Multi-channel statistical analysis of background fine particle aerosols

G. Gramotnev[1] and P. Madl[2]

[1]Applied Optics Program, School of Physical and Chemical Sciences, Queensland University of Technology, GPO Box 2434, Brisbane, QLD 4001, Australia, GPO Box 2434, Brisbane, QLD 4001, Australia.

[2] Department of Molecular Biology, Division of Physics and Biophysics, Division of Physics and Biophysics, University of Salzburg, A-5020 Salzburg, Austria

Universität Salzburg

Division of Molecular Biology – Department of Physics and Biophysics
Dosimetry and Modelling Working Group

Abstract

Nano-particles are one of the major types of air pollutants in urban indoor and outdoor environments. Therefore, determination of mechanisms of formation, evolution, and transformation of combustion aerosols near major sources, e.g. busy roads and road networks, is one of the most essential and urgent goals. This work addresses the particular direction of research by filling some gaps in the existing physical understanding of aerosol behavior and evolution. The applicability of the Gaussian plume model to combustion aerosols near busy roads is discussed and used for the numerical analysis of aerosol dispersion. New methods of determination of emission factors from the average fleet on a road from different types of vehicles are presented. Strong and fast evolutionary processes in combustion aerosols near a busy road are documented experimentally, interpreted, modelled and statistically analysed. A new major mechanism of aerosol evolution based on intensive thermal fragmentation of nano-particles is proposed, discussed and modelled. A comprehensive interpretation of mutual transformations of particle modes, a strong maximum of the total number concentration at an optimal distance from the road, and an increase of the proportion of small nano-particles far from the road is suggested. Modelling of the new mechanism is developed on the basis of the theory of turbulent diffusion, kinetic equations, and theory of stochastic evaporation degradation processes.

Introduction

In this paper, new statistical methods for the determination and investigation of particle modes and their mutual correlations in One and ultra-fine panicle aerosols in the presence of strong turbulent mixing (Gramotnev & Gramotnev, 2005a) are applied for the analysis of urban background aerosols. In particular, several distinct modes will be obtained from the background particle size distribution, and their possible sources will be discussed. The meteorological parameters during the measurements are presented in Table 1.

Monitoring of background aerosols was conducted in the Brisbane area, Australia (Gramotnev et al., 2004), on the upwind side of the Gateway Motorway, so that traffic emissions from the road did not affect the obtained background data. There were no buildings within ~150 m from the measurement area, which was practically a flat grass field with isolated scattered bushes and trees. No particular sources of air pollutants are known within at least several kilometers upwind from the sites of measurement. The distance from the curb of the Motorway was ~60 m. This distance was sufficient for not registering any noticeable particle concentrations coming directly from the road due to turbulent diffusion.

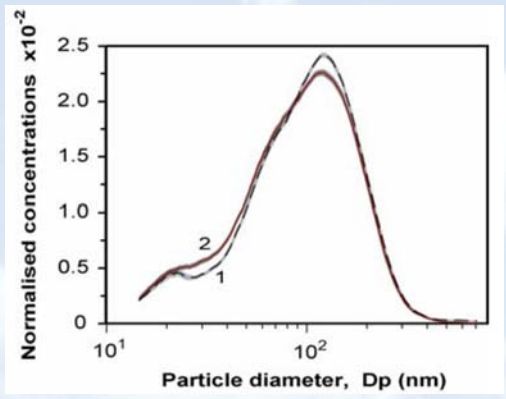

Figure 1. The moving average size distributions for the two sets of scans: (I) scans from 11 to 27 (higher solar radiation), and (2) scans from 22 to 38 (negligible solar radiation).

Methods

Some 38 scans of particle size distribution in the background aerosol were measured in the range from 13 um to 763 nm in the afternoon on 30 July 2002 using a TSI SMPS-system. All concentration measurements were conducted simultaneously along with measurements of temperature (as 20 °C), humidity (a 40%), solar radiation (<300 W·m^{-2}), wind speed (~1.5 m·s^{-1}) and wind direction in intervals of 20 seconds. Two separate sets of 17 scans (from 11 to 27 and from 22 to 38) were chosen out of the overall 38 scans. This choice was made primarily on the basis changing solar radiation. The entire campaign was conducted during time of sunset with the second set of scans corresponding to nearly zero solar radiation, while for the first set of scans it was still significant. As shown in figure 1, the average particle size distributions for both sets of scans does not reveal significant features and/or noticeable changes.

Table 1: Meteorological Parameters for the sampling campaign

Temperature, [°]	20.5 ±0.9
Humidity, [%]	39 ±4
Solar radiation, [W·m^{-2}]	200 ±100
Wind speed, [m·s^{-1}]	1.7 ±0.8
Wind direction to the road, [°]	46 ±50

Results

Nevertheless, the statistical analysis based on the moving average correlation coefficient (Gramotnev & Gramolnev, 2005a) reveals substantial differences in particle modes for the two sets (Figure 2). Correlations between neighbouring channels of the size distribution significantly change indicating noticeably different processes of particle evolution/transformation. Considering a moving interval and calculating the average correlation coefficient for all pairs of channels within this interval, gives the moving average correlation coefficient and its standard error of the mean as functions of particle diameter for the two selected sets of 17 scans.

