Separate Appendix Report

For the PhD Thesis A Geographic Approach to Modelling Human Exposure to Traffic Air Pollution using GIS

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Contents

List of Publications, Presentations and Courses

List of Publication:

Jensen, S.S. (1995): Driving Patterns and Emissions from Different Types of Roads. The Science of the Total Environment 169 pp. 123-128.

Fenger, J., Jensen, S.S., Larsen, P.B. (1995): Health Impacts from Traffic Related Air Pollution in Danish Urban Areas, Conference Proceedings No. 2 of "Trafikdage på AUC 1995", pp. 375-384.

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Jensen, S.S.:Driving Patterns and Emissions from Different Types of Roads. Presentation at the 3rd International Symposium "Transport and Air Pollution" in Avignon, France 6-10 June 1994.

Jensen, S.S.: Modelling Population Exposure to Traffic Air Pollution Using GIS. Presentation at "Trafikdage på Aalborg Universitet 19-20 August 1996".

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Poster presentations:

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Jensen, S.S. , *Fenger, J.*, *Larsen, P.B.* (1998): Health Effects of Urban Air Pollution in Denmark. Poster presentation at the Atmospheric Chemistry and Air Pollution 6th FECS Conference on Chemistry and the Environment, 26-28 August 1998, Copenhagen.

Jensen, S.S. (1998): Modelling Human Exposure to Traffic Air Pollution using GIS - Application in Epidemiological Studies. Poster presentation at the SOEH Conference: International Environmental and Occupational Health - Creating Global Linkages. Society for Occupational and Environmental Health (SOEH). Washington DC 18-23 October 1998.

Sabbaticals:

One day stay on the 27th of October 1997 at the RIVM (National Institute of Public Health and the Environment), the Netherlands at the Laboratory of Exposure Assessment.

Teaching experience:

Participation in the organisation of five seminars within "Transport and the Environment" held at University of Roskilde, Department of Environment, Technology and Social Studies during October and November 1995. Gave one lecture on "Environmental Problems in the Transport Sector".

Participation in the organisation of the course and eight lectures within air pollution during a course on "Traffic Noise and Air Pollution" held by the In-Service Courses of the Danish Road Sector (VEJ-EU) during 12-14 May 1998: Introduction to air pollution; Vehicles, EU regulation and the future; Health effects of traffic air pollution with a focus on particles; Air quality standards and national targets Monitoring of air pollution; AirGIS - a future decision-support tool for air quality planning; Emissions from different means of transport; and National programmes within air pollution.

Courses:

Ph.D. course in "Environmental Assessment and Scientific Techniques for Predicting Impacts" at the University of Roskilde, Department of Environment, Technology and Social Studies, 13-17 November 1995.

Course at the Technical University of Denmark (DTU), Department of Planning. "Urban traffic and GIS in Traffic Planning". 24 sessions including assignments during September to December 1995.

Ph.D. course in GIS, Technical University of Denmark (DTU). Six days during November 1995.

Course in "Modelling of Air Pollution Transport and Chemistry", University of Copenhagen. Six sessions including assignments during November and December 1995.

Seminar on Air Pollution Epidemiology organised by the University of Århus, Institute of Environmental and Occupational Medicine. Held at the Danish Cancer Society 28-30 November 1995.

Course in Avenue Programming. Held by the National Environmental Research Institute 6-8 August 1996.

Ph.D. Methodology Course. Held by "Forskeruddannelsesnetværk. Teknik og Samfund. AUC,DTU, HHK, AUC" (Science Education Network of Universities). 14-16 August 1996.

Ph.D. course on "Philosophy of Science and Technology", University of Aalborg (AUC). 30.9 - 2.10 1996.

Participation in more than 20 short seminars held at the National Environmental Research Institute and Risø National Laboratory.

Selected Papers

Paper one

Driving Patterns and Emissions from Different Types of Roads

by

Jensen, S.S.

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Driving patterns and emissions from different types of roads

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Abstract

The project evaluates the relationship between emissions and travel speeds on different types of roads such as city streets, highways, express roads and motorways. Approximately 800 measured driving patterns of 13 streets and roads have been analysed and emissions have been predicted in an emission model, also taking into account deterioration factors and cold start emissions. The result of the analysis is a clear relationship between travel speed (trip length divided by trip time) in the range of 10–120 km/h and emissions from all vehicle types. For petrol-powered passenger cars catalysts reduce HC, CO and NO_x emissions by 70–80% on main roads and by 60–75% on city streets. The difference is due to the proportion of cold engines in city traffic. In city streets, when cars with cold engines are included, the emissions of CO and HC from petrol-powered passenger cars are found to be 10–20% and 5–10% higher, respectively. Travel speed — and not the type of road — is crucial to the level of emissions. However, express roads have slightly higher emission levels than motorways at similar travel speeds, presumably because traffic flows are less steady on express roads than on motorways.

Keywords: Driving patterns; Cold start emissions; Travel speed; Road type; Petrol-powered passenger cars; Catalytic convertors

1. Introduction

The Danish Environmental Protection Agency and the Danish Road Directorate conducted in 1989/1990 a project that evaluated relationships between driving patterns, emissions and traffic management measures in four streets in Copenhagen with heavy traffic. The project showed a strong relationship between travel speed and emissions. The travel speeds in the four streets were between 10–60 km/h [1][1].

This project [2][2] is a follow-up study of the above mentioned. It includes streets in a middle-sized town outside Copenhagen as well as roads

[1] In Danish with an English summary. Air Pollution from Individual and Public Transportation, Danish Environmental Protection Agency, Copenhagen.

[2] In Danish with an English summary, Driving Patterns and Air Pollution — in the Provinces, The Danish Road Directorate, Copenhagen.

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on the main road network. The objective of the study is to examine whether or not the strong relationship between travel speed and emissions is valid for travel speeds >60 km/h and for other types of roads. Another aim of the study is to analyze the relationship between emissions and steady and unsteady traffic flows on main roads. Travel speed is defined as trip length divided by trip time, including stops.

The Danish Road Directorate has financed the project, Spencer C. Sorenson, Laboratory for Energetics, Technical University of Denmark, 2800 Lyngby, Denmark has developed the emission model and COWIconsult has carried out the analysis.

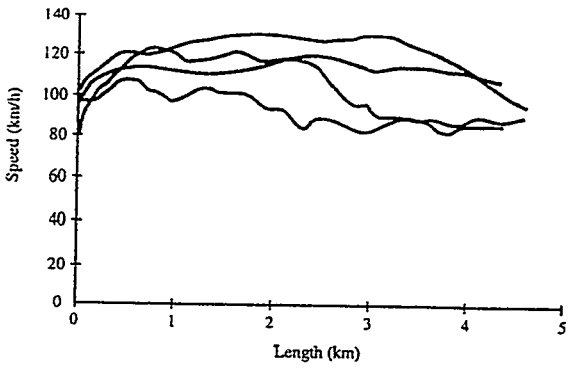

Fig. 1. Examples of driving patterns of four passenger cars on a motorway.

