

Aerosol Evolution from a Busy Road in North-West England

BRIAN DAVISON^{1*}, DUNCAN WHYATT¹ and CARL BOARDMAN²

¹Lancaster Environment Centre, Lancaster University, Lancaster, United Kingdom

²Department of Earth Sciences, The Open University, Walton Hall, Milton Keynes, United Kingdom

(Manuscript received December 14, 2006; in revised form May 29, 2008; accepted July 06, 2008)

Abstract

Motor vehicle emissions are the most significant source of particulate matter (PM) in urban environments. This study was undertaken to observe the evolution of aerosols downwind from a busy road, concentrating specifically on the aerosol total number maximum and number size distribution. A Grimm Aerosol Technik (5.400) CPC and DMA 5.5-900 classifier were used to measure ultra-fine particles from 9.8 nm to 1.1 μm at varying distances up to 100 m from the road side. Contrary to current accepted aerosol theory particle number concentration was seen to rise with increasing horizontal distance from the road side up to a maximum distance of about 100 m. As this occurred the number of fine particles was seen to increase as numbers of larger particles declined. These findings are discussed in light of existing aerosol literature on aerosol formation and dispersion.

Zusammenfassung

Kraftfahrzeuge stellen die wichtigste Aerosolquelle in städtischen Umgebungen dar. Im Rahmen der vorliegenden Arbeit wurde die Entwicklung von Aerosolen im Lee einer verkehrsreichen Straße beobachtet. Dabei wurde das Hauptaugenmerk auf das Maximum der totalen Aerosolanzahl und auf die Anzahlgrößenverteilung gerichtet. Zur Messung ultrafeiner Partikel von 9.8 nm bis 1.1 μm Durchmesser in unterschiedlichen Abständen von bis zu 100 m von der Straße wurden ein Grimm Aerosol Technik (5.400) CPC und ein DMA 5.5-900 Klassifizierer verwendet. Im Gegensatz zur gegenwärtig anerkannten Aerosoltheorie wurde beobachtet, dass die Partikelanzahlkonzentration mit zunehmender horizontaler Entfernung von der Straße bis zu einer maximalen Entfernung von etwa 100 m zunimmt. Dabei stellte sich eine Zunahme der Anzahl der kleinen Partikel heraus, während die Anzahl der größeren Teilchen abnahm. Diese Ergebnisse werden im Bezug zur verfügbaren Literatur über Aerosolentwicklung und Dispersion diskutiert.

1 Introduction

Road vehicles are one of the main sources of ultra fine particles (particle diameter $<0.1 \mu\text{m}$) in the urban environment and a growing body of evidence recognises their effect on human health and global climate change (DONALSDON et al., 1998; WICHMANN and PETERS, 1997; VEDAL, 1997; JACOBSON, 2001; HOUGHTON et al., 2001). In urban areas 80 % of particle matter in terms of number is associated with ultra fine ($<100 \text{ nm}$) particles (MORAWSKA et al., 1999). While the USA and European Union have legislation regulating atmospheric concentrations of coarse particulate mass less than $10 \mu\text{m}$ (PM_{10}) and fine particle mass less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) (STOKSTAD, 2006) there is nothing to govern particle numbers density which are normally dominated by fine and ultra fine particles in the atmosphere.

It is currently unclear whether mass concentration or number concentration is the most important in relation to adverse health effects (OSUNSANYA et al., 2001; PETERS et al., 1997; TUCH et al., 2006). Ultra fine particles can penetrate deep into the lungs and epidemiological studies have shown an association with respiratory illness and increased cardiovascular disease (DOCKERY et

al., 1993; WARHEIT et al., 2004). Concerns have also been raised over the toxicity of nano particles and their entry into the body through the skin (OSUNSANYA et al., 2001; BROWN et al., 2000; WARHEIT, 2004; DONALDSON et al., 1998).

While studies have shown that ultra fine particles are more toxic than larger particles with the same chemical composition and the same mass concentration (BROWN et al., 2000; DONALDSON et al., 1998, 2001) the exact mechanisms of toxicity are unclear and at present only PM_{10} and $\text{PM}_{2.5}$ are regulated. A greater understanding of the effects on health could be gained from a more in-depth study of number concentration and size distribution of ultra fine particles. Understanding the decline of vehicle emitted particles as they are transported from the road and transform in the atmosphere would considerably aid this goal.

This study was undertaken in a region with relatively clean background air in order to assess the evolution of aerosol formation from a single busy road, concentrating on total number maximums and number size distributions.

