

ATMOSPHERIC PARTICLE SIZE DISTRIBUTIONS FOR SOURCE APPORTIONMENT IN DRESDEN, SAXONY

Holger Gerwig¹ Erika Brüggemann² Thomas Gnauk² Konrad Müller² Antje Plewka²
Hartmut Herrmann²

¹ Section Regional Air Pollution Control, Climate Change, LFUG - Saxon State Agency for Environment and Geology, Dresden, Germany, Holger.Gerwig@lfug.smul.sachsen.de

² Department of Atmospheric Chemistry, Leibniz – Institute for Tropospheric Research, Leipzig, Germany

ABSTRACT

Higher concentrations of PM₁₀ (about 50% HVS, 20% Berner, 90% MOUDI) and PM_{2.5} (HVS) respectively PM_{3.5} (Berner) / PM_{3.2} (MOUDI) were measured at the traffic site compared to a nearby urban background site.

At 9-10-2003 air masses from the North Sea reached Dresden by long range transport and sea salt was responsible for about 15 – 20 % of PM₁₀. There was also a high amount of coarse nitrate.

At the traffic site in the period before heating EC concentrations were highest between 50 and 140 nm AeD (Aerodynamic Diameter).

INTRODUCTION

Epidemiologists have indications on the negative health effects of finer, finest and ultra fine particles. The focus on atmospheric research is directed on particles and their physical and chemical nature.

Size distributions and chemical composition of ultrafine (10 nm) up to coarse particles (10 µm) by analysing the different size fractions are the starting point for calculations about source apportionments in an urban environment [1-3].

METHODS

Berner low-pressure and Micro Orifice Uniform Deposit Impactors (MOUDI) were used 8 times on weekdays and one time on a weekend. Sampling took 24 h for Berner (108 m³ at 75 L/min m³) (0.05 - 10µm aerodynamic diameter (AED), mass, EC, OC (by a thermographic two step method)), ions (capillary electrophoresis and ion-chromatography), elements (Al; As; Br; Ca; Cl; Cr; Cu; Fe; Ga; K; Mn; Ni; P; Pb; S; Se; Si; Sr; Ti; Zn by PIXE), PAH, organic acids, alkanes (by Curie-Point-GCMS) and usually 96 h for MOUDI (172.8 m³ for particles >0.056 µm and respectively, 57.6 m³ for particles <0.056 µm) (0.01 - 18µm, mass, EC, OC, ions). Filter Samples of PM₁₀ and PM_{2.5} (Digital DHA 80, 720m³ in 24 h) (mass, EC, OC (Coulometry), ions (Ion-Chromatography), 11 elements (As; Cd; Cr; Cu; Mn; Ni; Pb; Sb; Ti; V; Zn) (GF-AAS), PAH (HPLC)) were collected for 24-h periods, 11 August 2003 until

25 April 2004 every second week for 7 days. The same sampling settings were used, from 9 until 29 February (winter) at a station of urban background. Additional parameters: NO, NO₂, BTX, CO, soot, wind direction, wind speed, temperature, humidity, global radiation. The numbers of automatic counting of vehicles of nearby streets [4] and additional meteorological information [5] was used as well.

Sampling at the urban traffic site (A) was located in Dresden near a crossing with a traffic load of about 55,000 vehicles per day. The sample inlets were about 4 m apart from the kerb and 3.5 m above ground. The site of urban background (B) was located about 400 m to north east and at least 100 m away from streets with less than 5,000 vehicles per day.

Backward trajectories were used for characterising the meteorological conditions [6]. Above ground level in 200, 500 and 2000 m were calculated for Dresden (51.47 N and 13.73 E).

RESULTS

High variability of the distributions of mass and compound classes in the size classes were found in the samples of Berner impactor and MOUDI (fig 1, 2).

