Biomarkers, 2009; 14(S1): 23–28

informa
healthcare

REVIEW ARTICLE

Sources and source contributions to fine particles

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Abstract
Fine dust is currently seen is one of the major issues in air quality as being responsible for causing negative health effects. Hence it is important to derive information on particle sources and their contribution to the ambient concentrations. This paper reviews two recent studies related to fine particle sources in Germany. The first study deals with the attribution of source regions to PM10 mass concentrations in nine agglomeration areas of Germany. The second study focuses on the diffusive sources from traffic differentiating and quantifying the various non-exhaust related emissions at an Autobahn.

Keywords: *Particulate matter, source regions, diffuse traffic emission, PMF*

Introduction

Fine dust, particles of aerodynamic diameters <10 µm, has been associated with different health effects in various toxicological and epidemiological studies. It is currently not definitely known which particle characteristic (e.g. chemical composition, morphology, reactivity) is the one/are the ones actually leading to the determined health effects. Epidemiological studies have shown, for example significant correlations between fine dust concentrations and mortality (Dockery et al. 1993), as well as cardiovascular effects (Hoffmann et al. 2007) and airway diseases (Krämer et al. 2009).

The 'First European Framework Directive for Air Quality' which was based on these health effect findings came into effect 1996 (Directive 96/62/EC). Still, after 10 years about 67% of agglomeration areas exceeded the daily limit value for PM_{10} (Figure 1). The revision of this directive and the daughter directives was published 2008, this time also based on results obtained from Integrated Assessment Modelling (Amann et al. 2004). The new air quality directive of the EU (Directive 2008/50/EC, 2008) is extending the focus onto the $PM_{2.5}$-dust fraction and onto the exposure of the public. The latter is obtained by setting measurement obligations for the determination of an average urban background concentration for agglomeration areas or larger cities.

These mass-related air quality indicators that are linked to limit values and action plans to be set up in cases of exceedances is the background of the studies presented here. Further reports and publications of source apportionment studies in Germany can be found in the following references (Lenschow et al. 2001, Kuhlbusch et al. 2003, Quass et al. 2007a, John and Kuhlbusch, 2004, Werkstatt Feinstaub 2009).

The first study deals with the attribution of source regions to PM_{10} mass concentrations in nine agglomeration areas of Germany (Quass et al. 2007b). The second study focuses on the diffusive sources from traffic

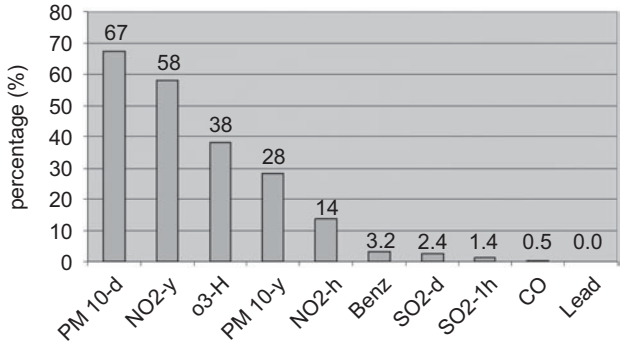

Figure 1. Percentage of zones and agglomeration areas in the EU exceeding the target and limit values of the EU in 2006 (De Leeuw & Vixseboxse 2007).

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ISSN 1354-750X print/ISSN 1366-5804 online © 2009 Informa UK Ltd
DOI: 10.1080/13547500902965377

differentiating and quantifying the various non-exhaust related emissions at an Autobahn (Quass et al. 2008).

A very recent overview on source apportionment studies in Europe was conducted within the framework of COST 633 and will soon be published (Viana et al. 2008).

Source regions of PM

The identification of source regions for air quality parameters and their specific contributions to the local air quality are the basis for assessments of possible abatement strategies and hence the potential success of action plans. Therefore a study based on PM_{10}-data for the years 2003–05 from the air quality networks in Germany was conducted (Quass et al. 2007b).

Agglomeration areas and methodology

The nine cities and urban agglomeration areas chosen were Berlin, Hamburg, Munich, Frankfurt and Hannover, Leipzig/Dresden, Ludwigshafen/Mannheim and Bremen as well as the Ruhr Area, represented by the cities of Dortmund and Duisburg at the easterly and westerly boarder. Figure 2 shows that the chosen agglomeration areas cover the whole area of Germany.

Two regional background, two rural background, and two urban sites were chosen for each of these areas. This selection of sites was extended by hot-spot sites, two traffic-related and, where available, two industry-related sites.