2 Location

Lancaster, situated approximately 3 km from the coast in north-west England, experiences predominantly clean

*Corresponding author: Brian Davison, Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, United Kingdom, e-mail: b.davison@lancaster.ac.uk

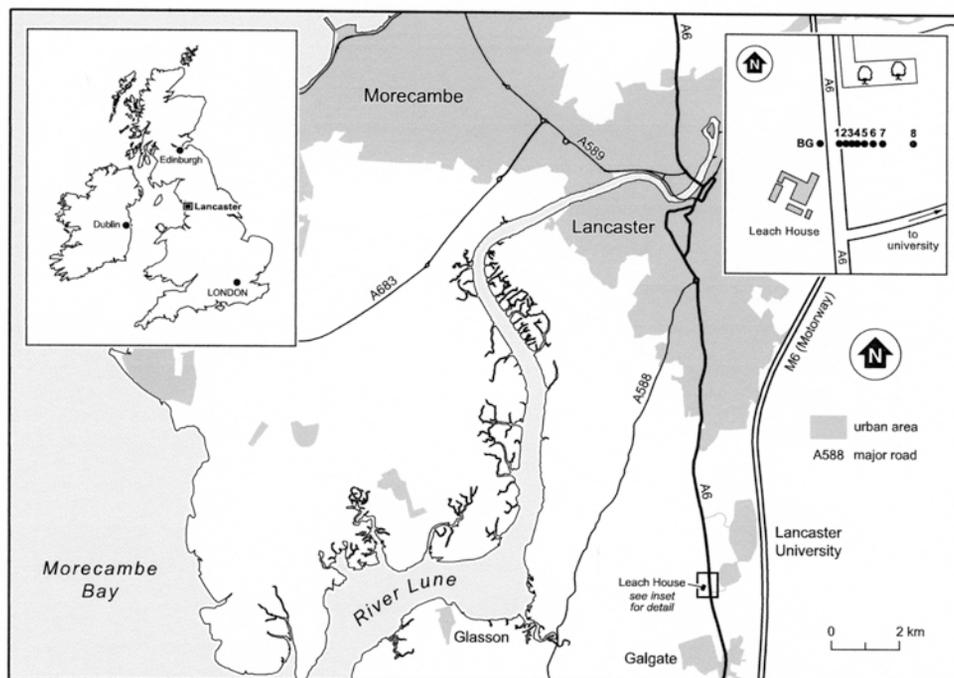


Figure 1: Map of sampling location. Inserts show the relative position of Lancaster in the UK and the positions of the background sampling site and the eight downwind sampling locations.

west-south-westerly winds and a maritime climate reflecting the passage of air masses across the Irish Sea. Background aerosol concentrations are low, typically around $1000 \text{ particles cm}^{-3}$ when summing the size range of the classifier used in this study. The A6 road to the south of the city runs parallel to the coast and therefore perpendicular to the prevailing wind direction making it ideal to study the dispersion and evolution of vehicle emitted aerosols.

Some locations such as large conurbations or multiple lane highways where urban or road aerosol studies have been undertaken are affected by multiple aerosol sources and a high traffic flow counts so leading to a high aerosol loading. In contrast this site with its prevailing maritime air mass, low aerosol count and few other sources of aerosols offers a ideal location to undertake this study.

Figure 1 shows the location of the site and the individual sampling stations to the south of Lancaster University. There are few other roads in the vicinity to cause contamination of the air masses crossing the sampling site.

The field and section of road used for sampling is not screened by vegetation. Such vegetation barriers are commonly used alongside roads in the UK to screen the road from nearby dwellings and also help cut down noise. This also has the effect of filtering gases and aerosols emitted from the vehicles.

Downwind sites were located at 3, 6, 10, 15, 25, 40, 60 and 100 m from the road side. Measurements were also taken at a background site 20 m upwind of the road.

3 Method

A Grimm Aerosol Technik (5.400) CPC and DMA 5.5-900 classifier were used to measure ultra fine particles from 9.8 nm to 1100 nm with a measurement error of 2 %. This was mounted on a trolley for ease of movement between sampling stations. The sampling inlet to the DMA stood approximately 90 cm off the ground and above any of the grass vegetation of the sampling area. The use of the system in this way ensured that no additional inlet line was necessary and any inlet losses were as characterised by the manufacturer.

Five replicate aerosol size spectra were collected at each sampling location taking approximately 20 minutes. These spectra were averaged to give the spectra shown. Additional repeat samples were taken at selected distances during the day to check for significant changes in the aerosol number and size spectra and verify reproducibility of the results.

