 | a | UT | 42 | 29 | 29 | 15 | 29 |
| DE | Dortmund | DENW_E_166 | a | UT | 25 | 47 | 28 | 42 | 28 |
| DE | Dortmund | DENW_E_166 | a | UT | 29 | 53 | 18 | 46 | 20 |
| DE | Dortmund | DENW_E_166 | a | UT | 21 | 14 | 65 | 8 | 65 |
| DE | Dortmund | DENW_E_166 | a | UT | 36 | 24 | 40 | 14 | 40 |
| DE | Düsseldorf | DENW_E_170 | a | UT | 17 | 17 | 66 | 9 | 66 |
| DE | Düsseldorf | DENW_E_170 | a | UT | 18 | 18 | 63 | 10 | 63 |
| DE | Aachen | DENW_E_158 | a | UT | 24 | 24 | 53 | 11 | 53 |
| DE | Aachen | DENW_E_158 | a | UT | 20 | 19 | 61 | 9 | 61 |
| DE | Mönchengladbach | DENW_E_190 | a | UT | 30 | 20 | 50 | 20 | 50 |
| DE | Mönchengladbach | DENW_E_190 | a | UT | 24 | 16 | 60 | 16 | 60 |
| DE | Duisburg | DENW_E_167 | a | UT | 39 | 36 | 25 | 12 | 25 |
| DE | Duisburg | DENW_E_167 | a | UT | 43 | 30 | 27 | 7 | 27 |
| DE | Leipzig | DESN_E_02 | | UT | 24 | 19 | 57 | 14 | 57 |
| DE | Dresden | DESN_E_06 | a | UT | 27 | 22 | 51 | 18 | 51 |
| DK | Copenhagen/Frederiksberg | DK0001 | a | UT | 20 | 20 | 60 | 16 | 60 |
| DK | Aarhus | DK0002 | a | UT | 25 | 25 | 50 | 20 | 50 |
| ES | Palma | ES0401_NO2_a | a | UT | 10 | 15 | 75 | 6 | 75 |
| FI | Helsinki | FI_HEL_1 | a | UT | 11 | 35 | 55 | 29 | 55 |
| HU | Budapest | 001/1 | a | UT | 29 | 39 | 32 | 21 | 25 |
| IT | Napoli | I15E01 | a | UT | 14 | 64 | 22 | 36 | 13 |
| IT | Napoli | I15E02 | a | UT | 10 | 47 | 43 | 27 | 25 |
| IT | Bari | I16E01 | a | UT | 12 | 38 | 50 | 28 | 28 |
| IT | Palermo | I19E01 | a | UT | 10 | 36 | 54 | 26 | 54 |
| IT | Catania | I19E02 | a | UT,SUT | 6 | 15 | 80 | 11 | 80 |
| IT | Firenze (Florence) | I09E02 | a | UT | 13 | 43 | 43 | 27 | 43 |
| IT | Firenze (Florence) | I09E03 | a | UT | 10 | 32 | 58 | 21 | 58 |
| LU | Luxembourg | LXBW_E_01 | a | UT | 14 | 13 | 73 | 3 | 73 |
| LV | Rīga | NO2_Rīga | a | UT | 8 | 82 | 10 | 56 | 10 |
| PL | strefa aglomeracja krakowska: Kraków | Mp07MKRNO2a01 | a | NA (UT) | 6 | 24 | 70 | 12 | 55 |
| PT | Lisboa | PT3001_NO2_T | h+a | UT | 14 | 41 | 45 | 29 | 41 |

