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Human exposure to traffic pollution. Experience from Danish studies*

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Abstract: Air pollution may have severe long-term as well as short-term health effects. The determination of possible links between pollution levels and impact on human health is, however, not a straightforward task. A key problem is the assessment of human exposure to ambient pollution levels. In later years, the possible role of particulate pollution as a health hazard has drawn major attention and is, therefore, the subject of research projects in many countries including Denmark. The present paper gives a review of recent and ongoing/planned Danish air pollution exposure studies. Furthermore, key results from Danish studies of ultrafine particles from urban traffic are outlined. The exposure studies show that air pollution models may be strong tools in impact assessment studies, especially when used in combination with personal exposure monitoring and application of biomarkers. Personal exposure measurements in Copenhagen indicate that indoor pollution levels may be very important for the personal exposure to fine fraction particles ($PM_{2.5}$). Measurements with a differential mobility analyzer (DMA) in Danish urban areas show that number concentrations of ultrafine particles (<100 nm) in busy streets are strongly correlated with classic traffic pollutants such as nitrogen oxides and carbon monoxide. The number concentrations in urban Danish streets have decreased considerably between two campaigns in 1999 and 2000, apparently as a result of reductions in sulfur contents in Danish diesel fuels that took place in July 1999.

INTRODUCTION

Human exposure to air pollution is believed to cause severe health effects, especially in urban areas where pollution levels often are high. The classic example is the severe London smog (smoke and fog) episode in 1952 where the mortality rate in the city increased dramatically [1]. However, health effects may also be significant when they cannot be detected as easily as in connection with such a severe episode. Studies of long-term exposure to air pollution (especially particles) suggest an increased risk of chronic respiratory illness (see, e.g., refs. 2–4), and of developing various types of cancer (see, e.g., refs. 5–7). In an apparently worst case scenario carried out on the WHO data sets, Künzll *et al*. [8] found that 6% of deaths in Austria, France, and Switzerland might be associated with exposure of the popu-

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lation to particulate air pollution. Many factors influence human health, and a good assessment of human exposure is crucial for a proper determination of possible links between air pollution and health. A discussion on how to carry out assessments of human exposure to air pollution can be found in Hertel *et al.* [9].

In Denmark as well as in many other countries, traffic is the most important source of high pollution levels in cities and thereby also high exposure of the population in the ambient environment. A number of Danish investigations have been carried out with the aim of characterizing pollution from traffic with main focus on nitrogen oxides, hydrocarbons and, in later years especially, particles. Over the last five to six years, studies of personal exposure to traffic air pollution have been part of these investigations.

EXPOSURE MODELING

Dispersion of air pollution from traffic in urban streets is reasonably well described for the classic pollutants like nitrogen oxides (NO_x), carbon monoxide (CO), and benzene (see, e.g., refs. 10, 11). This is reflected in the generally good results obtained from the street pollution models like the CAR model [12], the CPBM [13] and the Danish Operational Street Pollution Model (OSPM) [14].

In a Danish research project [15] the Operational Street Pollution Model (OSPM) was tested for assessment of exposure of bus drivers in Copenhagen, the capital of Denmark. During a three-week campaign nitrogen dioxide was monitored inside and outside a bus on a route in central Copenhagen. Sampling was performed for entire working days. Similar sampling was performed on postmen during the delivery of post, and the postmen were used as a lower exposed reference group. Traffic data for the streets of the routes were obtained from local authorities. Street characteristics (including building height, orientation of the street, street width, etc.) were obtained from maps and on location observations and used together with the traffic data as input for the calculations for 23 selected street sections along the routes and covering the three-week campaign. From diaries of the postmen and time schedule of the bus, an average exposure was determined and compared to the monitoring data. Sampling was performed for the working day of the bus driver and for the mail route for the postmen. The measurements showed that the NO_2 concentrations inside and outside the bus were practically identical. Similarly the NO_2 concentrations on the postman and the bicycle were similar. The OSPM was able to reproduce the measurements well (Fig. 1), and the study therefore showed that this type of model could