2. Methodology

2.1. Driving pattern

A total of ~800 measured driving patterns of 13 streets and roads have been registered and analyzed. During 1991 driving patterns were registered by a measuring car which followed selected vehicles. The velocity of the selected vehicle was registered every second. Based on this information a driving pattern can be established which describes the speed profile of the vehicle on the section of the road in question (Fig. 1). Six streets in the city of Roskilde outside Copenhagen represent streets with different traffic flows in a middle-sized town (50 000 inhabitants). On the main road network, seven roads have been analyzed, representing steady and unsteady traffic flows on two motorways, two express roads and three highways. At the time of the study the speed limit on city streets was 50 km/h and on highways, express roads and motorways 80 km/h, 90 km/h and 100 km/h, respectively. During 1992 the speed limit on motorways was raised to 110 km/h. The average length of the sections analyzed was 1.5 km for city streets and 6.8 km for main roads.

Furthermore, a survey of the proportion of traffic which have cold engines was carried out for the six city streets assuming no cold engines on main roads.

An existing emission model was extended to cover energy consumption and travel at high speeds. The emission model is developed by the Laboratory for Energetics, Technical University of Denmark [3]. The model predicts emissions and fuel consumption in g/km for HC, CO, NO_x and particles for a given driving pattern for the following vehicles: petrol-powered passenger cars with or without catalyst, diesel-powered passenger cars and diesel-powered heavy duty lorries (10 and 26 tonnes). Emissions and fuel consumption of light duty lorries are estimated from emissions and fuel consumption of passenger cars assuming that light duty lorries have 50% higher emissions than passenger cars due to differences in weight and size.

Emissions are predicted in the following way. The driving pattern is divided into driving conditions which can be characterized as either acceleration, deceleration, constant speed or idle. The driving condition that fits best in an established database of emissions related to different driving conditions is used and emissions can be calculated for the entire driving pattern. For petrol-powered passenger cars the emission model is based on a database established by the Environmental Protection Agency in Studsvik, Sweden (SNV) which contains many instantaneous measurements made during the different portions of various standard emission test cycles which cover speeds up to 100 km/h. The database for Swedish passenger cars has been adjusted to Danish conditions. Details on how the emission database is

established for vehicles other than passenger cars are given in [3].

The emission model predicts emissions and fuel consumption from a new and warm engine. Since emissions change as a function of mileage driven for petrol-powered passenger cars, predicted emissions of HC, CO and NO_x are adjusted according to deterioration factors. Furthermore, as a cold engine has higher emissions than a warm engine for petrol-powered passenger cars, predicted emissions of HC and CO as well as energy consumption are also adjusted according to cold start emissions.

The emission model has been tested using the standard emission test cycles as driving patterns and the variations were within 5–15% which is found to be satisfactory.

2.2. Modelling emissions at high speeds

Detailed emission measurements with acceleration and deceleration are not available for passenger cars at speeds > 100 km/h. Data has been obtained for a few vehicles for constant speed operations up to 120 km/h from the Swedish database. Only integrated emission factors are available for speeds with accelerations up to 140 km/h averaged over a large number of vehicles from Technischer Überwachnungs-Verein Rheinland, Germany (TÜV Rheiland) [4]. This German study shows a significant increase in measured emissions at high speeds as emissions of HC, CO and NO_x increase by factors of 2.3, 5.2 and 2.7, respectively, from a highway cycle with a mean speed of 78 km/h to a motorway cycle of ~ 116 km/h. The measured emissions are for new catalyst cars with warm engines. The levels for HC, CO and NO_x are 0.06 g/km, 0.98 g/km and 0.28 g/km, respectively, for the highway cycle and 0.14 g/km, 5.13 g/km and 0.75 g/km, respectively, for the motorway cycle.

Therefore, to predict emissions at speeds > 100 km/h it has been necessary to extrapolate from present conditions, although this is a very uncertain procedure. Extrapolation has been carried out in a fashion which has at least some basis in technical principles as it is assumed that vehicle power is a correlating variable for higher speeds. Extrapolation has been carried out based on the Swedish database for lower speeds, converting each speed-acceleration condition to the equivalent vehicle power, for details see [3]. The German measured emissions at high speeds were found to be 3–5 times higher when compared with the Swedish extrapolated emissions at high speeds for petrol-powered passenger cars equipped with catalysts. Therefore, it is clear that other factors must be influencing the results.

An explanation could be found in the electronic control system for vehicles with three-way catalyst control. The driving cycles currently in use for emission certification all have maximum speeds < 100 km/h, and weak accelerations with the higher speeds. The manufacturers do not need to pay attention to the emissions at these conditions in order to meet emission certification test procedures. Vehicles using three-way catalysts may change to a rich engine operating mode at the highest speeds to perform accelerations at high speeds. This is especially true for smaller cars with little extra power to give away. In addition to preventing the catalyst from overheating, emissions increase significantly [3].

Therefore, two alternative calculations have been carried out for passenger cars with catalysts at high speeds. 'Cat Low', where the catalyst is assumed to be functioning at all speeds, has extrapolated data for speeds > 100 km/h based on the Swedish database for constant speed operations up to 120 km/h. 'Cat High', where the catalyst is assumed to operate in an unregulated mode at high speeds, is based on the assumption that vehicle power is a correlating variable for higher speeds, extrapolating the Swedish database for lower speeds to high speed conditions and at the same time adjusting the level of modelled emissions to the German measured integrated emissions at high speed. Since the German measured emissions are for new catalyst cars with warm engines, these emissions have further been adjusted to an average Danish car according to deterioration factors.

A small follow-up study was carried out to test the 'Cat Low' and 'Cat High' issue. The exhaust emissions and fuel consumption were measured for three petrol-powered passenger cars with catalysts registered in Denmark according to the

exhaust emission standards of October 1990 when catalysts became a requirement. All cars had an engine size of 1.4 l. The tests were carried out at a test centre in Sweden. In addition to standard regulatory driving cycles the cars were also tested at constant speeds from 85 to 135 km/h and over a special motorway driving cycle. No clear conclusions could be made from the study since the average CO and HC emissions were closer to the 'Cat High' alternative while NO_x emissions were closer to the 'Cat Low' alternative [5].

3. Results

3.1. Emissions and travel speed

The result of the analysis is that there is a clear relationship between emissions and all travel speeds. The strong relationship between emissions and travel speeds for the four streets in Copenhagen is also valid for higher speeds and for city streets in a middle-sized town and for main roads (Fig. 2). However, at high speeds the predicted emissions are uncertain due to few available measured emissions at high speeds.

HC and CO emissions decrease when travel speeds increase for all types of vehicles for travel speeds up to 80–90 km/h. Above 90 km/h emissions increase. For passenger cars the relationship between NO_x emission and travel speeds is parabolic with a minimum at ~60 km/h. For heavy duty lorries a minimum is reached at travel speeds between 60–70 km/h. Energy consumption follows the same trends as NO_x emission. However, at high speeds the increase is weaker and at low speeds it is stronger. Emissions of particles have only been predicted for diesel-powered vehicles because data concerning emissions from petrol-powered vehicles is insufficient to determine emissions of particles as a function of travel speeds. For diesel-powered passenger cars the relationship between emissions of particles and travel speeds is parabolic with a minimum at 70–80 km/h. For heavy duty vehicles the relationship is less clear. However, emissions seem to decrease when travel speeds increase. In general, emissions and fuel consumption are at a minimum at travel speeds of 60–80 km/h.