Table 13 Urban and local traffic contribution to NO₂ concentrations at mixed sites. Information obtained from source apportionments in TENs. The information was normalized so the sources sum up to a total of 100%. r: regional; u: urban; l: local

| Description of the zone | | Description of the exceedance | | | Source apportionment | | | | |
|-------------------------|--------------------------|-------------------------------|-----|-------------------------|------------------------|-------|-------|--------------------------|-------|
| | | | | | Total contribution (%) | | | Traffic contribution (%) | |
| Member State | Cities or Municipalities | Code | L V | Station | r | u | l | u | l |
| BE | Brussels | BEB10a (NO ₂) | a | UI, 3UT, 1U | 41 | 19 | 26 | 10 | 15 |
| CZ | Praha | CZ010-1 | a | 3 UT 1 UB | 22 | 42 | 36 | 26 | 36 |
| IT | ROMA | I12E1 | a | IT, UB, UT | 11 | 52 | 37 | 40 | 37 |
| IT | Genova | I07E01 | a | IT, UT | 13-22 | 24-41 | 63-37 | 10-17 | 35-37 |
| IT | IT0301 Milano area | I03E01 | a | UT, UI, SI | 30 | 49 | 21 | 26 | 21 |
| BG | Пловдив (Plovdiv) | BG0002_PD_NO ₂ _a | a | Transport oriented (UB) | 55 | 45 | na | 35 | na |

Regional, urban and local contributions (all sources) to total NO₂

In summary, at urban traffic stations the regional contribution to NO₂ concentrations are between 3 and 51%, with an average of 22%; the total urban contribution ranges between 5 and 82% with an average value of 30%; and the total local contribution is from 10 to 80% with an average of 50%. This assessment is based on a total of 74 sources apportionments for sites classified as urban traffic stations plus one suburban traffic station (Köln), and one site (Kraków) not classified yet, according to its distribution of sources probably correspond to an urban traffic site. For those source apportionments that include several types of stations, the total regional contribution to NO₂ concentrations ranges from 11 to 55%, and has an average value of 28%; the total urban contributions is about 39% (range between 19 and 52%); and the total local contribution ranges between 21 and 63% with an average value of 37%.

Contribution of urban traffic

Road traffic within the urban area was assessed to contribute with 17% to NO₂ concentrations measured at urban traffic sites. The urban traffic contribution ranges from 0.2 to 56%. The lowest contribution corresponds to an UT site in the city of Hannover (Germany), and the largest to Riga (Latvia). Following the same rationale as

for PM_{10} , we consider suspicious data from those sites with contributions below 3% of the cities' own traffic to urban traffic sites. If those sites are left out (2 sites in two cities), the average for the remaining 74 sites is 17% with a range from 3 to 56%. The lowest contribution corresponds now to Luxembourg.

The urban traffic contribution to mixed stations is 23% as an average of 6 places, and ranges between 10 and 40%. The lowest contribution corresponds to Brussels (Belgium) and the largest to Rome (Italy).

Contribution of local traffic

The contribution of local traffic at urban traffic sites accounts for 47% of the NO_2 concentrations measured at urban traffic sites, and it ranges between 10 and 80%, as an average of 76 sites in 44 cities. The lowest contribution corresponds to Riga (Latvia) and the largest to Catania (Italy). For the statistics the null contribution of the UT site at Firenze (Italy) has not been taken into account since considering that a contribution of 0% of local sources to an UT site is not realistic.

As in the case of PM_{10} , the large range of traffic contribution to NO_2 concentrations may be due to the differences in the location of the station (close or nor to the road) and the ventilation in the street.

The local traffic contribution at mixed sites ranges between 15 and 37%, with an average value of 30%. The minimum corresponds to the city of Brussels (Belgium) and the maximum to Rome (Italy). These two cities have also the lowest and largest contributions of urban traffic to NO_2 concentrations, respectively.

Combining the urban and local traffic contribution to urban traffic sites for the 74 places that have reported non suspicious data (with >3% of urban traffic), the averaged total contribution of road traffic contributions measured at urban traffic sites is 64% (17% from urban and 47% from local).