Average diurnal variations (1/2 hourly values) were calculated for each weekday based on the monitoring data, thereby reducing the dataset to 336 data points per year, site and parameter. This dataset was further processed by averaging the two corresponding site types per region in case of regional, rural and urban background. This averaging was not done for the hot spots to be able to differentiate possible local differences.

Spatial and temporal variability of contributions

Figure 3 shows on the left side the weekly profile of PM_{10}-mass concentrations averaged for the different site types. A clear increase in concentration from

Figure 2. Location of the nine cities and agglomeration areas (stars).

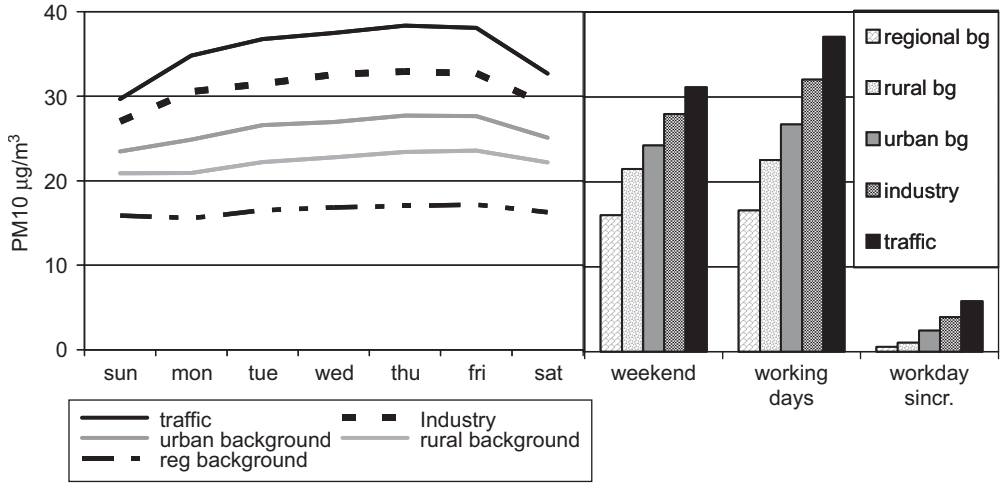

Figure 3. PM_{10} weekly variation of different site types (average of all sites and years) and comparison for weekend and work days.

regional background to hot spot location can be seen. Additionally an increase in PM_{10}-mass concentration during the course of the week with a maximum on Fridays is also evident. This trend depicts the build up of anthropogenic emissions during the week with decreasing concentration on Saturdays and Sundays, during periods of lower emission rates. Interestingly average minimum concentrations at regional background sites and often at rural background sites are determined for Mondays. This can be related to the lower emissions of particle precursors for secondary particles (NO_X, SO_2, etc.) on weekends which on average contribute around 30% to the PM mass and lead to lowest concentrations of for example PM-nitrate on Mondays.

Significant differences can sometimes be seen for industrial- and traffic-related sites. An example is given in Figure 4. PM_{10} mass concentrations at the industrial-related site in Ludwigshafen/Mannheim are more similar to the urban background than is the case for the example taken from the Ruhr Area.

Three-year average PM concentrations determined for the different site types are summarized in Figure 5 as a box-whisker plot.

Concentration gradients are evident between the different site types with increasing PM-mass concentrations from the regional background to the urban background and hot-spot sites. This gradient is larger for workdays than for weekend days. It is lowest if only Sundays are averaged (excluding Saturdays).

Gradients are in the range of 5–10 µg m^{-3}. This is in the range found in an analysis of the Landesamt für Natur, Umwelt und Verbraucherschutz NRW (LANUV, P. Bruckmann personal communication). Table 1 summarizes results obtained by the LANUV study for 2005 with the concentration increase, regression coefficient between the site types and ratio range of daily averages.

On average about 50% of the concentrations determined at traffic hot spots are measured at a corresponding regional background site. This value did not vary significantly between the years 2003 and 2005 despite quite significantly different meteorological conditions.