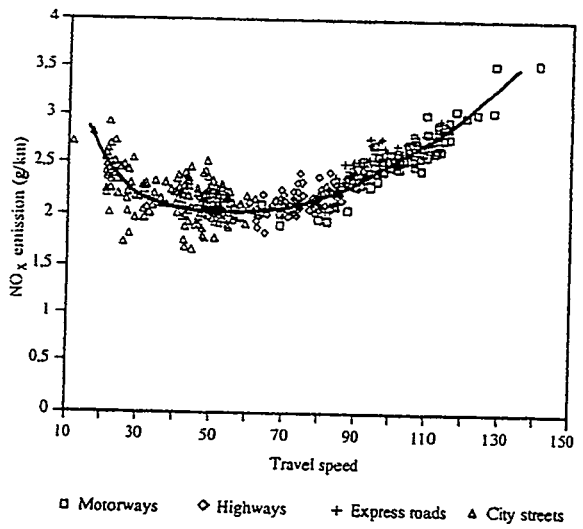

Fig. 2. NO_x emissions from passenger cars without catalysts as a function of travel speed. One dot represents a driving pattern.

Emissions at low travel speeds were found to be relatively high due to the variations in speed at low travel speeds in city streets.

For petrol-powered passenger cars catalysts reduce HC, CO and NO_x emissions by 70–80% on main roads and by 60–75% in city streets, provided the catalyst is functioning at high speeds ('Cat Low'). In the case of 'Cat High' for high speeds, reductions are considerably less on main roads. On main roads catalysts are more effective at reducing emissions of HC, CO and NO_x than in city streets, due to the fact that there are no or few cold engines on main roads (Figs. 3 and 4).

A cold engine has higher emissions of HC and CO as well as higher energy consumption than a warm engine for petrol-powered passenger cars. In city streets, when cold engines are included, the emissions of CO and HC from petrol-powered passenger cars are found to be 10–20% and 5–10% higher, respectively, than when only warm engines are considered, on the condition that 4–8% of all petrol-powered passenger cars have cold engines. Cold engines of petrol-powered passenger cars equipped with catalysts have 20–40% higher emissions of CO and 20–50% higher emissions of HC than warm engines. Cold engines of petrol-powered passenger cars without catalysts have 3–4 times higher emissions of CO and HC than warm engines. For petrol-powered passenger cars with catalysts emissions are 10–20

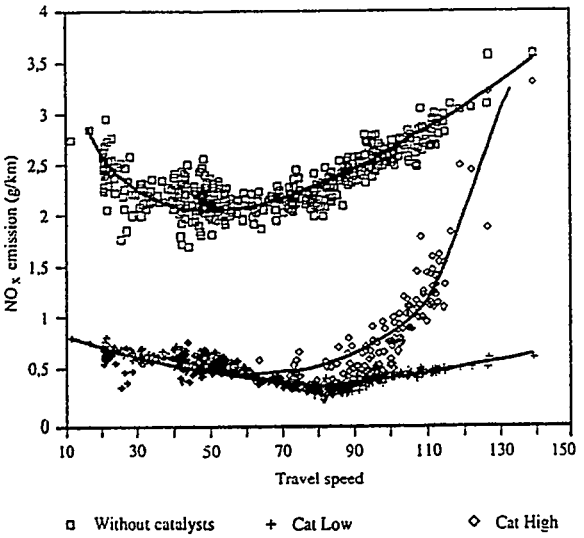

Fig. 3. NO_x emissions from passenger cars with and without catalysts as a function of travel speed.

times more than those of warm engines. A follow-up study has further analyzed the impact on national emissions of cold engines. This study concludes that although driving with cold engines accounts for only 9% of the time, it will generate 60% of HC and CO emissions when all passenger cars have catalysts [6].

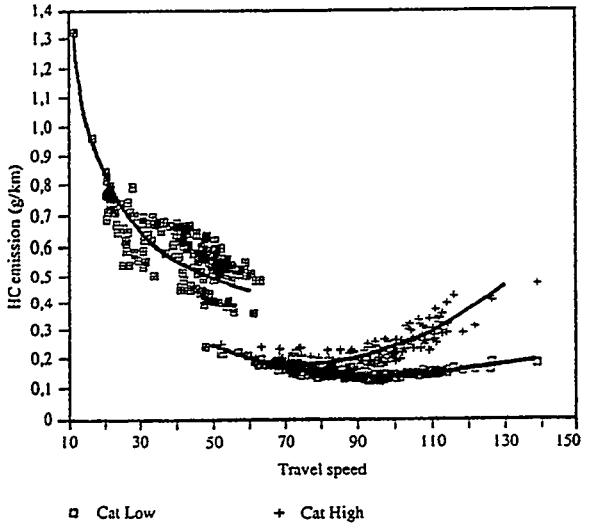

Fig. 4. Illustration of the impact of cold start emission for passenger cars with catalysts. HC emissions are substantially higher for city streets including cold engines than for main roads assuming no cold engines at the same travel speeds (50–60 km/h).

During the project period there was a debate about speed limits in Denmark. A tentative assessment of the effect of higher speed limits was carried out. Based on the relationship between travel speeds and emissions the effects of increased travel speeds from 80 km/h to 100 km/h and from 100 km/h to 110 km/h have been estimated. HC emissions will increase marginally and CO emissions slightly.

Increases of NO_x emissions will by and large follow the increases of petrol-powered passenger cars, that is, ~15% from 80 km/h to 100 km/h and ~10% from 100 km/h to 110 km/h ('Cat Low'). Increases will be higher in the 'Cat High' alternative (Figs. 3 and 4). Increased travel speeds will only increase emissions of particles slightly. Energy consumption will increase along the lines of NO_x emissions from passenger cars, that is, ~14% from 80 km/h to 100 km/h and ~8% from 100 km/h to 110 km/h. Eventually, the speed limit on motorways was raised from 100 km/h to 110 km/h and remained 80 km/h on highways and 90 km/h on express roads.