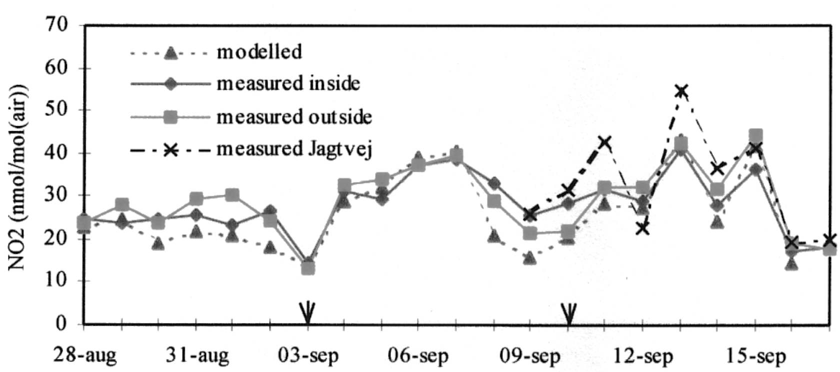

Fig. 1 Average nitrogen dioxide mixing ratios over a whole working day of the Copenhagen bus driver for the campaign period 28 Aug. to 17 Sept. 1995. The figure shows measurements for the entire working day of the bus drivers obtained inside and outside the bus, and results obtained from OSPM calculations representing the average concentrations along the bus route. Average concentrations for the working hours at the monitoring station Jagtvej (one of the streets on the bus route) are shown as well. Arrows on the time axis indicate Sundays [15].

serve as a useful tool for exposure assessment in occupational epidemiological studies of bus drivers in urban areas. The agreement is within the uncertainty of the measurements.

In another project, the aim was to study the possible link between exposure of Danish children to traffic pollution and development of childhood cancer. In this study front door pollution levels at the residence were taken as indicators of exposure of Danish children. The applicability of this indicator and ability of the OSPM to reproduce pollution levels at the address was tested in a pilot study [16]. Personal exposure to nitrogen dioxide and BTX (benzene, toluene, and xylenes) was monitored on 200 Danish children together with pollution at the front door and inside the bedroom. Traffic data and physical characteristics of the area at the address were obtained from registration scheme sent to local authorities. From the registration schemes input data needed for running OSPM was generated.

The OSPM calculations for the 200 addresses were shown to reproduce well the observed concentrations (see Fig. 2). Calculations were subsequently performed for 19 000 addresses in the period 1960 to 1990 to provide input for a case-control study of the possible link between exposure and the development of childhood cancer. The case-control study showed no association between development of the most common childhood cancers and traffic air pollution [17].

Establishing data for the characteristics of the streets (building heights, street width, etc.) have turned out to be a resource-demanding task when calculations for large numbers of locations have to be carried out. This was the background for the development of the so-called AirGIS system [18,19]. AirGIS is a geographic information system (GIS) that applies digital maps and various register data to generate input data for OSPM and subsequently performs calculations with the model. AirGIS has and will be applied for assessment of human exposures within a number of completed and ongoing epidemiological studies.

EXPOSURE MONITORING

In a European Project "Monitoring of Atmospheric Concentrations of Benzene in European towns and Homes" (MACBETH) [20] personal exposure, indoor, and outdoor concentrations were measured using