Sampling days were selected with similar characteristics to avoid variations due to meteorological or other unconsidered factors. Sampling data were collected over six days during two summer periods during sunny summer days (May–August) from mid morning until mid afternoon (approximately 10 am to 4 pm local time) to maintain a constant traffic flow and avoid possible increases in traffic during rush hour. Wind speed, direction and temperature were monitored and were typically $1 \pm 0.1 \text{ m/s}$, westerly with a temperature of around $21 \pm 2^\circ \text{ C}$. Cloud cover varied from 1 to 7 oktas, however, no link could be identified between this and the recorded

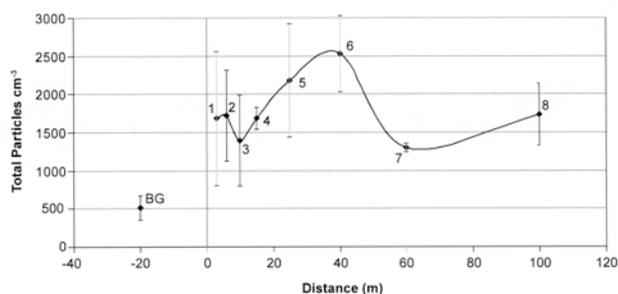


Figure 2: Change in mean total particle number density with distance from the road, June 2005. Error bars are for 1 standard deviation. The measurements at -20 m represent background readings.

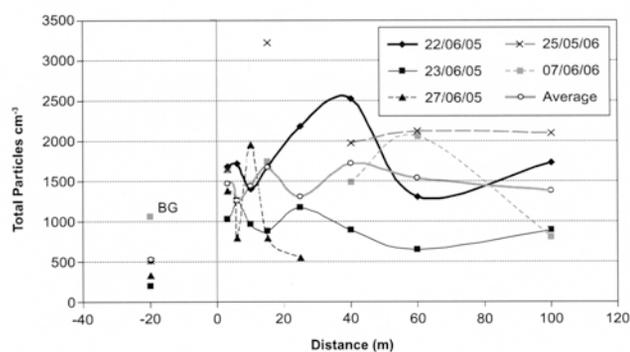


Figure 3: Change in mean total particle number density with distance from the road for several measurements over 2005 and 2006. The average is also shown. The measurements at -20 m represent background readings.

aerosol spectra. A traffic survey was undertaken during the sampling and in general about 1200 ± 300 vehicles an hour passed the site with a ratio of approximately 5:1 of cars to heavy goods vehicles.

4 Results

At each site total particle numbers were summed across the size range of the instrument. Figure 2 shows the changes in particle number density with horizontal distance from the road. From the sampled data presented in Figure 2, collected on 22 June 2005, the particle number maximum occurred at 40 m. Data collected on several other days presented in Figure 3 showed a similar trend with a total particle number maximum occurring around 30 to 60 m from the road. The error bars shown in Figure 2 are one standard deviation of the mean aerosol count for that sampling location and show the greatest variation in aerosol number occurred closer to the road. This greater variability can be explained by the higher turbulence caused by the passing of traffic. For ease of comparison with previous studies particle numbers were summed into selected size bands to investigate changes in particle size with distance from the road. As would

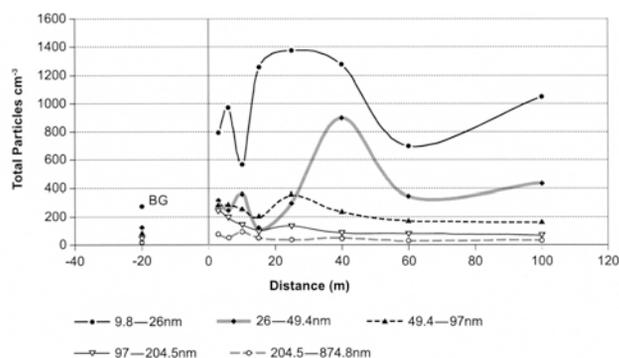


Figure 4: Averaged particles number summed over selected sizes. The measurements at -20 m represent background readings.

be expected the highest number of particles was found to be present in the smallest size range 9.8 to 26 nm which rose to a maximum of nearly 1400 particles cm^{-3} between 20 and 40 m from the road (Figure 4). The background aerosol concentration for this size range was generally less than 300 particles cm^{-3} , well below that measured at the downwind sampling sites.