3.2. *Emissions and different types of roads*

The relationship between emissions, types of roads and traffic flows has been illustrated by the example of NO_x emissions from petrol-powered passenger cars without catalysts. In general, the relationship between emissions and type of road shows that travel speeds and not the type of streets or roads are crucial to the level of emissions. However, NO_x emissions on express roads showed 6% higher emissions than on motorways for a travel speed of 80 km/h dropping to 0% for a travel speed of 140 km/h, presumably because traffic flows are less steady on express roads than on motorways (Fig. 5). Based on qualitative assumptions main roads with steady and unsteady traffic flows on motorways, express roads and highways were selected. The analysis of the relationship between emissions and travel speeds showed no significant difference between steady and unsteady traffic flows for the same travel speeds, indicating that driving patterns are much alike for the same travel speed regardless of the type of road. A follow-up study has further analyzed the relationship between speed fluctuations and travel speeds. This study concludes that travel

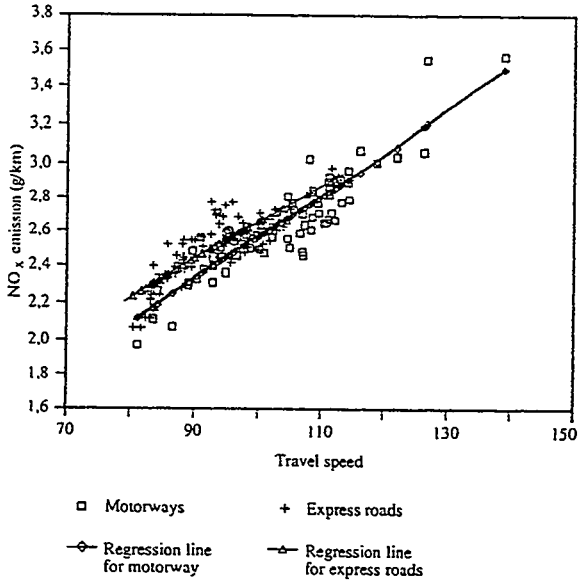

Fig. 5. NO_x emissions from passenger cars without catalysts as a function of travel speed on motorways and express roads. Regression lines for motorways and express roads are shown.

speed is the most important factor with speed fluctuation being of a lesser importance in relation to emissions [7].

3.3. Total emission loads

For the selected motorways, express roads, highways and city streets the total emission loads have been calculated based on average emission factors of the different types of vehicles and their numbers. HC and CO emissions per km travelled are higher for city streets than for main roads, while NO_x emissions and energy consumption per km travelled are higher on main roads compared with city streets. The distribution of emission load by type of vehicle has been calculated. Approximately 85–90% of HC and CO emissions come from passenger cars, whereas 50% of NO_x emissions originate from heavy duty traffic. On main roads, 50% of emissions of particles are derived from heavy duty traffic compared with 60% in city streets. In city streets light duty traffic causes ~ 25% of the total load of emissions of particles. Estimates are based on the 'Cat Low' alternative and on the assumption that 9% of the traffic flow in 1991 was petrol-powered passenger cars equipped with catalysts.

4. Discussion

The study revealed the need for further measurements of emissions at high speeds in order to predict reliable emissions based on driving patterns at high speeds. Furthermore, the impact of cold engines on emissions is significant for city streets and especially with a growing proportion of cars equipped with catalysts. Measures to reduce the impact of cold engines are essential.

The relationship between travel speeds, and emissions and energy consumption for all types of vehicles has gained widespread application in Denmark. Municipalities carry out mapping of the environmental impact of traffic in the preparation of local action plans based on these relations using spread sheet models or traffic models with environmental impact mapping facilities. Furthermore, the relationship between travel speeds, and emissions and energy consumption is used to carry out environmental impact assessments (EIA) of, for example, motorways, etc.

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Paper two

Sundhedsmæssig vurdering af luftforurening fra vejtrafik med særlig fokus på partikler

by

Jensen, S.S., Larsen, P.B., Fenger, J.

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Sundhedsmæssig vurdering af luftforurening fra vejtrafik
- med særlig fokus på partikler

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Abstract
En lang række undersøgelser peger på en sammenhæng mellem luftforurening og sundhedsbelastning i byområder. Disse forhold er nærmere diskuteret i rapporten "Sundhedsmæssig vurdering af luftforurening fra vejtrafik", der i foråret 1997 er udgivet som nr. 352 i Miljøstyrelsens serie af miljøprojekter. Blandt de mange forskellige stoffer, som udsendes fra vejtrafik udpeger rapporten de kritiske luftforureningskomponenter. Partikler og i særdeleshed små partikler vurderes som den mest sundhedsbelastende luftforureningskomponent. Derfor fokusseres i det følgende mere detaljeret på kilderne til partikelforurening, forureningens egenskaber og niveauer samt påvirkning af mennesker.

Indledning

Trafik dominerende forureningskilde
Luftforureningen i storbyer i Danmark og andre industrialiserede lande har i de senere år skiftet karakter. Tidligere var det alvorligste problem svovl- og sodforureningen fra mindre fyringsanlæg, der i ekstreme tilfælde gav anledning til den berygtede "London smog" (efter engelsk: smoke and fog). Dette problem er nu i det store og hele løst ved anvendelse af renere brændsler, forbedret fyringsteknik og indførelse af fjernvarme produceret på store anlæg med røggasrensning og høje skorstene. I dag er luftforureningen i gadeniveau - og dermed den potentielle sundhedsbelastning - domineret af udstødningen fra biltrafik, og selv om mange af stofferne går igen, er sammensætningen anderledes.

Udsættelse for luftforurening
En række forhold har indflydelse på udviklingen af sundhedsskadelige effekter i befolkningen. Jo højere niveauer en person eksponeres for og jo længere det varer jo større er risikoen for sundhedsskadelige effekter. Koncentrationen af luftforurenende stoffer vil, alt andet lige, stige med bystørrelsen og trafikintensiteten, falde fra bycenteret mod forstæderne og yderligere ud til landområderne. En persons tids- og aktivitetsmønsteret dvs. hvor lang tid en person opholder sig i forskellige mikromiljøer med forskellige forureningsniveauer (fx indendørs hjemme, udendørs hjemme, på arbejde eller i trafikerede gader) er derfor afgørende for personens samlede eksponering, og den kan være meget forskelligt fra person til person. Den dosis, dvs. den mængde som optages, er afgørende for de sundhedsskadelige effekter og afhænger af vedkommendes fysiologi (fx udtrykt ved alder) og fysiske aktivitetsniveau (fx udtrykt ved mængde inhaleret luft). Hertil kommer en stor variation i følsomheden overfor luftforurening, således at ikke alle befolkningsgrupper påvirkes i samme grad.

Luftforureningskoncentrationen i udemiljøet er afgørende for en persons samlede eksponering, fordi den påvirker såvel udendørs- som indendørsniveauet. Ophold indendørs i bygninger yder dog en vis beskyttelse mod indtrængende luftforurening. I nogle tilfælde kan indendørskilder dog bidrage med de samme stoffer som trafik. Hvis en person derfor er udsat for indendørs udslip fx fra passiv rygning, fra et gaskomfur eller fra frigivelse af forurenende stoffer fra byggematerialer, kan dette væsentligt påvirke den samlede eksponering, idet en dansker i gennemsnit opholder sig indendørs ca. 22 timer i døgnet.

Udsatte og følsomme befolkningsgrupper
Personer som har bopæl eller arbejdssted langs stærkt trafikerede gader vil være særligt udsatte. Det gælder specielt i lukkede gaderum, hvor luftforureningen spredes dårligt. Her vil pendlere med lang transporttid i bil eller bus være særligt udsatte. Det samme gør sig gældende for gående og cyklister. I forbindelse med særlige aktiviteter udsættes befolkningen for en forhøjet eksponering, det gælder fx ophold i lukkede parkeringshuse og -kældre, ved tankpåfyldning på benzinstation mv.