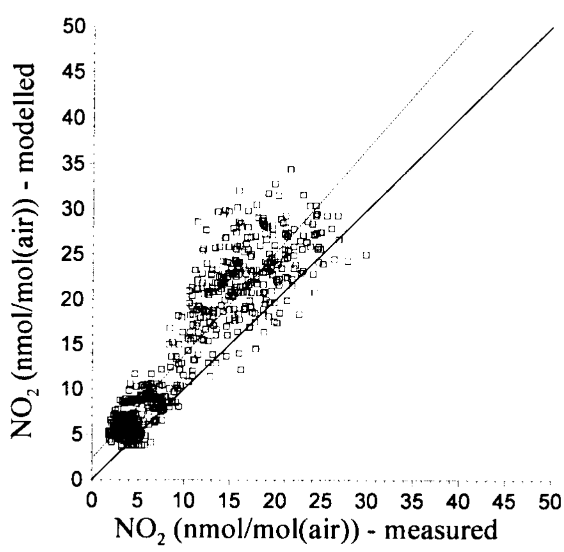

Fig. 2 Comparison of about 1200 observed and calculated monthly mean NO_2 mixing ratios at the address (front door) of 200 Danish children (100 in Copenhagen and 100 in rural areas). Data are identical to the ones presented in Raaschou-Nielsen *et al*. [16] and a similar figure will be presented in Hertel *et al*. [9].

a newly developed high uptake diffusive (Radiello) sampler [21] in six European towns simultaneously. The aim was to investigate a possible link between traffic pollution and human exposure to benzene. The principle of the applied Radiello sampler is following Fick's laws like other diffusive samplers. In general other samplers use axial diffusion from one surface to another, whereas Radiello samplers use radial diffusion over a microporous cylinder into an absorbing inner cylinder, which gives about a 100 times higher uptake rate. In total six five-day campaigns distributed over one year was carried out. Two groups of volunteers were selected—a "low-exposed group" (64% of volunteers), consisting of people with indoor work as, for example, clerks and teachers, and a "high-exposed group" (36% of volunteers) having outdoor work with direct exposure to traffic pollution as, for example, postmen and policemen. In each town personal exposure was measured on 50 volunteers and in their residences as well as in a monitoring net over the town. In the interpretation of the results, factors like occupation of the volunteers and variations in meteorological conditions are taken into account together with information from diaries filled in by the volunteers during the campaign.

The results showed higher exposure in South Europe compared with North Europe. This seems mainly to be due to differences in meteorological conditions, which strongly affects the benzene concentrations. The South European weather is typically controlled by high-pressure systems leading to low wind speeds, whereas in northern Europe the westerly winds dominate [20]. Neither urban background concentrations or concentrations in homes are found to be good measures for personal exposure and thereby as material for policy making.

The Danish part of the study indicates that outdoor levels of benzene (from traffic) strongly contribute to the personal exposure to benzene and furthermore contribute significantly to indoor benzene concentrations [22]. In accordance with this the high-exposed group had a generally higher benzene exposure than the low-exposed group (see Fig. 3).

The annual average benzene concentrations (taken as average of the six campaigns) at most outdoor monitoring sites were below the limit value of 5 µg/m^3 (only six sites had values above). The low-exposed group had a geometrical mean of the annual average just below 5 µg/m^3 whereas the high-exposed group had a geometrical mean of the annual average as high as 7 µg/m^3. In the high-exposed group 75% had an exposure higher than the limit value, whereas this is the case for 45% of the volunteers in the low-exposed group.

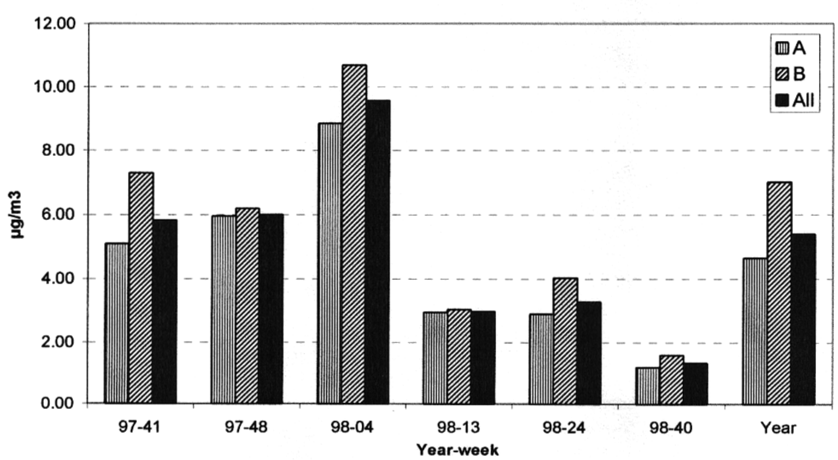

Fig. 3 Geometrical mean of the benzene exposure for the low-exposed group A (64% of volunteers), for the high-exposed group B (36% of volunteers) and for all data. Data for each of the campaigns are shown as well as the average of all campaigns as a proxy for the yearly exposure.