Diffuse traffic-related emission

Traffic contributions to ambient PM concentration can be related mainly to three emission sources: (1) direct

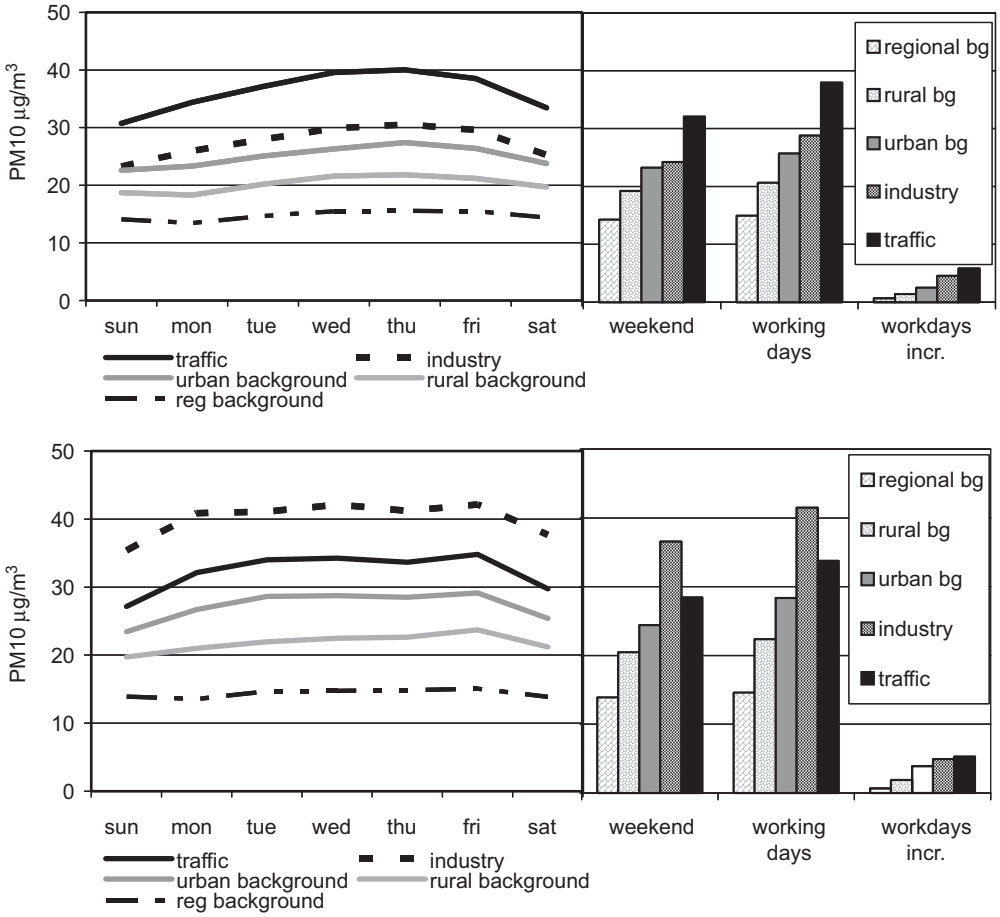

Figure 4. PM_{10} weekly variation of different site types for the areas of Ludwigshafen/Mannheim (above) and Ruhr Area (below).

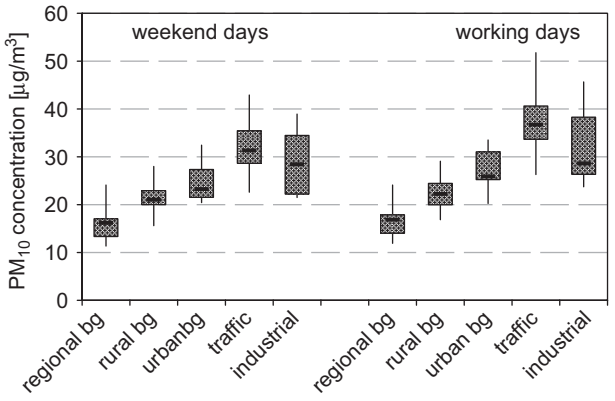

Figure 5. Box-whisker plot with minimum/maximum (vertical line), 25th and 75th percentile (Box), and median (horizontal line) of PM_{10}.

Table 1. Summary of concentration gradients of neighbouring sites in NRW.

Site	Difference (µg m^{-3})	r^2	Ratio of daily values (av.)
Regional/rural background	5–7		
Urban/rural background (medium size city)	4-9	0.2–0.7	1.10–1.60
Urban/rural background (agglomeration)	2–6	0.7–0.9	
Traffic site/urban background	3–7	0.7–0.9	1.1–1.4
Street canyon/urban background	8–15	0.7–0.8	1.3–1.6
Industry/urban background	6–15	0.4–0.6	1.2–1.5

primary particle tail pipe related emission (mainly soot with various amounts of (SV) OC, particle diameter <1 µm d_{ae} (Gehrig et al. 2003)); (2) diffuse emitted particles (tyre abrasion, break abrasion, street abrasion, resuspension of street dust, particles from corrosive processes; particle diameter mainly >1 µm d_{ae}); (3) secondary particle generated from gaseous precursors (mainly nitrate from NOx-emission, and VOC, SO_2; particle diameter <1 µm d_{ae}).