Erhvervsgrupper som udsættes for høj eksponering i udemiljøet er især de mennesker, som arbejder udendørs i trafikerede bymiljøer, og som samtidig er fysisk aktive fx postbude, cykelbude i større byer, arbejdere som udfører vedligeholdelsesarbejder i forbindelse med vejbelægninger og infrastruktur placeret i eller ved gader fx belysning, el, gas, vand, telefon mv. Chauffører og andre erhvervsgrupper som kører meget vil ligeledes være udsatte, især bus- og taxachauffører, som kører i trafikerede gader i de større byer.

Således skønner man, at luftforureningen forværrer tilstanden hos personer med luftvejslidelser og hjerte-karsygdomme samt øger forekomsten af visse kræftformer fx lungekræft. Der er således specielt følsomme grupper, som er i risiko for at blive berørt med øget sygelighed eller dødelighed til følge, hvor selv forholdsvis moderate luftforureningsniveauer kan påvirke svækkede og syge personer, personer med luftvejslidelser, samt evt. børn og ældre.

Sundhedsskadelige effekter

Undersøgelsesmetoder
Vurdering af de skadelige effekter af luftforurening sker på baggrund af en samlet vurdering af befolkningsundersøgelser, kontrollede laboratorieforsøg og dyreforsøg. Ved befolkningsundersøgelser (epidemiologiske undersøgelser) vurderes i princippet effekten af den samlede forurening, hvor sammenhængen mellem luftforurening og effekt typisk vurderes med effektmålene: dødelighed, sygelighed, antal lægebesøg/indlæggelser, lungefunktionsmål, forekomst af generende symptomer o. lign. I kontrollerede laboratorieforsøg undersøges effekten hos frivillige forsøgspersoner, som udsættes for bestemte koncentrationer af en eller flere luftforureningskomponenter i en veldefineret kortere varighed. I laboratorieforsøg med dyr udføres typisk et større antal forsøg, og der kan anvendes højere koncentrationer og mere tilbundsgående undersøgelser af væv og organer, end der er muligt med forsøgspersoner. Disse undersøgelser bruges til at bestemme kritiske stoffer og vurdere sammenhængen mellem eksponering og effekt (dosis-respons sammenhænge).

Kritiske stoffer, grænseværdier og effekter
De sundhedsmæssigt mest kritiske luftforureningskomponenter fra vejtrafik vurderet ud fra eksisterende niveauer i Danmark er vist i nedenstående tabel med angivelse af niveauer, grænseværdier og deres sundhedsskadelige effekt.

Tabel 1 Kritiske luftforureningskomponenter fra vejtrafik og deres sundhedsskadelige effekter[f]

Kritiske luftforurenende stoffer fra trafik	Byniveau, DK[a] $\mu g/m^3$	Bindende og vejledende grænseværdier, $\mu g/m^3$	Sundhedsskadelig effekt		
			Irritation	Kræft	Allergi
Partikler	døgnværdi: 109-139 $\mu g/m^3$ TSP, 95-percentil (sv.t. 60-76 $\mu g/m^3$ PM_{10}, 95-percentil)	døgnværdi: 300 $\mu g/m^3$ TSP, 95-percentil[b] (sv.t. 165 $\mu g/m^3$ PM_{10}, 95-percentil)	[e]	(x)[e]	(x)[e]
	årsværdi: 51-70 $\mu g/m^3$ TSP, middelværdi (sv.t. 28-39 $\mu g/m^3$ PM_{10}, middelværdi)	årsværdi: 150 $\mu g/m^3$ TSP, middelværdi[b] (sv.t. 83 $\mu g/m^3$ PM_{10}, middelværdi)			
NO_2	timeværdi: 100 $\mu g/m^3$ (98-percentil)	timeværdi: 200 $\mu g/m^3$ (98-percentil)[b]	x		
	årsværdi: 40-50 $\mu g/m^3$ (middelværdi)	årsværdi: 40-50 $\mu g/m^3$ (middelværdi)[c]			
Ozon	timeværdi: 100 $\mu g/m^3$ (98-percentil) 50 $\mu g/m^3$ (middelværdi)	8-timers værdi: 110 $\mu g/m^3$ (middelværdi)[b] 120 $\mu g/m^3$ (middelværdi)[c]	x		(x)
Hydrocarboner:					
- PAH	årsværdi: 1-4 ng/m^3 BaP (middelværdi)	årsværdi: 0,01 ng/m^3 BaP (middelværdi)[d]		x	
- Benzen	5-20 $\mu g/m^3$ (middelværdi)	årsværdi: 0,13 $\mu g/m^3$ (middelværdi)[d]		x	
- 1,3-butadien	ca. 1 $\mu g/m^3$ (middelværdi)	årsværdi: 0,01 $\mu g/m^3$ (middelværdi)[d]		x	
- formaldehyd		30-minuttersværdi: 10 $\mu g/m^3$ (98-percentil)	x	x	
	ca. 4 $\mu g/m^3$ (middelværdi, vinter)	årsværdi: 1 $\mu g/m^3$ (middelværdi)[d]			
- acrolein		30-minuttersværdi: 10 $\mu g/m^3$ (98-percentil)	x		
	ca. 1-2 $\mu g/m^3$ (middelværdi)	årsværdi: 1 $\mu g/m^3$ (middelværdi)[d]			

a: målt eller skønnet niveau på målestationer i større danske byer; disse stationer er normalt placeret i relativt stærkt trafikerede områder og repræsenterer derfor ikke gennemsnitlige forureningsniveauer
b: gældende dansk tærskelværdi for sundhed se EØF 1992 (bindende grænseværdi)
c: WHO-værdi (vejledende værdi)
d: kvalitetskriterie = 10^{-6} livstidsrisikoniveau for udvikling af cancer efter Miljøstyrelsens retningslinier
e: se iøvrigt efterfølgende beskrivelse i teksten, idet de nævnte effektområder ikke alene beskriver effekten for partikler
f: tabellen er baseret på Larsen et al. 1997
(x): indikerer en medvirkende faktor

Luftforureningskomponenter som CO, SO_2 og bly, som tidligere har været udpeget som kritiske, er gennem en årrække reduceret så meget, at de ikke forekommer i sundhedsmæssige kritiske niveauer. CO vil yderligere blive reduceret fremover som følge af udbredelsen af katalysatorer. SO_2, som primært stammer fra kul- og olieafbrænding, er blevet reduceret væsentligt gennem røgrensning og

brændsler med lavere svovlindhold. Bly tilsættes ikke længere i benzin, heller ikke i "blyholdig benzin".

Der er kun bindende grænseværdier for NO_2, ozon og TSP (Total Suspended Particles). Selvom de bindende grænseværdier ikke er overskredet vurderes luftforureningen at have væsentlige sundhedsmæssige konsekvenser for befolkningen. EU og WHO arbejder med revision af de bindende og vejledende grænseværdier, som vil blive skærpet i de kommende år.