5 Mitigation strategies

Mitigation strategies can be applied at several levels from the European level like the EURO standards down to city levels like the congestion charge in London or the studded tyre ban in one street (Hornsgatan) of Stockholm. Measures may have short term goals like the proposed diesel vehicle ban in Oslo on severely polluted days, or long term goals like incentives for cleaner fuels or change of vehicles. The measures can be focused, like a taxation of specific fuels, vehicles and areas, or more widely defined, like promoting public transport and informing the public about the effects of air pollution. Several measures are already applied in European cities and also many are planned. For instance in the TEN Berlin indicates 37 measures and Vienna 76, mainly focused on a local scale.

A problem when discussing mitigation strategies is that the effect of most measures is difficult to assess as there are many factors influencing the reduction in emissions and changes in concentrations, as for instance variations in meteorology. The most widely proposed traffic measures already implemented, reported in Hak et al. (2010), were promoting public transport and cycling, speed moderations and cleaner fuels/vehicles, and taking action on public fleet. The effectiveness of these measures was not assessed, but an interesting observation was that the cities reporting lower than average NO_x emissions had implemented more mitigation measures than the cities with higher NO_x emissions than average levels.

5.1 Reported effects of measures in literature.

5.1.1 *Low emissions zones and congestion taxation*

Low emissions zones (LEZ) (www.lowemissionszones.eu) have been applied in many cities either by pricing or by a ban of certain vehicles, following either the EURO classification or directed towards heavy duty vehicles. LEZ have been implemented in London, Lisbon, Prague and several cities in Sweden, Denmark, Netherlands, Italy and Germany. Most LEZ in Europe were introduced after 2007, even if the first LEZ was introduced in Sweden in 1996. In London has a central zone with congestion charge been in place from 2003 for all vehicles and a larger area for the LEZ from 2008 directed towards heavy duty vehicles. The effect reported for London after the introduction of congestion charge in 2003 was positive with emission reductions for NO_x, PM₁₀ and CO₂. (Beevers and Carslaw, 2005). The reduction was attributed to less congestion and vehicle km. In 2008 it was reported that the levels of congestion had increased steadily since the introduction of the congestion charging (Transport of London, 2008). Likely due to lower capacity on the road system. However, the annual average of PM₁₀ has been reduced and has been below the limit value since 2004. The number of days above the daily limit value was also visibly reduced, but is still above 35 days (Pires and Martins, 2012). After the introduction of the larger low emissions zone in 2008 and a demand for “sulphur free” fuel in 2007 the particle number concentrations dropped in London (Jones et al, 2012). The observed levels were mainly considered due to the low sulphur fuel rather than the LEZ.

An evaluation of the congestion tax in Stockholm concluded that traffic emissions were reduced, leading to a reduction in ambient concentrations of 12% for NO_x and 7 % for PM₁₀. The limit values would still be exceeded along main roads but 27 premature deaths would be avoided each year, highlighting the positive effect of the reduction (Johansson et al, 2009b).

As a general comment; LEZ (or congestion charging) according to EURO classes might however not be a good measure for NO₂ due to the increased NO₂ fractions in newer diesel cars, see section 2. It has also been reported that LEZs in Germany are unpopular and have met some resistance from the public (Wolff and Perry, 2010). High cost for businesses and a decline in revenues are some of the critics, also large percentage of the population believe that the LEZ have little effect (Wolff and Perry, 2010). Experience from London shows however that business did not get negatively affected by the congestion charging and the LEZ. It also had positive effect for traffic safety with less accidents and increased safety for both pedestrians and pedal cycles (Transport of London, 2008).

5.1.2 Speed moderations

Speed reduction is also applied in several cities and reported as one of the most commonly implemented measures. Listed benefits of reduced speed are; decrease in total fuel consumption, lower emissions of CO₂ and other components, in addition to having a positive influence on congestion, noise and traffic accidents. Oslo applied an environmental speed limit in 2005 (from 80 km/h to 60 km/h) with the goal of reducing the road dust, indicating up to 35 % reduction on the PM₁₀. Even if it was clear that the reduced speed limit had effect, a quantification of the effect of the speed limit reduction compared to the other factors like meteorology and changes in traffic was not possible at the time (Hagen et al, 2005). Also a reduction in NO₂ levels was observed.