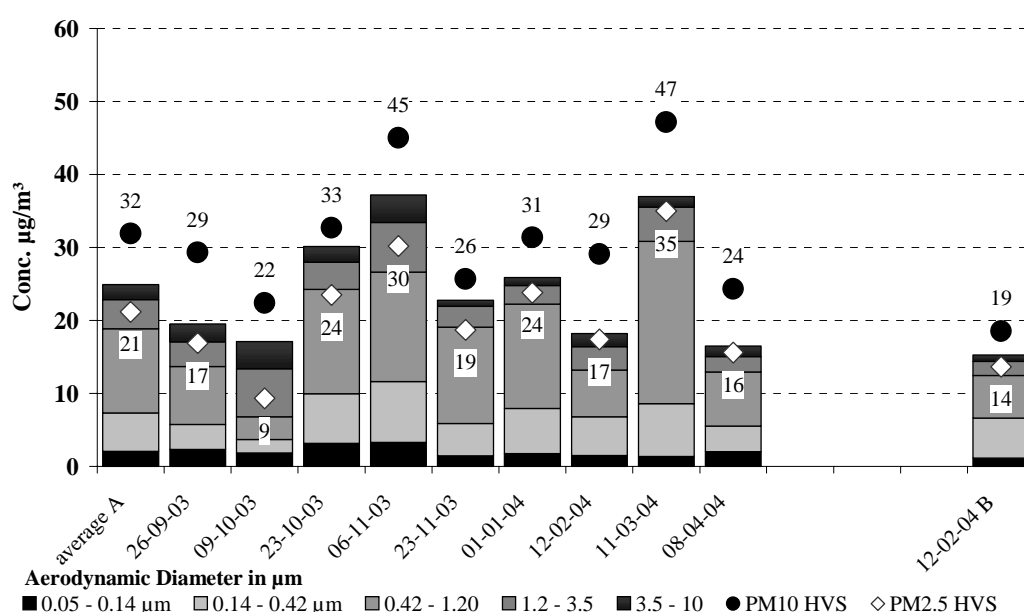


Figure 1: Mass in samples of Berner impactor and PM₁₀ respectively PM_{2.5} from HVS at traffic site (A, Sep-2003 to Apr-2004) compared with urban background site (B, Feb-2004).

During the winter measuring campaign at site A (near traffic) more coarse particles were found, than in site B (apart from highly frequented streets).

On the 12-02-2004 HVS Data are showing at A (29 µg/m³) one third more PM₁₀ than at B (19 µg/m³). The results of the Berner measuring are showing the same tendency but to a less great extent (A: 18, B: 15). During the 4 day sampling period of MOUDI (9 – 12-02-004) there was about the double amount found at site A (19) than at B (10 µg/m³) in PM₁₀. In the HVS samples A: 18, B: 12 µg/m³.

PM_{2.5} (HVS) respectively PM_{3.5} (Berner)/ PM_{3.2} (MOUDI) were less high in B compared to A.

Higher concentrations of Mn and Cu were found at the traffic site in Berner impactor and HVS samples. It was also found a significant correlation ($P > 99.9\%$, $R = 0,45$) between the sum of vehicle number of 4 different counting stations in the city centre, not direct at the site A and concentrations of Mn and Cu in PM₁₀ (HVS) [7].

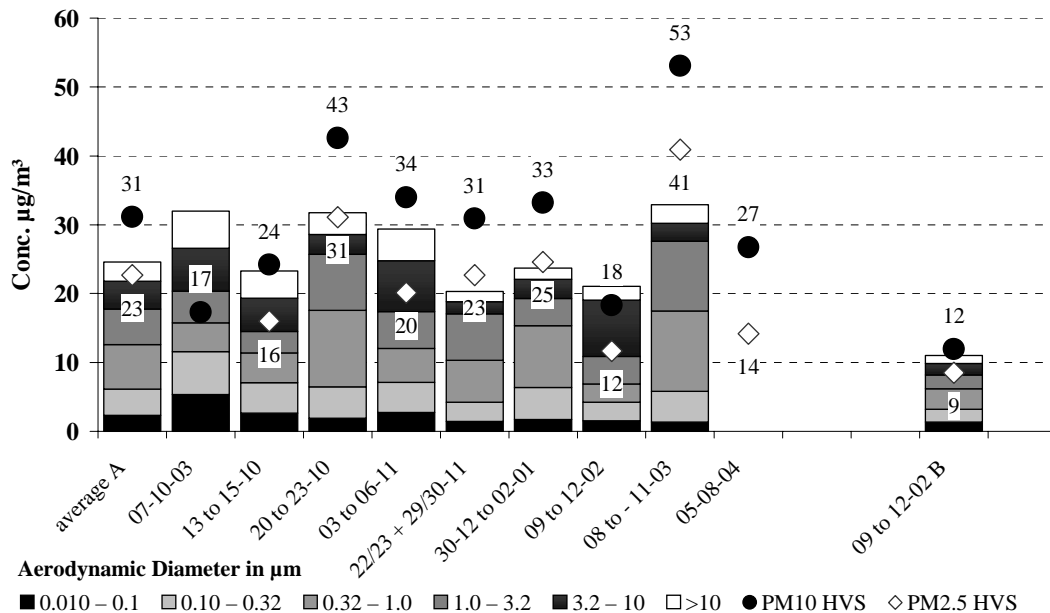


Figure 2: Mass in samples of MOUDI compared to PM₁₀ and PM_{2.5} from HVS at traffic site (A, Sep-2003 to Apr-2004) compared with site of urban background (B, Feb-2004).