In another just-finished monitoring study, personal exposure to particles ($PM_{2.5}$), and nitrogen dioxide and in one campaign also BTX was monitored during four seasons for 50 students in Copenhagen. For 30 of the students, monitoring was furthermore performed at the front door of their residence and in the bedroom inside the residence. The particles were sampled by using a system from BGI [23], a KTL $PM_{2.5}$ cyclone (developed for the European EXPOLIS study [24]), BGI400 pump (flow 4 L/min and weight 0.6 kg) and battery for 48 h operation (weight 2.1 kg). The equipment for the personal sampling was placed in a backpack that the study subjects carried during the campaign (or placed nearby when they where indoor). The equipment for the indoor sampling was installed in a plastic box, and during sampling it was placed on the floor in an open spot. The same plastic box was used for the equipment for the outdoor sampling, which was undertaken from old bicycles (bought at low cost from police auctions). Students employed in the project handled setting up and taking down the equipment. Sampling is performed on a 37-mm Teflon filter. Before and after the sampling the filters are weighted after conditioning for 24 h in the laboratory. Detection limit is found to be about 18 µg (defined as three times the standard deviation on the blank), which is typically about 5–20% for the performed samples. The nitrogen dioxide measurements were performed on Radiello passive samplers [21] (see earlier description). The sampler uses thiethanolamine as substrate, which absorbs nearly 100% nitrogen dioxide and converts it to nitrite. Afterwards, the nitrite is analyzed on segmented flow analyzer using Saltzman's reagent, followed by spectrophotometric detection at 540 nm. Measurements were performed over a two-day period for each study object and at the end of each two-day campaign, urine (sampled in a plastic container over the last 24 h) and blood samples were collected for analysis of various biomarkers.

The results from the study are still being analyzed, but the first comparisons between indoor and personal monitored $PM_{2.5}$ concentrations seems to show a strong correlation indicating that indoor concentrations play a significant role for the personal exposure; see Fig. 4 for present preliminary results from the first campaign. The correlation between indoor and outdoor $PM_{2.5}$ concentrations seems less pronounced than expected, but more detailed analysis is required before conclusions may be drawn. It should be noted that the Danish population spends more than 90% of the time indoor and that this campaign took place during a period with rather cold weather. Relationships between particle exposure and response in biomarkers are still being analyzed, and the modeling part has just been initialized. This work will be published in international journals.

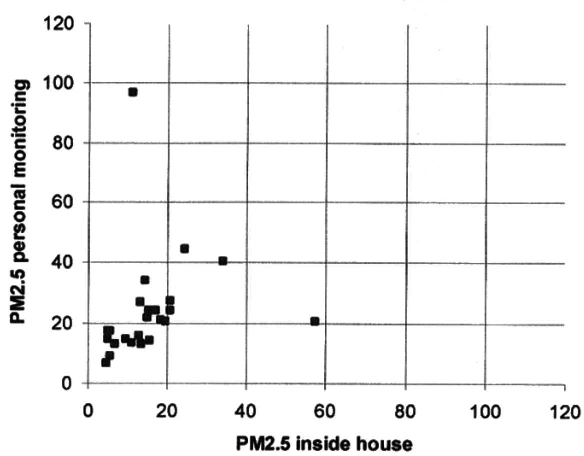

Fig. 4 Comparison of personal exposure and indoor concentrations of $PM_{2.5}$ during the first monitoring campaign in the Danish exposure study.