Luftvejsirritanter, kræftfremkaldende stoffer og allergi
Stofferne NO_2, ozon, formaldehyd og acrolein virker luftvejsirriterende, og er især et problem for personer som lider af luftvejssygdomme som astma og kronisk bronkitis, og kan være en medvirkende årsag til udvikling af luftvejssygdomme. Stoffer som PAH (Polycykliske Aromatiske Hydrocarboner), benzen, 1,3-butanien, formaldehyd og acrolein vurderes at være kræftfremkaldende. Partikler er en medvirkende faktor, som bærer af kræftfremkaldende stoffer. Risikoen for at udvikle kræft vil være forøget ved stigende eksponering for alle befolkningsgrupper, men personer som i forvejen har svækket immunforsvar har større risiko. Luftforureningen kan forværre tilstanden hos personer, som lider af luftvejsallergiske sygdomme, og undersøgelser antyder, at visse luftforurenende stoffer som partikler og ozon kan forstærke udviklingen af allergi, men resultaterne er ikke entydige.

Kombinationseffekter
I udeluften optræder stofferne ikke hver for sig men altid sammen, hvorfor kombinationseffekter i visse tilfælde kan forstærke effekten. Sundhedseffekter er derfor i høj grad knyttet til den samlede påvirkning fra luftforureningen snarere end til det enkelte stof. For luftvejsirritanter som fx NO_2 og ozon regner man med en additiv effekt. For de kræftfremkaldende stoffer har man på nuværende tidspunkt langtfra identificeret alle kræftfremkaldende enkeltkomponenter i trafikudstødningen, og kombinationseffekter mellem de kræftfremkaldende stoffer og øvrige komponenter er i vid udstrækning uafklaret. Foreliggende data tyder dog på, at den kræftfremkaldende effekt af den samlede udstødning/luftforurening overstiger summen af de enkelte kræftfremkaldende stoffer dvs. en synergistisk effekt.

Opgørelse af de sundhedsskadelige effekter
De irritative stoffer vil være til størst gene for personer, som lider af luftvejslidelser. Omkring 6 % eller ca. 233.000 personer blandt den voksne del af befolkningen har langvarige luftvejslidelser, som er meget hæmmende i dagligdagen fx astma og kronisk bronchitis (DIKE 1995).

For kræftfremkaldende stoffer anser Miljøstyrelsen et eksponeringsniveau, der svarer til en 10^{-6} livstidsrisiko for tolerabel (det niveau der over en 70-årig periode teoretisk set vil medføre et ekstra kræfttilfælde blandt en million personer). Ud fra sådanne livstidsrisikoberegninger og de målte niveauer af PAH og benzen og skønnede niveauer af 1,3-butadien i København kan det årlige ekstra antal kræfttilfælde pr. en million byboere skønnes til 3-6 for disse stoffer. Dette skøn undervurderer dog den samlede effekt pga. kombinationseffekter blandt andet med partikler.

Såfremt man kan overføre udenlandske resultater til danske forhold vil en reduktion af det aktuelle partikelniveau af PM_{10} på ca. 30% svare til et fald i den årlige dødelighed på omkring 500 personer pr. 1 millioner mennesker i de større byområder. Til sammenligning dør der årligt omkring 550 personer ved trafikulykker i Danmark. Hertil kommer, at et betydeligt større antal personer må forventes at blive påvirket af partikelforureningen med en forringelse af sundhedstilstanden og øget sygelighed til følge. De mest følsomme personer vil være at finde blandt personer med luftvejslidelser og hjerte-karsygdomme. Forekomsten af hjerte-karsygdomme i den voksne befolkning er ca. 6 % eller 259.000 (DIKE 1995).

Partikler

Som det fremgår af ovenstående er partikler den mest kritiske luftforureningskomponent mht. dødelighed og sygelighed. Problemstillingen er ikke helt enkel, fordi partikler - i modsætning til mange andre luftforureninger ikke er en veldefineret kemisk forbindelse. Alt efter kilderne og de atmosfæriske forhold varierer de både i fysisk form og kemisk sammensætning. Det giver vanskeligheder, såvel når man skal kortlægge forureningen, som når man skal vurdere effekterne.

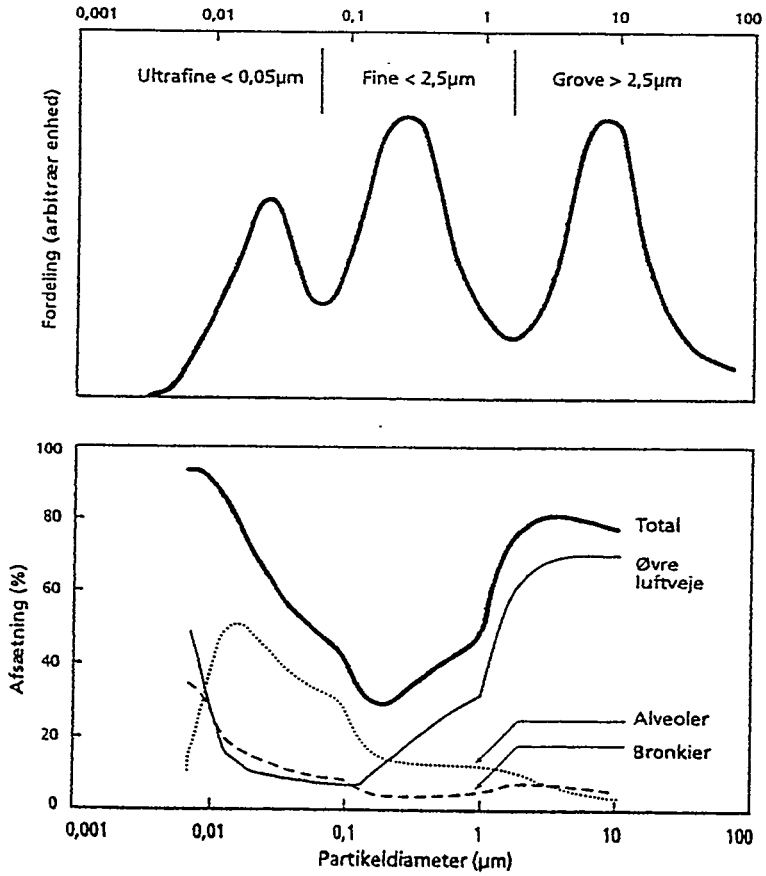
Hvor kommer partiklerne fra ?
Partikler i byluften kan opdeles i tre, næsten adskilte størrelsesfraktioner (figur, øverst). De fleste af de mindste partikler har en diameter på under 0,2 µm. Disse partikler er blevet dannet i forbindelse med forbrændingsprocesser eller ved efterfølgende kondensation af forbrændingsgasser. De har en relativ kort levetid, fordi de har tendens til at hæfte sig til hinanden; herved dannes den næste partikelfraktion med en diameter på mellem 0,2 og 2 µm. I trafikerede byer dominerer partikler fra disse to partikelfraktioner. Kemisk består de overvejende af nitrat, sulfat ammonium og kulstof. Desuden indeholder disse mindre partikler hovedparten af luftens indhold af PAH og tungmetaller. Den tredje fraktion består af partikler med en diameter over 2 µm; de skyldes støvformige emissioner fra virksomheder, friktionsprocesser (fx dækslid) eller ophvirvlet støv. Denne noget grovere fraktion har som regel et højt indhold af jordmineraler.