Reduced speed limit down to 80km/h was enforced in the Netherlands along urban highways in 2005. The effect of the emission reduction was estimated for Amsterdam and Rotterdam and found to be in the range of 5-30 % for NO_x and for 5-25% PM₁₀. The effect depended much on the change in congestion before and after the speed reduction and the amount of heavy duty vehicles (Keuken et al, 2012). Reduction in the concentration levels was found to be significant at the Amsterdam ring road reducing traffic's contribution to PM₁₀, PM₁ and Black smoke (Dijkema et al., 2008), both due to lower exhaust emissions and reduced non-exhaust emissions. NO_x levels on the contrary did not show any clear reduction. Keller et al., (2008) modelled a reduction of NO_x emissions when testing the effect of reduced speed limits down to 80 km/h applied from 12-17 August in 2003 on Suisse motorways.

The area around Barcelona had reduced highway speed limits down to 80 km/h, from 2007 but the reduction was removed in 2011 going back to higher speeds. The effect of the speed reduction was modelled and an estimated effect of up to 5.7 % 5.3 and 3 % was found for NO₂, SO₂ and PM₁₀ respectively. The model underestimated the PM₁₀ concentration levels which can partly be related to lack of non-exhaust and other

emission sources in the modelling (Goncalves et al., 2008). Reduction of up to 11% in the vicinity of the road ways was also found by Baldasano et al., (2010) for the Barcelona area, improving the air for 1.35 million inhabitants (41 % of the population).

Panis et al., (2011) use two different model approaches to study the effect of speed changes and they found that one approach showed an increase of PM₁₀ whereas the other gave a decrease for the chosen scenarios. The modelled effect of the speed change will be based on how the speed and the emissions are coupled in the model and important factors like congestion and non-exhaust and re-suspension are often not included. This need to be carefully considered when analysing the results of a model calculation of a measure and illustrate the scientific uncertainties the policy makers need to consider.

The actual effect of a speed reduction will likely be different for different compounds. Also at a specific site the change in traffic situation will vary according to how it was before the measure was put in place. The speed limit does not directly reflect the driving behaviour and speeding with consecutive slowing down passing radars or other controlling measure could possibly increase emissions rather than decrease them. If a reduction from 50km/h -30km/h is done where the average speed limit anyway is below 30 km/h the change might not make any noticeable difference. If the change in speed limit would increase flow it still could have a positive effect. For PM an important contribution is the non-exhaust and a reduction in speed is likely to reduce this contribution even if a quantification of this effect is missing (Baldasano et al., 2010, Oxley et al, 2012).

5.1.3 Other measures

Oxley et al. (2012), used the BRUTAL model to evaluate different measures towards the road transport defining several scenarios including; 1) switch from petrol to diesel, 2) switch to smaller vehicles, 3) reduced usage, 4) low carbon technologies (electric and hydrogen), and 5) increased use of bio ethanol. Also some combined scenarios were studied. The main focus in the article are effects on CO₂, but also NO_x and PM₁₀ (tailpipe emissions only) are modelled. The scenarios with reduction in use of passenger cars combined with reduction in size of the remaining cars had the best effect; also the use of electric cars was highlighted as a good option as long as the extra energy to charge the batteries came from renewable energy sources. The authors stressed that a holistic approach is needed when evaluating abatement strategies.

