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Review

Urban air quality: The challenge of traffic non-exhaust emissions

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HIGHLIGHTS

- Only few *in vivo* toxicity and epidemiological studies focused specifically on non-exhaust sources.
- Further experiments are needed to better separate individual contributions and health effects.
- Need of understanding of the interaction between road surface texture, moisture, chemistry, dust load and dust emission.
- Poor emission inventorying on resuspension and heavy metals.
- The optimal mitigation strategy for each climatic region is still unknown.

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ABSTRACT

About 400,000 premature adult deaths attributable to air pollution occur each year in the European Region. Road transport emissions account for a significant share of this burden. While important technological improvements have been made for reducing particulate matter (PM) emissions from motor exhausts, no actions are currently in place to reduce the non-exhaust part of emissions such as those from brake wear, road wear, tyre wear and road dust resuspension. These "non-exhaust" sources contribute easily as much and often more than the tailpipe exhaust to the ambient air PM concentrations in cities, and their relative contribution to ambient PM is destined to increase in the future, posing obvious research and policy challenges.

This review highlights the major and more recent research findings in four complementary fields of research and seeks to identify the current gaps in research and policy with regard to non-exhaust emissions. The objective of this article is to encourage and direct future research towards an improved understanding on the relationship between emissions, concentrations, exposure and health impact and on the effectiveness of potential remediation measures in the urban environment.

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Contents

1. Introduction

About 400,000 premature adult deaths attributable to air pollution occur each year in the European Region [1]. Road transport emissions account for a significant share of this burden. While important technological improvements have been made for reducing particulate matter (PM) emissions from motor exhausts, no actions are currently in place to reduce the non-exhaust part of emissions such as those from brake wear, road wear, tyre wear and road dust resuspension. These "non-exhaust" sources contribute easily as much and often more than the tailpipe exhaust to the ambient air PM concentrations in cities, and their relative contribution to ambient PM is destined to increase in the future, posing obvious research and policy challenges.

This report overviews the main outcomes of the international experts workshop "Urban Air Quality: The Challenge of Non-exhaust Traffic Emissions", held in Barcelona (Spain), in July 2013 jointly organized by BDebate (an initiative of Biocat and 'la Caixa' Foundation) and the Institute of Environmental Assessment and Water Research (IDAEA) of the Spanish National Research Council (CSIC) as an immediate follow up of a workshop held in Amsterdam, 2012 [2]. The aim of the present workshop was to highlight the major and more recent research findings in four complementary sessions and to identify the current gaps of research and policy with regard to non-exhaust emissions. The objective of this article is to encourage and direct future research towards an improved understanding on the relationship between emissions, concentrations, exposure and health impact and on the effectiveness of potential remediation measures in the urban environment. A complete overview of the workshop is available at http://www.bdebate.org/en/forum/urban-air-quality-challenge-non-exhaust-road-transport-emissions.

2. Health effects

Traffic-related PM plays an important role in the development of adverse health effects, as documented extensively in acute toxicity and epidemiologic studies [3–8]. Although there are few *in vivo* toxicity and epidemiological studies focused specifically on non-exhaust sources, the data that are starting to emerge indicate that non-exhaust PM can be as hazardous as tailpipe PM depending on the nature of the health effect studied.

Particle mass, size and (surface) chemistry all affect PM toxicity. One of the biological mechanisms causing toxicity is oxidative stress which is often related to transition metals and/or redox active organics such as quinone [9–11]. Brake and tyre wear particles have higher oxidative potential than other traffic-related sources and their effect is very local (50–100 m from the source) [12] yielding more oxidant PM (per $\mu g/m^3$) at road sites rather than at urban background sites. Tyre wear particles have been shown to induce Reactive Oxygen Species (ROS) formation and inflammatory reaction in human alveolar cells [13,14] as well as inflammatory response in mouse lungs [15,16]. Other important factors to be investigated are PM size and size distribution, particle number, composition (including coating and surface modifications), shape, surface area/specific surface area, surface chemistry, and charge and solubility/dispersibility.