Figure 4 shows a decline in particles >26 nm at 15 m from the road which could be explained by dilution, however this coincides with a rise in finer particles <26 nm. Whether this rapid rise in the $9.8 < \text{Particle diameter} (D_p) < 26$ nm particles is due to growth of ultra fine particles smaller than those measured by our instruments or some unknown process is uncertain.

The maximum in total particles at 40 m shown in Figure 2 is due to a combined maxima in two size ranges, $9.8 < D_p < 26$ and $26 < D_p < 49.4$ nm. At this distance from the road side source we would have expected dilution to have caused a reduction in particle numbers in this size range, so an increase in particle numbers in this size range must be occurring as we move from 25 to 40 m. This could be either a rapid growth of particles from outside the measurement range of our instrument, or an as yet unconsidered mechanism. Similar findings were observed on other days with similar meteorological and traffic conditions.

5 Discussion

5.1 Other road side studies

In recent years a number of groups have made measurements of total particle number concentration in the vicinity of roads (ZHU et al., 2002a; SHI et al., 1999; HITCHINS et al., 2000; MORAWSKA et al., 1999). These measurements were often conducted as part of other experiments in urban areas with other roads in close proximity. Hence the locations may not have been chosen specifically to investigate particle size distribution and their evolution with distance from the road source. This has led to a sometimes confusing array of findings.

MORAWSKA et al. (1999) found no appreciable decline in particle numbers with distance from roadside. An increase in particle numbers at a different site was attributed to vehicle activity. A higher absolute number concentration was attributed to greater traffic activity around a slip road and did show a decline in total particle number away from the road. This, however, was explained by the mixing of different air masses at the sites due to local topography.

One of the first attempts to study the size distribution of particles emitted from road vehicles and their subsequent evolution was made by ZHU et al. (2002a; 2002b). Measurements of aerosol number and size spectra in the range 6 to 220 nm were undertaken at a series of distances (17 to 300 m) away from a busy highway in Los Angeles. They found exponential decay a good estimator of particle total number decrease with distance from the highway. They suggested atmospheric dilution and coagulation to be important processes in the rapid decrease of particle number concentration away from the highway. A shift of 20 nm size particles to 30 nm as the air parcel moved from 20 to 30 m downwind of the highway was attributed to coagulation being the dominant factor of particle transformations in open air near a highway.

Other researchers have produced contradictory results. GIDHAGEN et al. (2003) and STURM et al. (2003) found coagulation to be significant only within the confines of a road tunnel and the interpretation of rapid coagulation has been questioned by some. KETZEL and BERKOWICZ (2004) reinterpreting the work of ZHU et al. (2002a) suggested this growth was in fact due to dilution which is related to the ratio of particle numbers in background air to that emitted from the vehicle.

GRAMOTNEV and RISTOVSKI (2004) also reinterpreted the findings of ZHU et al. (2002a) in light of the results from their own similarly designed road side sampling in Australia. They suggest the 20 nm mode observed by ZHU et al. (2002a) at the location 17 m from the road centre did not grow into the 30 nm mode at the 20 m location but shifted to smaller size particles. This shifting of particle size modes to smaller sizes contradicts most generally accepted aerosol growth and dispersion theories. GRAMOTNEV and RISTOVSKI (2004) went on to present their own findings obtained over several days of observations. Aerosol size spectra were measured at varying distances downwind from the road side with a crosswind passing over the road. Ultra fine particles from 4 to 710 nm were measured at varying distance up to 300 m from the road. A traffic count showed around 4000 vehicles per hour passed the site, 20 % of which were trucks. A maximum in total particle number was observed some distance from the road (generally between 40 to 100 m) and, pertinently, they suggested a growth in a 12 nm size mode away from the road up to a distance of approximately 90 m. The decline of a maximum in the 30 nm mode is attributed to a decrease in

particle size as particles of 20 nm size are seem to increase in number with distance from the source.

GRAMOTNEV and RISTOVSKI (2004) observed a maximum at around 80 m which was attributed to a strong growth of particles in the 30 nm mode. The data of ZHU et al. (2002a) shows a maximum in 25 to 50 nm particles around 40 m from the road. In that data this coincided with a decline in finer 6 to 25 nm particles and also those in the next larger size range 50 to 100 nm. In this study we observe a maximum in total particles approximately 40 m from the road side and a gradual increase up to 100 m which is also shown in the data of ZHU et al. (2002a). As their study extended to 300 m from the road they were able to observe a subsequent decline after this small increase.