HVS and impactor data agree well for sulphate but not for ammonium, especially on warm days [8].

Date	day of week	Temp. a) [°C]	Wind-direction [°] b)	Wind speed [mm/s] b)	deposition a) [mm]	vehicles per day d)	heavy duty vehicles per day d)
26-09-2003	Fr	13.9	120	1.8	0	154560	17196
09-10-2003	Th	9.1	270	3.7	0.5	145551	16592
23-10-2003	Th	-0.7	30	1	0.1	138941	16580
06-11-2003	Th	4.1	30	0.7	0	144997	16381
23-11-2003	Su	6.5	120	3.4	0	91587	4268
01-01-2004	Th	-3.0	30	1	0	-	-
12-02-2004	Th	-4.5	270	1	0 ^{c)}	-	-
11-03-2004	Th	2.9	120	1.9	0 ^{c)}	-	-
08-04-2004	Th	4.9	270	2.1	0.9	-	-

a) [5] DWD site Dresden-Klotzsche. b) Traffic site (A). c) Deposition occurred the day before. d) [4] Sum of 4 counting stations near the city centre.

Table 1: Weather conditions during Berner impactor sampling periods and sum of vehicles at 4 counting stations in the city centre of Dresden.

Sea salt in aerosol particles reacts in the atmosphere with acid anthropogenic compounds, like SO_2 , H_2SO_4 and HNO_3 under release of HCl and building of NaNO_3 and Na_2SO_4 [9].

Air masses came directly from North Atlantic to Dresden on 9-10-2003 (fig 1).

In the PM fraction 1.2 - 10 μm was 77 % of Nitrate (fig. 2), 28 % of sulphate and 92 % of chloride found. In PM_{10} the relation between Cl / Na ratio was 1.175 (Berner) und 1.018 (HVS). This is almost equal to the ratio in sea water with 1.164.

At the other sampling days stronger deviations from the Cl/Na -ratio were found, although with the exception of 23.11. and 11.3. air masses came from the sea (Tab 2). The mentioned compounds were more frequently found in the smaller fractions. In AED 1.2-10 μm less than the half was found: Nitrate 5– 33 % (Fig. 4); sulphate 5- 11 %. The Backward trajectories show that the air masses were about 6 to 12 hours above the continent.

Date	Weather conditions / source of backward trajectories
26-9-2003	High pressure bridge over central Europe. No Rain./ From North Sea, air masses were going over black triangle region (South west of Poland and North west of Czech Republic).
09-10-2003	North west condition, cyclonal; marine air masses from arctic to Central Europe. Moderate rainy weather. / From Northern America and North Atlantic straight to Dresden.
23-10-2003	High north over north east Atlantic. Dry cold arctic air masses. Moderate rainy weather. / From Baltic states.
06-11-2003	High Fenoskandia, anti cyclonal; partly inversions. Foggy weather without precipitation. / From North and Baltic Sea over Poland.
23-11-2003	South West condition, anti cyclonal. Almost no Rain in southern part of Germany. / From Northern Italy, Swiss and Southwest Germany.
01-01-2004	Arctic cold air masses. Slightly snow fall. / From North sea, North Atlantic over Scandinavia along the German - Polish borderline.
12-02-2004	High over British Islands. Cold air masses from north. Slightly snow or drizzle. / Air masses from Scandinavia and arctic sea.
11-03-2004	South East conditions, cyclonal. Snow fall on the 10. no rain on 11. / Air masses coming from south east and eastern directions.
08-04-2004	Trough over Western Europe. Low pressure area. Shower of Rain. / From North and Baltic sea and with longer time over northern part of Germany.

Table 2: Conditions during Berner impactor sampling periods: weather characteristic [5] and source of 96 h backward trajectories [6].

The share of NaCl in measured PM_{10} was 20 % (Berner) respectively 15 % (HVS). That was 3.4 $\mu\text{g}/\text{m}^3$ sea salt (NaCl) of 17.4 $\mu\text{g}/\text{m}^3$ (Berner) respectively 22.4 $\mu\text{g}/\text{m}^3$ (HVS). In 2003 at the traffic site an annual mean of daily averages of 36 $\mu\text{g}/\text{m}^3$ PM_{10} was found.