One of the important features of Figure 2 is the existence of strong and distinct maxima of the moving average correlation coefficients for both sets of scans. Concentrations of particles in the channels within every such maximum tend to increase / decrease in maximum correlation with each other. This implies that a significant fraction of these particles is likely to come from the same source or originate from the same physical / chemical processes. Therefore, we use the term mode for a group of particles corresponding to a maximum of the moving average correlation coefficient (Gramotnev & Gramotnev, 2005a). Any such local maximum can be called mode, if it is statistically significant, i.e. if its height is larger than the width of the error band, and it lies above an acceptable level of confidence for the determined correlations.
Contrary to the original size distributions shown in figure 1, at least four statistically significant modes can be seen in figure 2. This is a clear demonstration of the effectiveness of this statistical approach. It is likely that the modes with smaller particle diameters originate from transport off the road network, because they were previously observed in combustion aerosols near a busy road (Gramotnev & Gramotnev, 2005a).

The second important feature of figure 2 is that the particle modes substantially change between the two sets of the 17 scans. If the sets of 17 scans is moved through the 38 scans (by taking the 17-scan set between 1 and 17, 2 and 18, 3 and 19, etc.), it can be seen that the strong differences between the correlation patterns shown in figures 2 develop in a consistent way through several steps. This is an indication of significant evolutionary processes in urban background aerosols, which strongly depend on and interact with solar radiation.

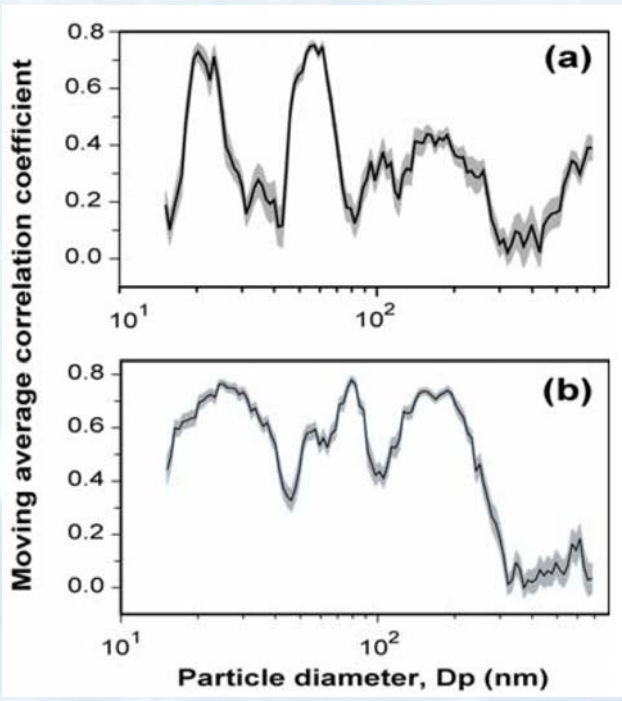

Figure 2. The moving average correlation coefficients wilh their errors as functions of particle diameter for the two sets of scans: (a) before sunset, and (b) alter sunset.

Conclusion

Multi-channel correlation analysis has been used for the determination of the effect from solar radiation, temperature, and humidity on aerosol particles. It has been shown that humidity has a negligible effect *on* background particle modes. This suggests that, at least at these observed low levels of humidity, mechanisms of formation of background particle modes are not related to humidity. Whereas decreasing temperature results in a stronger increase of the 170 nm mode (see Figure 1). This can be related to the fact that these particles experience thermal fragmentation (Gramotnev & Gramotnev, 2005a, 2005b), which becomes inefficient at lower temperatures, leaving more particles of the same type in this mode. Statistical weights and loadings for particles between ~20 nm and ~35 nm are both negative for solar radiation, which suggests that concentration of these particles significantly increase with decreasing solar radiation. Explanation of this effect can again come from the fragmentation theory. Indeed, fragmentation of these modes into smaller particles is expected to become weaker with decreasing solar radiation, leaving more particles in the considered modes.

This paper demonstrates that fragmentation mechanism of aerosol evolution is characteristic not only to combustion aerosols near busy roads (Gramotnev & Gramolnev, 2005a, 2005b), but also to background urban aerosols. Which of the particles fragment and at what rate fragmentation occurs, remains unclear at this stage and requires further investigation. Yet we demonstrated that the developed statistical methods of analysis based on the moving average correlation coefficients are indeed important for all types of aerosols and can reveal new fundamental features and processes of evolution of different types of particles in the atmosphere. Yet, determination of cross-correlation coefficients could provide further evidence and is scheduled for a follow-up investigation using the same data sets used so far.

Nonetheless are the so far obtained results important for accurate prediction of aerosol pollution levels in both outdoor and indoor environments. Already at this stage they can be used for a reliable determination of human exposure and impact of transport emissions on the environment both on local as well as on global scales. This work will also be important for the development of reliable scientifically-based national and international standards for nano-particle emissions.

References

Gramotnev G., Z. D. Ristovski, R.J. Brown, & P. Madl. (2004) New Methods of Determination of Average Particle Emission Factors for two Groups of Vehicles on a Busy Road. *Atmospheric Environment* 38 (2004): 2607-2610

Gramotnev, D. K. & Gramotnev, G. (2005a). A new mechanism of aerosol evolution near a busy road: fragmentation of nano-particles. *J. Aerosol Science*, 36, 323-340.

Gramotnev, D, K. & Gramotnev, G. (2005b). Modelling of aerosol dispersion from a busy road in the presence of nanoparticle fragmentation, *J. Applied Meteorology*, 44, 888-899.