Some of these studies have been conducted in major conurbations such as Birmingham (UK) and Los Angeles (US) (SHI et al., 1999; ZHU et al., 2002a) with significantly higher traffic counts (12000 vehicles per hour, as opposed to 1200 per hour during this study). The associated higher background count encountered at such sites may mask the detailed trends observed in this and other studies conducted under more moderate traffic flow regimes.

These findings of a maxima in total particle numbers away from the road and increases in the number of finer particles as those of larger particles decline are not easily explained by conventional aerosol dynamics or represented with the current generation of dispersion models. Such models tend to consider aerosols to behave in a simplified standard manner at ambient temperatures with a reduction in particle number with distance travelled from the source due to dilution, deposition and particle growth occurring due to condensation and coagulation processes (CARRUTHERS et al., 2003).

5.2 Possible Mechanisms

There is a growing body of evidence showing that particle number distribution increases away from a busy road and this appears to relate to increases in fine and ultra fine particles coinciding with a decline in the number of larger particles. The process is rapid, occurring in less than one minute and as yet the mechanism is unclear.

GRAMOTNEV and GRAMOTNEV (2005) suggest a mechanism of thermal fragmentation. The dispersion of the exhaust plume as it moves away from the road lowers the concentration of volatiles within the plume due to dilution. As the concentration of volatiles in the plume approach ambient concentrations the volatile molecules then begin evaporating from the surface of the particle so the particle size begins to reduce. If losses due to evaporation are sufficiently high the energy required for thermal fragmentation may be reached energy producing greater numbers of smaller particles.

KETZEL and BERKOWIEZ (2004) suggested the apparent growth in particles is due to increased levels of dilution of finer particles. The effect of dilution on an aerosol is more efficient the smaller the particles and so as the exhaust plume moves away from its source and is diluted, proportionally the larger aerosol modes appear to increase.

Another possible mechanism is that of a “blast” or “plough wave” effect which has also been proposed to explain the decline in particle numbers close to the road. The shock wave formed by an explosive emission can cause the air to rise from ground level and sink again some distance from the source. In the case of an explosion this can have the effect of leaving areas less affected by the blast. The passage of vehicles along the road may have the same effect upon the exhaust plume causing it to rise, so reducing the measured particle numbers at ground level close to the road. Particle numbers then increase some distance from the road when the plume touches the ground again.

The data observed in this study is not easily explained by conventional mechanisms of particle growth and dispersion. Work is ongoing to elucidate the mechanism which causes these transformations of aerosol from traffic emissions near a busy road.

6 Summary

Particle size spectra have been measured at increasing horizontal distances downwind from a busy road in North-West England. The maximum of total particle was found some distance (40 m) from the road side rather than close to the source of particles. An increase in particles (<50 nm) was also observed to occur around this distance from the road so causing the observed maximum in total particle numbers. Such observations cannot be accounted for by current aerosol growth theories or represented with the current generation of dispersion models. Alternative mechanisms to explain these observations have been considered and further work is in progress to understand the process and ascertain the extent and significance of these increases in particle numbers. Only when the mechanisms are understood can we appreciate the significance of this phenomenon and develop regulative aerosol dispersion models accordingly.

Acknowledgments

The authors would like to thank Simon CHEW, Division of Geography, Lancaster Environment Centre, for his assistance in the preparation of the figures.

References

BROWN, D.M., V. STONE, P. FINDLAY, W. MACNEE, K. DONALDSON, 2000: Increased inflammation and intracellular calcium caused by ultrafine carbon black is independent of transition metals or other soluble components. – *Occup. Environ. Medicine* **57**, 685–691.