5.1.4 Non-exhaust mitigation

Remediation measures have been tested in Europe with the specific goal to mitigate road dust emissions. Often the effectiveness has been calculated for a single road and therefore their effectiveness on a city or meso-scale still need to be demonstrated as well. These measures can be classified into five groups:

1. Road sweeping and washing. Road cleaning activities are generally performed by municipalities for sanitary and esthetical purposes. However, an improved (in frequency and/or intensity) road cleaning protocol has been proved to have a beneficial effect on road dust load; the result for PM₁₀ concentrations is however more unclear and street

sweeping alone did not show any effect on PM₁₀ levels (Amato et al., 2010a). Some city-studies report reduction in the PM concentrations by street flushing, some do not (Keuken et al., 2010, Ang et al., 2008 and John et al., 2006). In addition, the duration of effectiveness varies but being limited to the first 8-24 hours after treatment. The difference in the results could be due to local conditions and the method applied. The indication of possibly positive effect of flushing could be due to temporarily damping rather than removing the dust and which possibly explain the observed short term effects. (Amato et al., 2009a; 2010b; Karanasiou et al., 2011) Light and frequent washings are maybe more effective than episodic and intense ones (Amato et al., 2012), also due to the large variability in road dust loading the large scale road cleaning campaigns are questionable (Mathissen et al., 2012). However Zhu et al. (2012) found that sweeping shortly after sanding events on the whole road system reduced PM concentrations and was better than seasonal sweeping and only sweeping on main roads. This study was particular in the sense that it was timed according to sanding events. This could indicate that the main effect of sweeping will be to remove larger particles to prevent crushing and a increased sand paper effect rather than removing the PM fraction.

2. Chemical suppressants. Several chemicals can be used to suppress dust, the most common being salts. The chemicals reduce the vapour pressure keeping the road moist which damp emissions and can also demonstrate some binding effects on particles making a sticky paste. Many products have been actively used on unpaved roads and sand outtakes etc, but the use in a city environment on a regular basis is fairly uncommon. Most products can be dispensed as liquids by a truck equipped with a tank, spray bar and a flow-controller. Some suppressants are likely to reduce the friction between road and tires, and effective cleaning needs to be adapted and dosage needs to be controlled to avoid effects on traffic safety. The environmental impact of the chemicals has been a concern, also effects on road constructions, vehicles and the surrounding build environment need to be considered. In the Netherlands, a positive impact of CaCl₂ was found, reducing the share of the road dust in the background concentration by over 10%. Calcium Magnesium Acetate (CMA) has been tested in Stockholm, Stuttgart, London and Austria indicating in most cases some effectiveness against road dust emissions (Norman and Johansson, 2006; see also <http://www.tfl.gov.uk/corporate/projectsandschemes/17246.aspx>, and <http://www.life-cma.at/english/project-contents-89.asp>). In Oslo MgCl₂ has been used on a regular basis since 2006 to damp the suspension from major roads, with apparent success as levels of PM₁₀ have been below the limit values since 2007 (Oppegaard and Løseth, 2012)
3. Change in road pavement. Road pavement characteristics influence the rate of wear and the potential of emission. Swedish researchers (Gustafsson et al., 2009) have performed facility tests to describe the behaviour of different road pavements with regard to the wear rate. Granite pavement was found to be more prone (70% higher) to PM₁₀ production compared to the quartzite pavements. Also the total wear of a pavement normally decreases with increasing aggregate size. These results indicate that by making proper choices regarding pavement material, can somewhat reduced wear. Unfortunately is small aggregate size favourable for noise reduction. Finnish tests described different materials' resistance to wear and stating that mafic, volcanic rock is

the most resistant compared to granite(Raisanen et al, 2005). Conditions of pavement might also be important: fresh abrasion particle emissions from pavements in good condition are reported to be quite low.

4. Reduced use of studded tyres, salting and sanding. Even if it is only a few countries in Europe where studded tyres are used, it is a major contributor to road dust in several cities. In Stockholm around 60 % of the vehicles use studded tyres. Studded tyres are only allowed during the winter period. Some cities in Norway practice taxation on the use of studded tyres reducing the share, like in Oslo the share is now below 20 %. Salting and sanding are also common traction control methods contributing with extra dust load in the cities. Even if contribution from salting and sanding can be removed from the concentration for compliance with the limit values, it is often not done as it is difficult to quantify the contribution. It is also likely that the effect on health is anyway present. Salting has clear negative effect on the biological environment.
5. Other non-technological measures. Strategies aimed to reduce traffic volume and vehicle speed have been applied in several European cities as discussed above of LEZ, congestion charge etc. Even if these measures often are not only applied for the non – exhaust emissions it can be a positive added effect. Reducing traffic volumes reduces the total wear, the direct emission and the re-suspension. Vehicle speed is also directly related to the road wear and emission strength.