Secondary particles, according to process (3) above, are of minor importance for local traffic-related PM contributions (nevertheless a contribution of 1–2 µg m^{-3} in some cases is discussed). On the other hand, a huge fraction of NOx stems from traffic-related combustion processes and therefore a significant amount of ambient background PM-nitrate is due to traffic emission. NOx-emissions are quite well described due to regulatory obligatory emission testing and measurement of emissions.

Particulate tail pipe emissions are also determined during the regular emission test and emission factors are known for various types of vehicles and driving situations. The emission factors applicable to the German vehicular fleet are summarized in the handbook of emission factors HBEFA (UBA 2004).

Huge uncertainties on the other hand exist for diffuse traffic-related particle emissions. Some studies have been conducted especially in urban areas. These show that between 20 and 80% of the local traffic-related PM_{10} emission can be attributed to diffuse emissions, depending on traffic flow, fleet composition, road conditions, etc. General emission factors for diffuse emission are given for passenger cars and HDV (heavy-duty vehicle) for example by Düring and Lohmeyer (2004).

So far the contributions of the different diffuse sources (abrasion, resuspension, etc.) are not known with sufficient accuracy and therefore effect assessment of activities such as street sweeping cannot be modelled. Studies published so far focused on heavy trafficked tunnels and urban street canyons which lead to relatively high concentrations of PM; therefore facilitating studies in this direction (Rauterberg-Wulff 2000, Luhana et al. 2004). Results from these studies however can not simply be used for high speed rural roads and the Autobahn (motorways). To fill this gap a special project was conducted.

Data and methodology

Measurements were conducted from 01.09.2005 to 07.01.2007 at the A61 between AK Meckenheim and AD Bad Neuenahr. Average daily traffic was about 72 000 vehicles/day, with an HDV fraction of 20% (hourly averages 28%, min 2%, max 82%).

PM_1 and PM_{10}-mass concentrations were determined manually as well as automatically (1/2 hour resolution) at upwind and simultaneously downwind conditions along with NO and NO_2 concentrations. The mass concentration difference of the two sites during upwind–downwind conditions could directly be related to the traffic emissions, traffic counts, and NO–NO_2 emissions. The difference in PM_1 concentration was evaluated at first approximation as traffic-related tailpipe emission while the difference in $PM_{1\text{-}10}$ concentration (calculated from PM_1 and PM_{10} measurements) was interpreted as diffuse traffic-related emissions.

The NOx tracer method (Gehrig et al. 2003) was used for the determination of the total emission factors for PM_{10}, PM_1 and $PM_{1\text{-}10}$.

A collection of manually sampled PM_{10}-filters (24 h samples of 68 days) were chosen for chemical analysis based on meteorological conditions (wind direction, rain, wind speed). Filters of the PM_1-fraction were

analysed for the EC and OC content for the verification of tailpipe emissions. EC concentration difference in PM_1 and PM_{10} served as indicator for tyre abrasion. These data were subsequently used in a positive-matrix factorisation (PMF) analysis to obtain emission factors for tyre, break and street abrasion as well as resuspension of dust.

Results

Emission factors determined by the NOx tracer method are shown in Table 2. The values obtained for PM_{10} compare well with other studies (Gehrig et al. 2003, Düring & Lohmeyer 2004) and for PM_1 with values calculated based on the information given in HBEFA (UBA 2004). Significant differences were determined for emission factors of the PM_{1-10} fraction in comparison to values published by Gehrig et al. (2003). Nevertheless, a difference in the range of a factor of 2 can still be viewed as comparable taking the uncertainties in the determination of these factors into account.

Figure 6 shows the chemical composition of the selected PM_{10} samples. The typical secondary particle components sulphate, nitrate and ammonium contribute significantly to the total PM concentrations but are negligible for the local traffic-related contributions (difference downwind–upwind). Significant contributions were detected especially for carbonaceous compounds, NaCl and metals.

Table 3 summarizes the results obtained with PMF-modelling based on the chemical composition data.

Table 2. Emission factors for PM_x.