Monitering af partikler
Gennem mange år har man registreret luftens indhold af partikler med den såkaldte OECD metode, der er baseret på måling af sværtning af filtre, hvor igennem der er suget en given mængde luft. Resultatet angives som "sodindhold", på engelsk forkortet BS (black soot).

Senere er man gået over til også at veje filtrene før og efter eksponering og har herved bestemt luftens totale indhold af partikulært stof, på engelsk forkortet TSP. Først fra omkring 1990 er man enkelte steder i Europa begyndt at måle størrelsesfraktioner af partiklerne ved at bestemme luftens indhold af såkaldt PM_{10}, der er vægten af partikler (particulate matter) med en diameter under 10 µm. Imidlertid indeholder PM_{10} naturligt dannede partikler, der generelt er større end de menneskeskabte, og det vil derfor på længere sigt være vigtigt også at bestemme $PM_{2,5}$ (partikler med en diameter under 2,5 µm) samt endnu finere partikelfraktioner.

Anvendelsen af forskellige målemetoder vanskeliggør en vurdering af udviklingen i forureningsniveauer. Resultaterne kan til en vis grad omregnes, men der er for forskellige områder ikke samme sammenhæng mellem BS og TSP, fordi partiklerne kan have forskellig farve. Cementstøv, der er lyst, registreres således ikke særligt effektivt ved sværtning.

Også størrelsesfordelingen kan have betydning. Målinger fra Erfurt, en by i det tidligere DDR, viser således, at partikelindholdet (her målt som $PM_{2,5}$) er faldet markant efter genforeningen, hvor der blev indført opvarmning med gas i stedet for brunkul og udskiftning af østtyske biler med nyere vesteuropæiske modeller. Alligevel er indholdet af endnu mindre partikler (0,01-0,1 µm) mere end fordoblet, og antallet af partikler er forøget.



Øverst:
Størrelsesfordeling af luftbårne partikler i byluft.
Nederst:
Partikelafsætningen i forskellige regioner af luftvejene hos en voksen mand. Sandsynligheden for afsætning afhænger af partiklernes størrelse og er generelt størst for små partikler. En høj totalafsætning for store partikler skyldes, at disse afsættes i de øvre åndedrætsorganer, hvor de ikke har større helbredsmæssige virkninger (figuren er baseret på HMSO 1995).

Forureningsniveauer

I Europa varierer de årlige middelværdier af PM_{10} fra 10 µg/m^3 i renluftområder til over 100 µg/m^3 i stærkt industrialiserede områder. De seneste tre år har der ikke været nogen signifikant udvikling. Emissionsopgørelser for Storbritannien, Tyskland og Holland antyder dog, at der er sket et fald i udslip.

I danske byer er partikler blevet moniteret under LMP (Det landsdækkende luftkvalitetsmåleprogram), der blev oprettet i 1982 og i forskelligt omfang har været fortsat siden da (Kemp, Palmgren 1995). I Hovedstadsregionen driver de fem amtskommunale enheder i fællesskab et overvågningsprogram (HLU), der er en videreførelse af aktiviteter under det nu nedlagte Hovedstadsråd. Samlet foreligger der målinger tilbage fra slutningen af 1960'erne.

I Københavns centrum faldt den årlige middelkoncentration af sod fra næsten 100 µg/m^3 i 1968 til under 20 µg/m^3 i slutningen af halvfjerdserne. Derefter steg den i nogle år - muligvis pga. forøget kulfyring og vækst i antal af dieselbiler, men den er nu igen under 20 µg/m^3 (HLU 1996).

TSP har været målt siden 1988 i København, Odense og Aalborg og niveauerne er siden da faldet ca. 25%. I Københavns centrum ligger årsmiddelværdien nu på ca. 60 µg/m^3 med ca. 140 µg/m^3 som 95-percentil.

Foreløbige målinger af PM_{10} i København, antyder et bybaggrundsniveau på 20-25 µg/m^3, mens niveauer på 30-35 µg/m^3 er observeret langs en stærkt trafikeret vej (Frøsig & Moseholm 1996). Indikative målinger af $PM_{2.5}$ i København viser gennemsnitsniveauer på omkring 19 µg/m^3 langs en trafikeret gade (Jagtvej) målt over 5 måneder og et bybaggrundsniveau omkring 13 µg/m^3 målt over 1 måned (Frøsig et al. 1997). Miljøkontrollen i København har ligeledes i en stærkt trafikeret gade (H.C. Andersens Boulevard) målt $PM_{2.5}$ til 9 µg/m^3 i gennemsnit over en et årig periode (Miljøkontrollen, 1997). Forskelle i niveauer på Jagtvej og H.C. Andersens Boulevard kan være begrundet i forskelle mellem målemetoder, tidsperioder og lokaliteter. Forholdet mellem TSP, PM_{10} og $PM_{2.5}$ for en stærk trafikeret vej i København (Jagtvej) er 1.00/0.52/0.30, hvilket er i god overensstemmelse med observationer fra andre lande, idet grove omregningsfaktorer er 1.00/0.55/0.33.

Befolkningens eksponering
Ved indånding af partikler (figur, nederst) opfanges de grovere partikler (diameter over 10 µm) i de øvre luftveje i næse og svælg. Partikler under 10 µm afsættes også længere nede i luftvejene i bronchier og bronchioler, mens partikler under 5 µm vil være i stand til at kunne trænge helt ud i lungernes yderste forgreninger, alveolerne. Jo længere nede i lungerne partiklerne afsættes, jo længere tid går der, før de elimineres af lungernes forsvarsmekanismer. I alveolerne kan eliminationstiden være måneder til år. Det betyder i sig selv, at små partikler har større sundhedsskadelig effekt. Hertil kommer, at de finere partikler modsat de grovere er i stand til at trænge ind i bygninger og opretholde stort set de samme niveauer som i udemiljøet, hvorved man bliver udsat for dem både ude og inde. Samtidige målinger af $PM_{2.5}$ udendørs og indendørs i København har vist at indendørskoncentrationerne var 85 % af udendørskoncentrationerne (Frøsig & Sherson, 1997).

Folk udsættes fortrinsvis for partikelforurening, og i særdeleshed fra trafik, hvis de bor i de større eller mellemstore byområder. I Danmark gælder det 1,8-2,9 millioner personer. Andre betydende faktorer for personers partikeleksponering er den tid, de opholder sig i gademiljøer med direkte eksponering fra trafikken, samt hvor fysisk aktive de er. Den mængde luft som indåndes øges således betydeligt ved løb og cykling. Yderligere synes den generelle sundhedstilstand at spille en rolle. En forholdsvis ny amerikansk laboratorieundersøgelse har således vist, at personer med kroniske luftvejslidelser pga. ændret vejrtrækningsmønster og luftgennemstrømning i luftvejene ved realistiske partikelniveauer tilbageholdt op til fem gange så mange partikler som raske (Bennett et al. 1997).