6 Comparisons and discussions

The road traffic contribution to ambient air concentrations of air pollutants vary largely in space and time (dependent on the emissions, distance to the sources and meteorology). This makes it difficult to draw general conclusions applicable to the ensemble of European cities. The data analysed are often based on measurements from a limited number of stations which represent a small area, and do not necessarily represent road traffic's contribution to the whole city or other cities in the same region or country. This can easily be demonstrated from analysis within one city at two different stations showing differences in traffic contributions, or in two cities in the same country (see tables). The representativeness of the stations is also highly relevant when doing source apportionment. Especially in the TENs several cities reported numbers that are clearly suspicious. When zero or very low local traffic contribution to a traffic station is reported, something is wrong, indicating that either the methods used are not good, the station classification is wrong or that the people reporting the data misunderstand the terminology.

Often, source apportionments are performed from campaign data. Some studies have data covering one year; some only for shorter periods (for instance 2 weeks in summer/winter). Differences in data coverage also make it difficult to compare one study with another, and results from short periods cannot be directly analysed towards the annual limit values. Methods for finding the traffic contributions are also quite different with some methods only giving the local contribution. Chemical speciation needs further to be understood as well as accounting for the non-exhaust contribution. Road traffic contribution to secondary particles and long range transport are often not addressed.

In spite of data and method limitations, it is clear both from the literature review and from the TENs that traffic is a large contributor to NO_2 and PM levels in cities.

6.1 NO_2

Emissions from traffic vary according to several parameters like car fleet, annual daily traffic (ADT), speed and driving pattern. In European cities the exhaust emissions of several pollutants have been reduced generally except for NO_2 which has increased due to the dieselisation of the car fleet and a larger NO_2/NO_x ratio in newer diesel cars. NO_x emissions vary largely for the light and heavy vehicles tested in different studies, but it is clear that in general they show larger emissions than stipulated by the EURO standard. The increase in NO_2 direct emissions is also not evenly distributed in Europe as the increased mileage and dieselisation of the car fleet varies quite distinctly from country to country/city to city.

Increase in NO_2 emissions is not only relevant for the NO_2 concentrations but also since NO_2 is a precursor of ozone - the levels of which have seen an increase in urban areas. Furthermore, nitrogen oxides play a major role in secondary particle formation (see sections on ammonia and secondary inorganic aerosols).

NO₂ concentrations exceedances of the LV occur widely over Europe. Also urban background concentration levels are often close to the annual LV which then easily leads to exceedances at the road side. From the articles reviewed traffic is considered the only or at least dominant source at traffic stations in urban areas even if quantifications are not given. In Anttila et al. (2011) a quantification was done at a site in Helsinki heavily influenced by traffic. Contributions were found as follows: 18 % from the regional background, 31 % from the direct traffic NO₂ emissions and 51% from photochemical transformation of NO as contribution to measured NO₂ levels corresponding to the years 1994-2004. However the contribution from direct emissions increased towards 2009 which was then similar to the photochemical contribution of 43% due to an increase in the diesel share in the vehicle fleet.

In the TENs the average reported contribution from urban and local traffic was 64%. The range for the local contributions was 10-80% and for the urban traffic contribution the range was 3-56%. The regional contribution might of course be considered to also have road traffic as origin but this was not further investigated.