The average size distribution of NH_4^+ compared to SO_4^{2-} was very similar. It is found in the fine fraction like described by Funasaka [2] . The most of the unimodal distributed mass was found between 0.4 – 1.2 μm (Berner) and 0.32 – 1.8 (MOUDI) (Funasaka [2] < 2.1 μm).

At the traffic site with about 55,000 vehicles per day in the period before heating EC (elemental Carbon) concentrations were highest between 50 and 140 nm AeD, probably originated by soot from traffic.

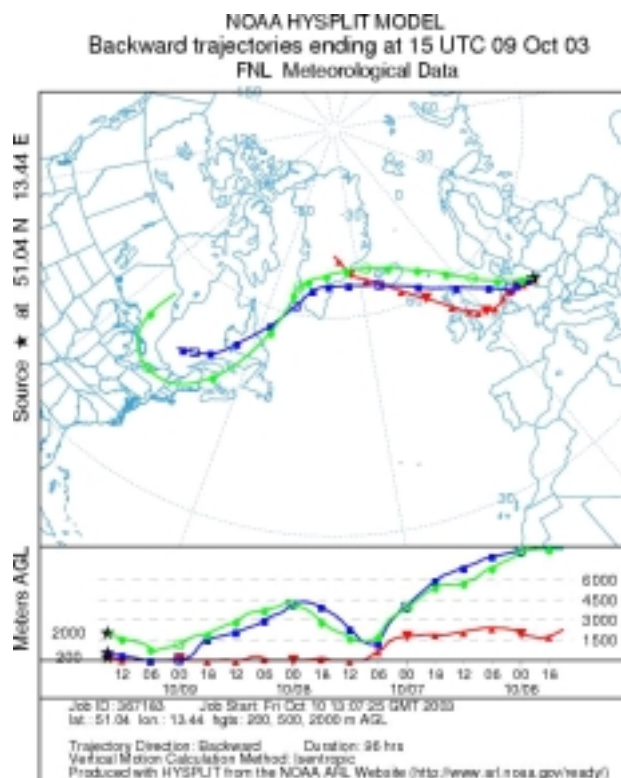


Figure 3: 96h backward trajectories at 9-10-2003, 15:00 UTC with target Dresden [6].

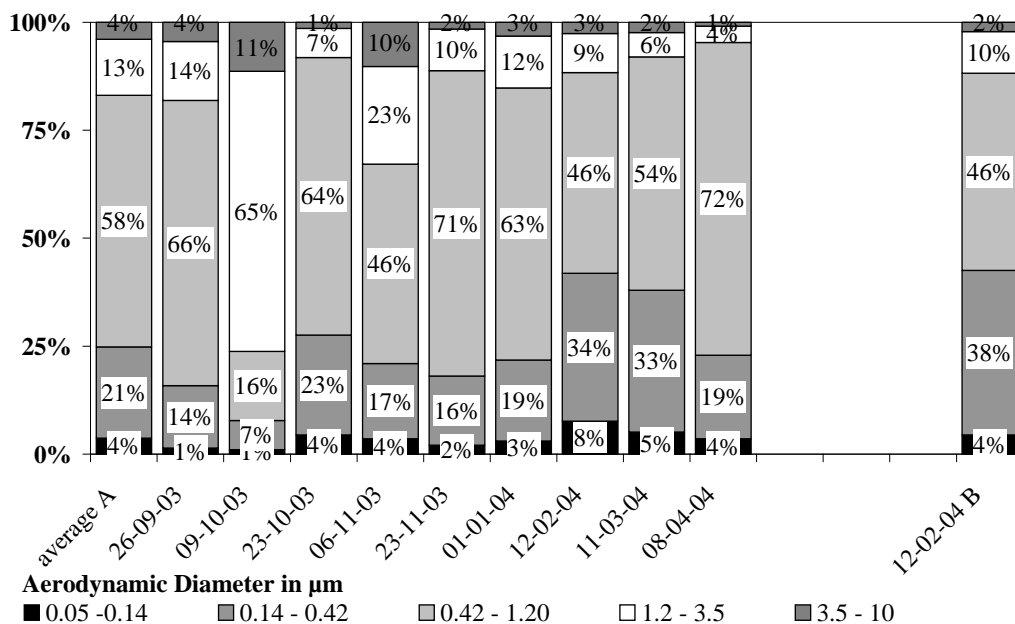


Figure 4: Nitrate in samples of Berner impactor and PM_{10} respectively $\text{PM}_{2.5}$ from HVS at traffic site (A) compared with site of urban background (B).