A global positioning system (GPS) was used together with a diary to trace the route of the study subjects during the campaigns. Until May 2000 the American Army deliberately put a distortion on the satellite signals and therefore the GPS was only accurate within 50 to 100 m. This was insufficient for our purpose, and in the first three campaigns a Spot-FM corrector system developed by the Danish National Survey and Cadastre was applied in order to ensure accuracy within few meters. The Spot-FM was power demanding, which made it necessary to include a battery pack of extra 3 kg. In May 2000 the distortion was removed from the satellite signals and the Spot-FM was therefore not used in the last campaign. Fig. 5 shows an example of data from a GPS device. The results from the GPS will be used together with the personal exposure data in testing and further developing the AirGIS system as a tool for personal exposure assessment.

PARTICLE CHARACTERIZATION

A number of studies have been designed with the special aim of characterizing fine and ultra fine particles from traffic in Danish streets. A differential mobility analyzer (DMA) has been used to monitor size distribution in the ultra fine fraction. Statistical analysis of number concentrations and distributions in traffic density of different vehicle categories has been applied to determine the contribution to the number concentrations from gasoline and diesel powered vehicles [25]. The number concentrations have been shown to correlate well with other traffic-generated pollutants like CO and NO_x. A significant decrease in number concentrations observed in two campaigns in 1999 and 2000 (Fig. 6) is believed to be associated with a reduction of sulfur contents in diesel that took place in July 1999 [26].

Fig. 5 Example of GPS data-logging during an air pollution exposure study in Denmark. Positions are plotted as black dots for every six seconds. Data are obtained from a GPS device in a backpack and an adjustment is performed by use of a corrector that makes use of information from FM radio-masts with known positions (differential GPS). This figure has been presented in Hertel *et al*. [9].

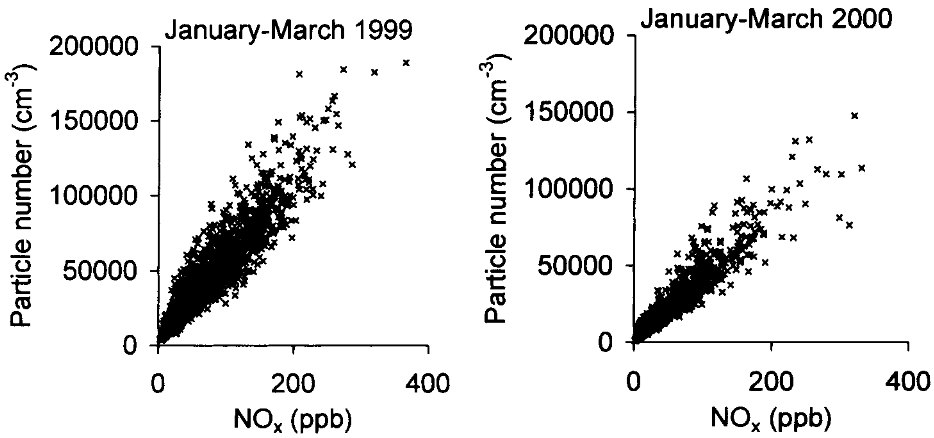

Fig. 6 Half-hour measurements of particle numbers and NO_x in a Copenhagen street canyon during two campaigns in 1999 and 2000.

PERSPECTIVE

In a new series of Danish research projects, studies on personal exposure to particle pollution and the possible link to health effects will be further explored. These projects will take advantage of a number of large ongoing epidemiological studies to address the most pertinent aspects of the health effects related to exposure to traffic-generated air pollution, in particular particles. Modeling (application of the AirGIS system) will be used for exposure assessment, mainly in Copenhagen and surroundings, and a special effort will be made for characterization of particle pollution in Danish urban environments and the important relationship between indoor and outdoor pollution levels.

