Available online at www.sciencedirect.com
ScienceDirect
Atmospheric Environment 41 (2007) 3521–3534
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Multi-channel statistical analysis of combustion aerosols. Part I: Canonical correlations and sources of particle modes

G. Gramotnev, D.K. Gramotnev*

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

Received 27 January 2006; received in revised form 28 December 2006; accepted 9 January 2007

Abstract

A major problem with the analysis and investigation of combustion aerosols in the real-world environment is related to strong stochastic variations of the external and environmental parameters and factors (e.g., atmospheric turbulence, traffic fluctuations, etc.). Therefore, this paper develops new powerful statistical methods based on the canonical correlation analysis and the moving average technique, applied to combustion aerosols near a busy road. As a result, a new physical insight into the evolution of combustion aerosols and possible sources of nano-particle modes is presented and discussed. Several new particle modes are identified, analysed and associated either with trucks or cars on the road. In particular, liquid and solid particle modes are identified, and the mechanism of thermal fragmentation of solid nano-particles is used for the interpretation of the obtained results.

Keywords: Combustion aerosols; Statistical analysis; Canonical correlations; Sources of particles

1. Introduction

Recently, increasingly convincing evidence appear that fine and ultra-fine aerosol particles (within the ranges <1 and $<0.1\,\mu$m, respectively) emitted from combustion sources may present significant health risks for humans in the urban environment (Schwartz et al., 1996; Stone, 2000; Brown et al., 2000). Since busy roads are the main source of such particles (Shi et al., 1999, 2001), major theoretical and experimental efforts have been concentrated on the analysis of dispersion and evolution of fine and ultra-fine particle aerosols from vehicles on roads (Shi et al., 1999; Hitchins et al., 2000; Zhu et al., 2002; Gramotnev et al., 2003; Gramotnev and Ristovski, 2004; Zhang and Wexler, 2004; Zhang et al., 2004; Gramotnev and Gramotnev, 2005a, b; Davison et al., 2006). Attempts were also made to investigate particle modes from vehicle exhaust under laboratory conditions, using hot dilution and thermodesorption for separating volatile and solid components in the aerosol (Abdul-Khalek et al., 1998; Sakurai et al., 2003; Fierz and Burtscher, 2003).

Zhu et al. (2002) and Gramotnev and Ristovski (2004) demonstrated that significant transformation of particle modes may occur near a busy road. This could hardly be explained using the known

*Corresponding author. Tel.: +61 7 3864 2593; fax: +61 7 3864 1521.
E-mail address: d.gramotnev@qut.edu.au (D.K. Gramotnev).

1352-2310/$ - see front matter
doi:10.1016/j.atmosenv.2007.01.031

mechanisms of aerosol evolution, such as particle formation, coagulation, condensation/evaporation and deposition (Gramotnev and Gramotnev, 2005a; Davison et al., 2006). Therefore, a new major mechanism of aerosol evolution, based on intensive thermal fragmentation of nano-particles, was developed (Gramotnev and Gramotnev, 2005a). A complex evolution pattern was suggested, including several stages such as formation of solid and liquid nano-particles inside and near the exhaust pipe, their coagulation near the exhaust pipe by means of condensed volatile molecules, evaporation of volatile compounds from the surface of solid particles as the aerosol is transported away from the road, loss of bonding between coagulated nano-particles due to evaporation of bonding volatile molecules and, finally, thermal fragmentation of nano-particles (Gramotnev and Gramotnev, 2005a). Numerous experimental evidence supporting this evolution model were presented, including substantial transformation of particle modes and their shift towards smaller particle size (Gramotnev and Ristovski, 2004; Gramotnev and Gramotnev, 2005a), observation of a maximum of the total number concentration at an optimal distance from the road (Gramotnev and Ristovski, 2004; Gramotnev and Gramotnev, 2005a, b; Davison et al., 2006), observations of strong modes that would be expected as a result of fragmentation (Gramotnev and Gramotnev, 2005a), etc.

A new method of statistical analysis of particle modes in combustion aerosols near a busy road was developed based on the moving average correlation coefficients between neighbouring channels in the particle size distribution (Gramotnev and Gramotnev, 2005a). This method allows determination and analysis of particle modes in the presence of strong turbulent mixing, even if these modes are not directly seen (as distinct maxima) on the size distribution. However, detailed statistical analysis of these modes and their relationship to meteorological and traffic parameters, determination of their possible sources and origins have not been done so far. At the same time, these questions are important for the detailed understanding of fundamentals of aerosol evolution, accurate forecast of aerosol pollution levels, and reduction of the impact of transport emissions on human health and environment.

Therefore, the aim of this paper is to develop and use new statistical approaches for the detailed analysis of modes of the particle size distribution near a busy road, including their possible sources, mutual transformations, correlations with atmospheric and meteorological parameters. This will be done by the extension of the previously developed method based on the moving average technique (Gramotnev and Gramotnev, 2005a) to the multivariate canonical correlation analysis. In particular, modes resulting primarily from heavy diesel trucks and cars will be identified, the dependence of these modes on temperature, humidity and solar radiation will be analysed.

2. Experimental data and particle modes

The development of the new statistical methods and their application for the analysis of particle modes will be conducted on the basis of the experimental data previously discussed in (Gramotnev and Gramotnev, 2005a). The data were obtained during the field campaign on 25 November 2002 near Gateway Motorway, Brisbane, Australia. The particle size distributions were measured by means of a scanning mobility particle sizer (SMPS-3936) and a condensation particle counter (CPC-3025). Fifty scans were taken during ~ 3 h of measurements at the distance of $L \approx 40$ m from the centre of the road, within the range of particle diameters from 4.6 to 163 nm in 100 equal intervals (channels) of $\Delta\log(D_p)$, where D_p is the particle diameter in nanometres (Gramotnev and Gramotnev, 2005a). The time for 1 full scan was 2.5 min, with a 1 min down-scan. The width of the Motorway was ≈ 27 m, and its elevation above the surrounding area was ≈ 2 m.

Traffic was recorded on a video camera and subdivided into 2 groups: heavy-duty trucks and cars (the car group included gasoline cars, diesel cars and light trucks). Numbers of vehicles in each of these 2 groups were determined within the 2.5 min intervals by means of direct counting from the videotape. The beginning of each of the 2.5 min intervals was taken L/v_n seconds earlier than the beginning of the corresponding scan; v_n is the 1-h average normal component of the wind. Thus, we took into account average time delays associated with the aerosol transport from the road to the point of monitoring.

Wind speed, wind direction, temperature, humidity, and solar radiation were measured every 20 s by a automatic weather station at the same distance from the road. In the paper by Gramotnev and Gramotnev (2005a), the 2 sets of scans (out of the