CONCLUSIONS

Long range transports during eastern high pressure periods with wind from South East resulted in the highest PM₁₀ concentrations (11-3-04). Transport over sea results in lower PM₁₀ concentrations with higher amounts of sea salt and coarse NaNO₃ because of not deposited or washed out coarse particles from aged marine aerosol of NaNO₃ and Na₂SO₄.

Traffic emissions and other fossil fuel emissions (especially from heating) are influencing the EC-concentrations.

The number of vehicles nearby the measuring rise PM₁₀ and the concentrations of Mn and Cu.

ACKNOWLEDGEMENTS

The authors wish to thank the Staatliche Umweltbetriebsgesellschaft for sampling PM₁₀ and PM_{2.5}. The city of Dresden (Straßen- und Tiefbauamt, Abt. Verkehrstechnik) provided numbers of automatic counting of vehicles. The Department of Umwelt Service, TÜV Bau und Betrieb GmbH, Dresden, Germany made the filter analyses. The authors wish also to thank Erik Swietlicki and Per Kristiansson, University of Lund, Sweden for analysing the trace elements in Berner Impactor samples by PIXE.

The Saxon state Agency provided data regarding online measurements. The project is financed by the Saxon State Agency for Environment under reference number 13-8802-3520/10 (<http://www.umwelt.sachsen.de/lfug> (see „Luft, Lärm, Klima“, Veröffentlichungen, Materialien zur Luftreinhaltung).

REFERENCES

- [1] Brüggemann, E.; Franck, U.; Gnauck, T.; Müller, K.; Neusüß, Ch.; Plewka, A.; Spindler, G.; Stärk, H.-J.; Wennrich, R. & Herrmann, H. (2000): Korngrößendifferenzierte Identifikation der Anteile verschiedener Quellgruppen an der Feinstaubbelastung, final report for Saxon State Agency for Environment and Geology, Zur Wetterwarte 11, 01109 Dresden, Germany (Reference number 13-8802.3521/46).
- [2] Funasaka, K.; Sakai, M.; Shinya, M.; Miyazaki, T.; Kamiura, T.; Kaneco, S.; Ohta, K.; Fujita, T. (2003): Size distributions and characteristics of atmospheric inorganic particles by regional comparative study in Urban Osaka, Japan, Atmospheric Environment - International ASIA 37, 4597-4605.
- [3] Pakkanen T.A.; Loukkola, K.; Korhonen, C.H.; Aurela, M.; Mäkelä, T.; Hillamo, R.E.; Aarnio, P.; Koskentalo, T.; Kousa, A. and Maenhaut W. (2001a): Sources and chemical composition of atmospheric fine and coarse particles in the Helsinki area, Atmospheric Environment, 35, 5381-5391.
- [4] Landeshauptstadt Dresden (Straßen- und Tiefbauamt, Abt. Verkehrstechnik (2004): Automatische Straßenverkehrszählungen in Dresden für das Jahr 2003.
- [5] DWD Deutscher Wetterdienst (2003, 2004): Witterungs-Report express.
- [6] Draxler, R.R. and Rolph, G.D. (2003): HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (<http://www.arl.noaa.gov/ready/hysplit4.html>). NOAA Air Resources Laboratory, Silver Spring, MD.
- [7] Gerwig, H. (2004): Near traffic source apportionment in the City of Dresden, Saxony (PART I: PM₁₀ and PM_{2.5}); Poster presentation at the European Aerosol Conference 6.-10.9.2004 in Budapest.
- [8] Gerwig, H.; Herrmann, H. (2004): Impaktorprobenahmen zur Klärung der Herkunft von PM₁₀ an einer Straßenkreuzung in Dresden; in: 39. Messtechnisches Kolloquium, 17. - 19. Mai 2004 in Hamburg.
- [9] Plate, E. (2000): Variabilität der Zusammensetzung anorganischer Aerosole - insbesondere der reaktiven Stickstoffverbindungen - in küstennahen Gebieten der Nordsee und Ostsee; Dissertation, Universität Hamburg Fachbereich Chemie, in Schriftenreihe Angewandte Analytik, Hrsg. W. Dannecker, Bd. 37 (<http://www.sub.uni-hamburg.de/opus/volltexte/2000/135/pdf/Gesamt.pdf>).