The differences in percentage and fairly large span of NO₂ contribution reported in the TENs might be due to some errors related to methods, misunderstandings and station classification as mentioned, with some countries reporting suspicious data. It could also be due to the actual level of the NO₂ concentrations hence to the possible difference in relative importance of the different processes, e.g. NO oxidation, direct emissions of NO₂ and dispersion. Factors influencing the levels and contributions, as discussed by many authors in the review literature, are for instance amount of sun light, depending on latitude and season, local meteorology and ozone background concentrations. The amount of emissions is of course also crucial which is affected by the car fleet, mileage, etc. There is a large difference in the fraction of diesel passenger cars, going from almost none in Athens to above 70 % in Paris.

The photochemical processes influence largely the NO₂ concentration levels, however the importance of the extra contribution from the primary NO₂ emissions is clear at several sites, especially traffic stations. The discussion of the primary NO₂ contribution is mainly done with the yearly average in mind and more insight in the high pollution episodes would be interesting. The influence of high concentration hours on the yearly average is also not largely explored in the literature reviewed.

6.2 PM

The TENs give an average estimate of 31 % of urban and local traffic contribution to the measured PM₁₀ concentrations at traffic sites, and 19% to urban background sites. This is lower than what is found for NO₂, as expected. The local traffic contributions to traffic stations reported in the TENs have a range of 6-34%. The overall regional total contribution (from all sources) is ranging from 15-75% with an average of 51%.

In the reviewed articles the range of the traffic contribution to urban PM concentrations is 13-70% for PM₁₀ and 19-66% for PM_{2.5} (Table 3 and Table 4 and references therein).

The number of studies is too low and limited in both spatial coverage and sometimes based on short periods' campaign data to derive a useful statistical analysis for urban areas across Europe. Differences seem to be more specific to measurement location and related parameters (like station type, traffic numbers and size of agglomeration). Seasonal variations are also not necessarily the same, even within one country. For instance it was found for Italy that concentrations in Milan had higher concentration levels in winter while it was the opposite for Lecce, due to differences in meteorology and source profiles.

The road traffic contributions reported in the articles seem to be larger than what is reported in the TENs, the latter having relatively high regional contributions from all sources. It is unknown for what reason this difference appears but it is likely to be partly connected to the difference in methods. Several TENs report very low traffic contributions at traffic stations. The main uncertainty related to quantifying road traffic contribution to PM is to quantify the non-exhaust contribution from traffic and a quantification of the contribution to the formation of secondary particles (SOA and SIA).

No direct quantification of SOA and SIA was possible in this review, but it is evident that secondary particles contribute with rather large fraction to the PM levels. Focus in better understanding formation of both SOA and SIA in the urban environment is hence crucial for setting up targeted measures for reducing PM concentrations in most of Europe. A few studies looking into the processes of SIA formation recommended further reduction of NO_x emissions as the best way to reduce the SIA contribution to PM. The contribution to secondary particles should be kept in mind also when introducing new technologies or fuels like bio-diesel and bio-ethanol, possibly emitting other non regulated compounds relevant for the urban air quality.

For PM the emission from non-exhaust processes has increased its relative contribution due to increased mileage and reduced exhaust emissions per vehicle. A few studies estimating specifically the non-exhaust contribution found larger contributions from the non-exhaust compared to the exhaust. An extra focus on quantifying the non-exhaust emissions is also important as it is often not accurately modelled or not fully included in modelling concentration levels. This is a large drawback when doing scenario runs either for future projections or when estimating effects of a mitigation measure.

Black Carbon (BC) levels vary proportionally to those of traffic-related gaseous pollutants (CO, NO and NO₂) and increased levels are found close to traffic at kerbside stations. Road traffic contribution to BC is however not directly quantified but is likely to depend on the vehicle fleet. EC emission calculations (see section 2) show a reduction over the last decade for the average over European cities.

Even if the data amount from the latest years is fairly scarce compared to the number of cities in Europe reporting exceedances of LV and too scarce to do a further analysis, one can conclude that road traffic is an important source and that its contribution ought to be larger when including road dust and secondary particles (organic and inorganic).