- CARRUTHERS, D., L. BLAIR, K. JOHNSON, 2003: Comparison of ADMS-Urban, NETCEN and ERG Air Quality Predictions for London. – In: Cambridge Environmental Research Consultants Ltd (DEFRA).
- DOCKERY, D.W., C.A. POPE, X.P. XU, J.D. SPENGLER, J.H. WARE, M.E. FAY, B.G. FERRIS, F.E. SPEIZER, 1993: An Association Between Air-Pollution And Mortality In 6 United-States Cities. – *New England J. Medicine* **329**, 1753–1759.
- DONALDSON, K., X.Y. LI, W. MACNEE, 1998: Ultrafine (nanometre) particle mediated lung injury. – *J. Aerosol Sci.* **29**, 553–560.
- DONALDSON, K., V. STONE, A. CLOUTER, L. RENWICK, W. MACNEE, 2001: Ultrafine particles. – *Occup. Environ. Medicine* **58**, 211.
- GIDHAGEN, L., C. JOHANSSON, J. STROM, A. KRISTENSSON, E. SWIETLICKI, L. PIRJOLA, H.C. HANSSON, 2003: Model simulation of ultrafine particles inside a road tunnel. – *Atmos. Environ.* **37**, 2023–2036.
- GRAMOTNEV, D.K., G. GRAMOTNEV, 2005: A new mechanism of aerosol evolution near a busy road: fragmentation of nanoparticles. – *J. Aerosol Sci.* **36**, 323–340.
- GRAMOTNEV, G., Z. RISTOVSKI, 2004: Experimental investigation of ultra-fine particle size distribution near a busy road. – *Atmos. Environ.* **38**, 1767–1776.
- HITCHINS, J., L. MORAWSKA, R. WOLFF, D. GILBERT, 2000: Concentrations of submicrometre particles from vehicle emissions near a major road. – *Atmos. Environ.* **34**, 51–59.
- HOUGHTON, J., 2001: The science of global warming. – *Interdisciplinary Sci. Rev.* **26**, 247–257.
- JACOBSON, M.Z., 2001: Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. – *Nature* **409**, 695–697.
- KETZEL, M., R. BERKOWICZ, 2004: Modelling the fate of ultrafine particles from exhaust pipe to rural background: an analysis of time scales for dilution, coagulation and deposition. – *Atmos. Environ.* **38**, 2639–2652.
- MORAWSKA, L., S. THOMAS, D. GILBERT, C. GREENAWAY, E. RIJNDERS, 1999: A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road. – *Atmos. Environ.* **33**, 1261–1274.
- OSUNSANYA, T., G. PRESCOTT, A. SEATON, 2001: Acute respiratory effects of particles: mass or number? – *Occup. Environ. Medicine* **58**, 154–159.
- PETERS, A., H.E. WICHMANN, T. TUCH, J. HEINRICH, J., HEYDER, 1997: Respiratory effects are associated with the number of ultrafine particles. – *Amer. J. Respiratory and Critical Care Medicine* **155**, 1376–1383.
- SHI, J.P., A.A. KHAN, R.M. HARRISON, 1999: Measurements of ultrafine particle concentration and size distribution in the urban atmosphere. – *Sci. Total Environ.* **235**, 51–64.
- STOKSTAD, E., 2006: Environmental regulation - New particulate rules are anything but fine, say scientists. – *Science* **311**, 27–27.
- STURM, P.J., U. BALTENSPERGER, M. BACHER, B. LECHNER, S. HAUSBERGER, B. HEIDEN, D. IMHOF, E. WEINGARTNER, A.S.H. PREVOT, R. KURTENBACH, P. WIESEN, 2003: Roadside measurements of particulate matter size distribution. – *Atmos. Environ.* **37**, 5273–5281.
- TUCH, T.M., O. HERBARTH, U. FRANCK, A. PETERS, B. WEHNER, A. WIEDENSOHLER, J. HEINTZENBERG, 2006: Weak correlation of ultrafine aerosol particle con-

- centrations < 800 nm between two sites within one city. – *J. Exp. Sci. Environ. Epidemiol.* **16**, 486–490.
- VEDAL, S., 1997: Ambient particles and health: Lines that divide. – *J. Air & Waste Manag. Ass.* **47**, 551–5–81.
- WARHEIT, D., 2004: Nanoparticles: Health impacts? – *Materials Today* **7**, 32–35.
- WARHEIT, D.B., B.R. LAURENCE, K.L. REED, D.H. ROACH, G.A.M. REYNOLDS, T.R. WEBB, 2004: Comparative pulmonary toxicity assessment of single-wall carbon nanotubes in rats. – *Toxicological Sci.* **77**, 117–125.
- WICHMANN, H.E., A. PETERS, 2000: Epidemiological evidence of the effects of ultrafine particle exposure. – *Philosophical Transactions of the Royal Society of London Series A: Mathematical Physical and Engineering Sciences* **358**, 2751–2768.
- ZHU, Y.F., W.C. HINDS, S. KIM, C. SIOUTAS, 2002a: Concentration and size distribution of ultrafine particles near a major highway. – *J. Air & Waste Manag. Ass.* **52**, 1032–1042.
- ZHU, Y.F., W.C. HINDS, S. KIM, S. SHEN, C. SIOUTAS, 2002b: Study of ultrafine particles near a major highway with heavy-duty diesel traffic. – *Atmos. Environ.* **36**, 4323–4335.