The health related studies include:

- Annoyance and symptoms in relation to exposure to air pollution from road traffic will be addressed for 20 000 people in the National Health Survey 2000.
- The risk of acute morbidity and mortality of cardiovascular and pulmonary diseases requiring admission to hospital will be addressed in relation to traffic-generated air pollution at the residence, workplace, and commuter route. Nested case-control and time-series designs will be applied in four large population studies in Copenhagen providing a population at risk of approximately 25 000 (the Copenhagen Male Study, Copenhagen City Hearth Study, Diet and Cancer Study and Glostrup Study). Exposure assessment will be particularly detailed by data from an ongoing measurement campaign of individual exposure.
- Intrauterine growth retardation and prenatal/infant mortality will be addressed in relation to exposure to traffic-related air pollution at the residence of 15 000 pregnant women from the Danish National Birth Cohort.
- Exposure to traffic-generated air pollution as a causal factor in asthma development will be addressed by individual measurements of exposure to fine particles, NO_x and carbonyls (acetone, formaldehyde and acetaldehyde) in a study of 400 infants at high risk of asthma development (COPenhagen Prospective Study on Atopy in Childhood)
- The development of self-reported asthma and bronchitis in relation to exposure to traffic-generated air pollution at the small area level will be studied in a health survey conducted on 3000 persons from the Glostrup Study.

A large number of health effects of traffic-generated air pollution are suspected. However, quantitative estimates required for risk assessment and management are extremely difficult due to lack of data and/or difficulties with extrapolating data from other geographical areas and time periods with large differences in composition of air pollution. Moreover, so far very few and mainly small studies have attempted to assess individual exposures and provide necessary confounder control.

Since particle concentrations in urban environments are only scarcely described, a series of projects will look into characterization of particle pollution and establishing relationships between indoor and outdoor pollution levels. The particle characterization projects will include:

- Determination of emission factors for particles from traffic. A number of different monitors will be applied in order to characterize size spectra in vehicle exhausts.
- Contribution from traffic to indoor particle concentrations. Particle and indoor climate measurements will be performed in office rooms close to the street pollution monitoring station in Jagtvej, Copenhagen. A part of the study will be devoted to particle generation from primary pollutants.
- Risk characterization of particles—effect of improved technology. DNA damage on isolated DNA and cell systems, etc.

The results from the particle characterization studies will be used in the further development of the AirGIS system, which in turn will be applied for exposure calculations in the above-mentioned health studies.

The projects will provide data concerning the risk of the most pertinent health outcomes in relation to exposure to traffic-generated air pollution under the present conditions in mainly Copenhagen and surroundings. It should contribute to better decision-making as well as estimation of health effects and associated cost estimation related to traffic policy scenarios.

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REFERENCES

1. E. T. Wilkins. *J. R. Sanitary Inst.* **74**, 1 (1954).
2. J. Schwartz. *Environ. Res.* **94**, 36–52 (1994).
3. A. C. Pope, M. J. Thun, M. M. Namboodiri, D. W. Dockery, J. S. Evans, F. E. Speizer, C. W. Heath. *Am. J. Respir. Crit. Care Med.* **151**, 669–674 (1995).
4. D. W. Dockery and C. A. Pope. *Ann. Review of Public Health* **15**, 107–132. 1994).
5. K. Hemminki and G. Pershagen. *Environ. Health Perspectives* **102** (suppl. 4), 187–192 (1994).
6. E. G. Knox and E. A. Gilman. *J. Epidemiol. Health* **51**, 151–159 (1997).
7. F. Nyberg, P. Gustavsson, L. Järup, T. Bellander, N. Berglind, R. Jakobsson, G. Pershagen. *Epidemiology* **11**(5), 487–495 (2000).
8. N. Künzll, R. Kalser, S. Medina, M. Studnicka, O. Chanel, P. Filliger, M. Henry, F. Horak Jr., V. Poybonnieux-Texier, P. Quenel, J. Schneider, R. R. Seethaler, J.-C. Vergau, H. Sommer. *The Lancet* 795–801 (2000).
9. O. Hertel, F. de Leeuw, O. Raaschou-Nielsen, S. S. Jensen, D. Gee, O. Herbarth, S. Pryor, F. Palmgren, E. Olsen. “Human exposure to outdoor air pollution”, IUPAC Recommendation. Accepted by the IUPAC Commissions for Atmospheric Chemistry and Toxicology. Submitted for publication in *Pure Appl. Chem.* (2001).
10. R. Berkowicz, F. Palmgren, O. Hertel, E. Vignati. “Using measurements of air pollution in streets for evaluation of urban air quality - meteorological analysis and model calculations”, Paper at the 5th International Symposium on Highway and Urban Pollution, WHO/EURO Copenhagen, 22–24 May 1995, *Sci. Total Environ.* **189/190**, 259–265 (1996).