	VEH*[g km^{-1}*veh]	LDV*[g km^{-1}*LDV]	HDV*[g km^{-1}*HDV]	VEH**[g km^{-1}*veh]
EF PM_{10}	0.067	0.033	0.187	0.066
EF PM_1	0.039	0.017	0.119	0.030
EF PM_{1-10}	0.027	0.016	0.068	0.035

Veh, vehicle; LDV, passenger cars, light duty vehicle; HDV, heavy-duty vehicle.

*Based on 30 min values (TEOM); **based on daily averages (of days chosen for chemical analysis, $n = 68$).

Summary

The study related to source regions for PM_{10} showed that about 50% of the concentrations determined at hot spots are also determined at regional background sites. Nevertheless, significant weekly variations in PM concentrations even at regional background sites show the anthropogenic influence on PM concentrations.

Rural and urban contribution of about 15% add to the regional background concentration leaving on average about 20% of the hot-spot PM_{10} mass concentration to local activities, here traffic. Variance analysis of regional background concentrations as a fraction of hot-spot concentration did not reveal any significant differences for the years 2003, 2004, and 2005, this despite the highly different meteorological conditions between 2003 and 2005.

Diffuse traffic-related emissions which are currently not regulated can significantly contribute to local traffic-related emission. Only very few studies differentiating the various source processes have been conducted so far, and no study related to rural or autobahn driving conditions to our knowledge.

A corresponding study revealed that about 55–60% of the local traffic-related PM_{10} emission can be attributed to tailpipe emissions. The remaining 40–45% is attributed to break abrasion (ca. 14%), earth crust resuspension

Table 3. PMF results (estimated uncertainty range in brackets).

Source process	Fraction of PM_{10} emission [%]	Emission factor [mg km^{-1}*veh]
Salt resuspension (only winter period)	*13*	*9 (5–18)*
Break abrasion	14	9 (5–18)
Tyre abrasion	3	2 (1–3)
Resuspension, street abrasion	7	5 (3–10)
Tailpipe emission	57	38 (20–70)
Not identified, mainly resuspension	*6–19*	*~12 (6–24)*

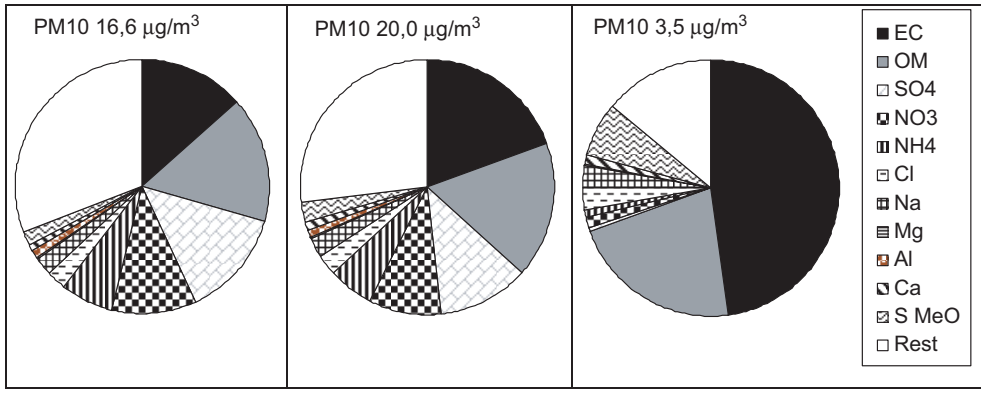

Figure 6. Average chemical composition of the selected PM_{10} samples (left: upwind samples, middle: downwind samples; right: traffic contribution).

(ca. 7%), resuspension of street salt (ca. 13%) and tyre abrasion (ca. 3%). HDV generally showed the highest emission factors, for total PM emission as well as for the diffuse source processes.

Acknowledgements

Thanks to the Umweltbundesamt, especially Arno Graff, for the financial support (360 11 007) and his contributions to the study related to source regions of PM in Germany. Thanks also to the BASt, especially Anja Baum, for the financial contribution (FE 02.0254/2004/LRB) and input to diffuse traffic-related emission. Thanks is extended to M. & M. Sulkowski and A. V. Hirner, Institut für Umweltanalytik an der Universität Duisburg-Essen, as well as J. Lindermann, M. Beyer, A. Hugo and U. Rating at IUTA e.V. for their contributions to this research project.

***Declaration of interest*:** The authors report no conflicts of interest. The authors alone are responsible for the content and writing of the paper.

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