Happo et al. [17] found significant inflammatory response in rats lungs exposed to coarse PM in Helsinki and correlated this with Fe and Cu content. A recent assessment of using ascorbic acid depletion (marker for presence of redox active metals), electron spin resonance (marker for $OH^{\bullet}$ radical) as well as DTT consumption (marker for redox active organics), showed a clear much higher oxidation potential of brake pad particles compared to diesel engine exhaust and tyre or road dust (F. R. Cassee, personal communication). Gustafsson et al. [18] showed at least as high inflammatory potential from road wear PM10 compared to diesel engine exhaust particles. Brake wear particles damage have been linked to oxidative stress and inflammatory responses in the lung using incubations of lung cells with brake wear particles [19].

Epidemiological studies related specifically to non-exhaust sources are still very few, again due to the difficult task of obtaining long time series of specific tracers and the lack of personal exposure data for risk assessment studies. Perez et al. [20] analyzed the respiratory, cardiovascular and cerebrovascular mortality risk associated with different PM size fractions in Barcelona, and found a significantly increased risk ratio (for coarse PM) of 5.9% and 9.8% for cardiovascular and cerebrovascular causes, respectively. Similarly, in Stockholm, Meister et al. [21] found that coarse particles (PM10–2.5) had a significant effect on daily mortality (1.7% per 10 $\mu g/m^3$ increase), while, across the Mediterranean region, Stafoggia et al. [22] reported associations between PM2.5–10 (and PM2.5) levels with cardiovascular and respiratory admissions. Source apportionment studies help identify the source-related health effects: Ostro et al. [23] identified a 4% increase of all-causes mortality risk for an interquartile range increase of road dust contributions only (in PM2.5), which was larger than the risk from vehicle exhaust emissions. Unpublished results from the MED-PARTICLES project, suggest an association between Fe and cardiovascular disease in Rome and Barcelona (J. Sunyer, personal communication), as well as for other non-exhaust tracers (Mn, Ti and Cu), supporting similar findings in the literature [24–26]. Although more research is necessary, especially that implementing source apportionment methods, there is already enough clear evidence to demonstrate the need for stricter PM10 guidelines.

3. Measurements and source contributions

There is no doubt about the serious environmental impact of non-exhaust emissions. Ambient air measurements across Europe have revealed a total non-exhaust contribution (wear emissions + resuspension) to PM10 comparable to that of tailpipe emissions, with a clear exacerbation in Scandinavian and Mediterranean countries due to winter tyres and drier climate, respectively [2,27–29]. Ketzel et al. [30] estimated that in several European countries a large part (about 50–85% depending on the location) of the total traffic PM10 emissions originates from non-exhaust sources. Moreover the lack of abatement measures for non-exhaust emissions has led to their increasing contribution to the PM airshed. In Southern Spain for example, from 2004 to 2011 road dust contributions to PM10 levels measured at a number of sites did not decrease, while motor exhaust contributions decreased ($p < 0.001$) by 0.4 (0.57–0.24) $\mu g\, m^{-3}\, year^{-1}$ [31]; similarly, at various European sites PM2.5 trends over last decade declined more rapidly than coarse PM [32]. Further increase in Europe in the coming years may also be boosted by the economic crisis, and the consequent poor maintenance of vehicles and roads and the possible increase of low-cost materials and technologies used with worse quality and faster degradation/erosion.

Further research is necessary to better separate individual contributions from road dust resuspension, brake, tyre and road wear given that the relative toxicity and mitigation measures are different. In this sense, valuable information can be offered by size and time-resolved PM chemical characterization and particle size distribution measurement, as well as improved source apportionment tools. Generally resuspension seems dominant in terms of mass, although its contribution can vary widely across Europe since road humidity dominates the emission potential: ~12% of PM10 in UK, 20–35% in Spain and Greece, up to 90% in Scandinavian countries during late winter and spring when studded tyres and road sanding contribute [31,33–39]. Wear emissions (and resuspension) are a major source for some metals [40]. A comprehensive inventory of tyres and brake composition in Europe is needed to serve as emission source profiles so future constrained source apportionment analyses would provide more reliable outputs. The analysis of elemental ratios in brake material and ambient air PM has revealed the significant contribution of brake disc abrasion [41]. Fractionation of total wear into size classes is also a major uncertainty. Average brake wear contributions vary from negligible up to 4 $\mu g/m^3$, or higher at specific traffic hotspots. At a regional scale, modelled brake wear emissions contribute up to 2 $\mu g/m^3$ [29,36,37,39,42,43] and were essential to explain observed ambient air concentrations of copper even at background sites [44]. Although the contribution of brake particles is not dominant in terms of mass, their health concern might be the most relevant due to their high bioreactivity as shown in session 1.