11. R. Berkowicz. Ch. 12 in *Urban Air Pollution: European Aspects*, Fenger, O. Hertel, Jensen (Eds.), pp. 223–251, *Environmental Pollution* Vol. I, Kluwer, Amsterdam (1998).
12. J. den Boeft, H. C. Eerens, W. A. M. den Tonkelaar, P. Y. J. Zandveld. "CAR International: A simple model to determine city street quality", 5^{th} International Symposium on Highway and Urban Pollution, Copenhagen 22–24 May 1995, *Sci. Total Environ*. 189/190, 321–326 (1996).
13. R. J. Yamatino and G. Wiegand. *Atmos. Environ.* **20**, 2137–2156. (1986).
14. R. Berkowicz, O. Hertel, N. N. Sørensen, J. A. Michelsen. "Modelling air pollution from traffic in urban areas". In *Proceedings from IMA meeting on Flow and Dispersion Through Obstacles*, Cambridge, England, 28–30 Mar. 1994, R. J. Perkins and S. E. Belcher (Eds), pp. 121–142 (1997).
15. O. Hertel, P. Wilhardt, R. Berkowicz, H. Skov. *Proceedings of NOSA/NORSAC Symposium 1996*, Poul Hummelshøj (Ed.), Elsingore, Denmark, 15–17 Nov. 1996, Risø-R-934 (EN), pp. 20–22 (1996).
16. O. Raaschou-Nielsen, O. Hertel, E. Vignati, R. Berkowicz, S. S. Jensen, V. B. Larsen, C. Lohse, J. H. Olsen. *J. Exposure Analysis Epidemiol.* **10**, 4–14. (2000).
17. O. Raaschou-Nielsen, O. Hertel, B. L. Thomsen, J. H. Olsen. *Am. J. Epidem.* **155**(3), 433–443 (2001).
18. S. S. Jensen. *J. Hazard. Mater*. **61** (1–3), 385–392 (1998).
19. S. S. Jensen. *A Geographic Approach to Modelling Human Exposure to Traffic Air Pollution Using GIS*, PhD Thesis, National Environmental Research Institute, Roskilde, Denmark (1999), 165pp., accesible as a pdf file at www.dmu.dk.
20. V. Cocheo, P. Sacco, C. Boaretto, E. De Saeger, P. P. Ballesta, H. Skov, E. Goelen, N. Gonzalez, A. B. Caracena. *Nature* **404**, 141–142 (2000).
21. V. Cocheo, C. Boaretto, P. Sacco. *A. Hyg. Ass. J.* **58**, 897–904 (1996).
22. H. Skov, A. B. Hansen, G. Lorenzen, H. V. Andersen, P. Løfstrøm, C. S. Christensen. *Atmos. Environ.* (2001), submitted for publication.
23. L. C. Kenny and R. A. Gussman. *J. Aerosol Sci.* **28**(4), 677–688 (1997).
24. M. Jantunen, O. Hänninen, K. Katsouyanni, H. Knöppel, N. Kuenzli, E. Lebret, M. Maroni, K. Saarela, R. Srám, D. Zmirou. *J. Exposure Analysis and Environmental Epidemiology*, **8**(4), 495–516 (1998).
25. P. Wåhlin, F. Palmgren, R. Van Dingenen. *Atmos. Environ.* (2001), in press.
26. P. Wåhlin, F. Palmgren, R. Van Dingenen, F. Raes. *Atmos. Environ.* (2001), accepted for publication