6.3 Mitigation

The evaluation of the effect of measures is difficult and trends in pollution levels can only be proven after several years, due to e.g. variations in meteorology from year to year. The results from models estimating the effect of a mitigation measure are highly dependent on the emissions applied, which often tend to exclude non-exhaust and parameters like flow/congestion or driving pattern. Due to relatively few published studies, strong recommendations only based on this review are difficult to outline. See also discussion in section 5. However action needs to be taken fast as LVs are exceeded at present. Experience and knowledge of what works need to be shared among stakeholders, municipalities and countries. More interactions among decision makers are highly recommended.

Due to the increased fraction of NO₂ in diesel passenger cars and enhanced NO_x emissions in urban driving, continued increase of diesel passenger cars in the car fleet in urban areas would anyway not be beneficent for this compound. Diesel engines are also the main contributors to primary BC emissions (Colvile et al., 2001), whereas petrol emissions dominate over diesel in formation of secondary organic aerosol mass (SOA). The contribution of road traffic to the SIA is not addressed explicitly in this report but the recommended action is to reduce NO₂ for reducing SIA. This would lead to a recommendation to reduce the amount of diesel cars as modern petrol cars have negligible NO₂ emissions. It should always be a goal to not affect other compounds negatively when introducing a measure or new technology. For the upcoming EURO 6 the use of urea in diesel cars after treatment might reduce the NO₂ emissions but might increase emissions of NH₃. The first tests of these systems for passenger cars show however NH₃ levels as for modern petrol cars (Heeb et al., 2011).

The reduction of direct NO₂ emissions from road traffic will be important to possibly reduce NO₂ exceedances in most places as a general reduction of NO_x has shown not to influence the local NO₂ levels to a large extent the latest years. Reducing NO₂ from diesel cars under all kinds of driving situations should be demanded and work with changing the type approval routines will help. Initiating a shift towards electric or hybrid cars and also smaller petrol cars should also be a goal in cities. Taxation regimes have proved to be efficient for the change from petrol to diesel vehicle so similar effect would be possible toward lower polluting vehicles if taxation rules are formed in a constructive way. Such vehicle change could be important for emissions of air pollutants and also for the CO₂ emissions.

It can take years before the vehicle fleet will be renewed and an effect of reduced fleet emissions will be noticeable. This means that other traffic measures like reducing the ADT or changes to flow and driving behaviour by e.g. speed moderation seem to be inevitable for most cities. The contribution from non-exhaust emissions will be present independently of exhaust contribution, making traffic reduction a recommendation for reducing emissions from this source. Studies looking at street washing or sweeping so far did not show a large effect and dust suppressant might be useful but has potentially negative consequences. Less use of studded tyres would improve the air in several Nordic cities and optimal winter maintenance and traction control measures, e.g.

sanding and salting, should be a priority task to reduce the emissions. Speed limit reduction is likely to give effect also for the non-exhaust, but as it depends on the local situation, and factors like car fleet, level of congestion and driving behaviour, a general recommendation cannot be given.

As it is mainly urban traffic stations which record exceedances of the LVs it is wise to reduce the emissions at hot-spots and through local traffic mitigation. This local focus seems to already be in place in the cities as most measures reported in the TENs were indeed on a local level.

The need for modal change from passenger cars and motorbikes to low emission public transport, walking and biking will be important, considering the predictions of further urban growth. When accounting also for heavy duty vehicles and busses new technology and fuels will give emission reductions in the future, but an integrated urban planning will also be important to make good transport solutions also for this group with the end result of reducing the ADT. Measures like restriction in private car use will probably meet resistance in the population and may become unpopular. Better quantification of the actual traffic exhaust emissions on the road together with further focus on understanding the non-exhaust processes and formation of secondary particles would lead to more knowledge based recommendations for mitigation with the goal of reducing concentration levels.

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