Tyre wear generally contributes the least of the non-exhaust sources (10% at Marylebone Road in London) [37] with a mean contribution to PM10 always below 2% [45]. Road wear contribution estimates are more common in Scandinavian countries where studded tyres generate high abrasion and airborne mineral dust is dominated by road wear particle suspension [35], but road wear can be substantial also where studded tyres are not used, due to low quality pavement material and constructions [46]. The contribution of road sanding and deposited Saharan dust are difficult to separate from that of pavement wear due to comparable chemical composition, as well as the interaction (sand wearing pavement) and its variation in time and space.

Overall there is need for new measurement studies aimed at understanding the interaction between road surface texture, moisture, chemistry, dust load and dust emission.

4. Emission inventory and modelling

EU Member States (MS) are committed, through the Convention on Long-range Transboundary Air Pollution (CLRTAP), to report their emission inventories (EI) including non-exhaust sources from road traffic. Current NFR (nomenclature for reporting) source categorization however only includes tyre wear together with brake abrasion as one source category (NFR 1.A.3.b.vi) and road surface wear emissions as a second source category (NFR 1.A.3.b.vii), and ignores the resuspension process which dominates PM emissions in some countries. Encouragingly in 2012 most MS reported national inventories for wear emissions, but the quality and consistency of data are questionable; for example, emission factors for the sum of brake and tyre wear show great variability among different MS, ranging from 0.01 (The Netherlands) to 0.5 $g\, veh^{-1}\, km^{-1}$ (Malta) [47]. The variability is also high for road wear (0.01–0.14 $g\, veh^{-1}\, km^{-1}$), although in this case this is more easily explained given that the highest values are reported from Finland and Sweden where studded tyres are used causing higher road wear. The major uncertainties and shortcomings in EI are due to:

1. Resuspension is not included in EI. This will affect compliance/uncompliance to the forthcoming revised PM National Emission Ceilings Directive and it is one of the reasons why models underestimate PM10 concentrations. Further improvements in emission estimates are urgently needed, given that the EPA AP42 model (www.epa.gov/otaq/ap42.htm) is inappropriate for European urban roads. Base emission factors are still lacking for many countries and their spatial and temporal variations are generally unknown. Spatially, emission factors vary depending on climate and type of road. Since only a very few emission estimates are as yet available in the literature, current Chemistry Transport Model performances are severely hampered [48,49]. More experimental observations are needed to cover different climatic conditions (mostly the Mediterranean region) and rural roads since they represent more than half of total emissions in Europe. Temporally, resuspension is heavily affected by season, precipitation and road moisture content [50,51]. For instance, after precipitation, the recovery of road dust emission potential follows an exponential curve, reaching 99% after 24 h in Spain and 72 h in The Netherlands [50]. The timing of emissions needs to be improved both in the short-term and long-term. Current estimates indicate that the resuspension caused by HDV traffic is roughly one order of magnitude larger than from LDV [52–55] but more studies on non-exhaust emission factors of trucks versus passenger vehicles are needed.
2. Improving resuspension emission modules (including the impact of meteorology, road operations, vehicle speed) is also a priority. Some recent parameterizations showed satisfactory results [50,51,56–58] but more observational data need to be gathered concerning road surfaces, dust loading, sanding, removal processes and source characterisations of emissions.
3. Some countries do not report on wear emissions, and those reported are likely to be affected by incompleteness and inconsistency in approaches. Major uncertainties arise from the use of inadequate tracers, uncertainties in measurements, variability of brake/tyre composition (manufacturer, vehicle type, through time). Standardized driving cycles and emission estimate methods are necessary for harmonization across Europe. Since the NFR does not include resuspension as a source category, countries that do or would like to include resuspension emissions will report it either under tyre and brake wear or road wear (see above) and this is likely to further increase inconsistency in the country reporting.
4. The current state of EI on metals (Cu, Zn, Pb, Ba, Sb) is too poor to enable their use as suitable tracers. No consistency is found between countries on brake wear emission factors, and many values are still missing for road wear. Scientific studies [44,59–61] showed that consistent bottom-up calculations of heavy metal emissions are feasible and can greatly aid the interpretation of ambient concentrations of these valuable tracers of non-exhaust emissions. However, it is important to realize that completeness of sources in EI is needed, at some locations the metal concentrations may partly originate from entirely different sources like non-ferrous metal production or industrial combustion. We need scientific, harmonized and consistent bottom-up inventories for tracers which will provide a valuable input for modelling and possibly exposure calculations in order

to understand more on the relationship between emissions, concentrations, exposure and health impact.

5. Mitigation and policy

The optimal mitigation techniques and strategies needed to abate PM non-exhaust emissions from road traffic are still an open question. The possible strategies to reduce non-exhaust emissions can be categorized as those aimed at minimizing the sources by (i) improving wear properties of materials and (ii) reducing the wear potential of traffic (e.g. studded tyres) [62] and those aimed at minimizing suspension to air by (i) removing/immobilizing dust from road surface (road cleaning), (ii) binding dust to road surface and (iii) adjusting traffic (less traffic, lower speed, less heavy vehicles). The optimum strategy likely involves a combination of the three categories.

Although more research is needed, recent studies suggest that beside the wear resistance of rock materials used for road pavement [63,64], other factors can reduce road wear emission potential: larger stone size, lower texture depth, road pavement construction type (e.g. rubber mixed asphalt, porous asphalt) and good operation and maintenance [65–69].

The lack of research focusing on preventive measures has directed interest towards mitigating measures, aimed at minimizing suspension to air. Road cleaning activities were found to reduce road dust resuspension only when water was used, due to the increase of road moisture content (rather than actual removal of dust). However, road washing activities were more effective in drier climates such as the Mediterranean region (up to 10% of daily PM10) while vacuum-sweeping alone did not provide evidence of effectiveness in the short-term ([70], www.redust.fi).

Several road dust binders have been tested such as Calcium Magnesium Acetate (CMA), $MgCl_2$, $CaCl_2$ and Potassium formate. However, most of these studies have been conducted in wet, cool climates (Sweden, Austria, Norway and Finland) in order to test binders after the use of studded tyres and road sanding ([62,71–73], www.life-cma.at, www.redust.fi). Results indicate large reductions of emission potentials and ambient air PM10 (up to 35% of daily mean). On the contrary, no or low evidence of effectiveness was found yet in Central and Southern Europe (UK, Germany and Spain) [74–76], except on industrial-construction roads, where the road dust load is high [74]. More observational studies are highly needed, since too few studies are available for Central-Southern Europe and for optimizing techniques and operation strategies in Nordic countries. In addition, a recent study found that CMA can increase locally concentrations of secondary inorganic aerosols due to the formation of dissolved $Mg(OH)_2$ and the consequent volatilization of NH_3 ([76], www.airuse.eu); this may be an unwelcome side effect of CMA that needs to be taken in consideration.

The measures targeted at reducing tyre, brake and road wear emissions are generally unexplored and offer much scope for future research. During the workshop, a preliminary survey was sent to participants from 11 European countries asking for what kind of technological measures should be investigated. The most common response (70%) was materials improvement that comprises road materials grain size, porosity and minerals, tyre design, brake composition and technology including dust collectors. 85% of responses identified a gap of legislation regarding non-exhaust emissions: new emission regulation (standardized braking cycle, ban of toxics, implementation of dust collectors) and/or air quality metric (toxic tracers, non-exhaust PM threshold) to be added to the current air quality directive [77].

Another important policy issue was also raised: as MS can subtract (anthropogenic) winter sanding/salting events from the number of PM10 exceedances, then why should Mediterranean countries be punished for their drier climate and more frequent (natural) Saharan dust intrusions, both of which provoke higher road dust emissions? Despite the justness of the argument, however as these PM components seem to have health outcomes in both regions, measures to abate or avoid these emissions are nevertheless needed if air quality improvements are to be achieved.

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