Helbredsmæssige virkninger

Kortvarig eksponering
Der er udført talrige befolkningsundersøgelser til vurdering af partikelforureningens sundhedsskadelige effekter. I de fleste undersøgelser har man beskæftiget sig med de akutte effekter, der er optrådt i forbindelse med perioder med forhøjede partikelniveauer. Man har her registreret den umiddelbare påvirkning mht. en eller flere effektparametre, fx: dødeligheden i befolkningen, antal hospital-sindlæggelser forårsaget af luftvejslidelser, forekomsten af luftvejssymptomer og anvendelsen af

medicin i forbindelse hermed, lungefunktionen (bestemt ved lungefunktionsmålinger), samt fravær fra skole eller arbejdsplads.

En række udenlandske undersøgelser fremkommet inden for de seneste fem år har meget enstemmende fundet en øget dødelighed i tilknytning til dage med forhøjet luftforurening. Undersøgelserne angiver en øget dødelighed på mellem 0,8 og 1,6% for hver gang PM_{10}-partikelindholdet steg 10 $\mu g/m^3$. Især steg dødeligheden som følge af hjerte-karsygdomme og luftvejslidelser. Over for andre forureningskomponenter fandt man en meget svagere sammenhæng mellem niveauer og effekter.

For sygelighed er der ligeledes fundet markante sammenhænge med partikelforureningen. Ved en sammenfattende gennemgang af de seneste af denne type undersøgelser fandt man, at en stigning i det daglige partikelniveau på 10 $\mu g/m^3$ (PM_{10}) er forbundet med 0,5-3,4% stigning i antallet af hospitalsindlæggelser eller skadestuebesøg, en stigning på 1-3% i forekomsten af astmaanfald og luftvejssymptomer, og en reduktion af lungefunktionen på omkring 0,5%.

Langvarig eksponering
Andre undersøgelser har vurderet partikelforureningens kroniske effekter, idet man her har sammenlignet dødelighed, forekomsten af luftvejssygdomme eller lungefunktionen i områder med forskellige niveauer af partikelforurening (målt som gennemsnitsniveauer over lang tid).

Hvad angår dødeligheden skal specielt nævnes to større undersøgelser med henholdsvis 8.000 og 552.000 mennesker fra henholdsvis 6 og 151 byområder i USA. I undersøgelserne fandt man, at der forelå en statistisk sikker sammenhæng mellem dødelighed og luftens indhold af partikler. Det gennemsnitlige partikelniveau for alle byer lå i intervallet 9 til 33,5 $\mu g/m^3$ (målt som $PM_{2,5}$). I den første undersøgelse steg dødeligheden 14% ved en øgning af $PM_{2,5}$ partikelniveauet på 10 $\mu g/m^3$ (svarende til 8 % for PM_{10}) og i den anden var stigningen på 7% (svarende til 4 % for PM_{10}). I undersøgelserne var der indhentet oplysninger om en række samvirkende faktorer såsom alder, race, rygevaner, legemsvægt, alkoholforbrug, erhverv m.m., således at betydningen heraf kunne modregnes. Det skal nævnes at tilsvarende klare sammenhæng ikke kunne konstateres for de andre luftforureningskomponenter.

Sammenholdes dette med andre, tidligere undersøgelser er der således en velunderbygget sammenhæng med en øget dødelighed på ca. 1-8% for en 10 $\mu g/m^3$ stigning i det gennemsnitlige partikelindhold bestemt som PM_{10} med et gennemsnit på omkring 4%.

Tilsvarende er der udført undersøgelser, der sammenligner sygelighed i områder med forskellige niveauer af luftforurening. En analyse af de seneste af disse undersøgelser angiver, at en stigning af det gennemsnitlige partikelindhold på 10 $\mu g/m^3$ (PM_{10}) er relateret til 2% stigning i hospitalsindlæggelser og skadestuebesøg pga. af luftvejslidelser, en 2% nedgang i lungefunktionen og en øget forekomst på 10-25% i befolkningen af bronkitis og kronisk hoste.

Vurdering af effekterne
Samlet må partikelforureningen vurderes at medføre betydelige sundhedsskadelige effekter. Især personer, der lider af luftvejs- og hjertekarsygdomme anses for følsomme og i risiko for at få deres tilstand forværret. Den øgede dødelighed, der kan relateres til partikelforureningen, må således tages som udtryk for, at personer, der påvirkes, dør tidligere end de ellers ville have gjort (dvs. en negativ påvirkning af den gennemsnitlige levealder).

Ved en vurdering af disse data har man for USA beregnet at partikelforureningen er relateret til 60.000 dødsfald pr. år, mens tilsvarende tal for England og Wales er anslået til 10.000 dødsfald. En hollandsk vurdering peger på et fald i den gennemsnitlige levealder på 1,1 år pr. 10 $\mu g/m^3$ indhold af $PM_{2,5}$ partikler i luften.

En vurdering af partikeleffekterne i Danmark er naturligvis behæftet med stor usikkerhed, da der ikke rutinemæssiget har været målt PM_{10} og $PM_{2,5}$. De hidtidige målinger tyder dog på, at de danske niveauer er af samme størrelse som de niveauer, hvorved der er fundet effekter i udenlandske undersøgelser. Anvendes de anførte dosis-respons sammenhænge for danske forhold, hvor en 10 $\mu g/m^3$ stigning i det gennemsnitlige partikelindhold (bestemt som PM_{10}) giver en øget dødelighed på ca. 4% kan det anslås, at en reduktion af det aktuelle partikelniveau på ca. 30% ville svare til et fald i den årlige dødelighed på omkring 500 personer pr. 1 millioner mennesker i de større byområder. Hertil kommer, at et betydeligt større antal personer må forventes at blive påvirket af partikelforureningen med en forringelse af sundhedstilstanden og øget sygelighed til følge. Personer med luftvejslidelser eller hjerte-karsygdomme er de mest følsomme. Til sammenligning dør der årligt omkring 550 personer ved trafikulykker.

EU's grænseværdier, der gælder herhjemme, er på 300 $\mu g/m^3$ TSP som 95-percentil og 150 $\mu g/m^3$ som årsgennemsnit. I de ovennævnte undersøgelser er der konstateret effekter ved niveauer væsentligt under disse grænseværdier. WHO anfører i deres seneste vurdering af partikler i forbindelse med opdateringen af deres Air Quality Guidelines, at der ikke kan angives nogen vejledende grænseværdi for partikler, idet der ikke ud fra data kan fastsættes et nedre effektniveau for partikler. (I 1987 var WHO's vejledende grænseværdi for partikler på 120 $\mu g/m^3$ som TSP og 70 $\mu g/m^3$ som PM_{10}, som 24 timers gennemsnit).

Endnu kan man ikke fuldt ud forklare, hvorfor partikler medfører de nævnte effekter bl.a. savnes der en afklaring af, i hvor høj grad det er koncentrationen af de helt fine partikler (ultrafine partikler mindre en 0,05 µm), der er ansvarlig for de sundhedsmæssige effekter, og hvilken rolle den kemiske sammensætning af partiklerne spiller. Endvidere synes kombinationseffekter mellem partikler og de øvrige luftforureningskomponenter at kunne have stor betydning for forureningens samlede effekt.

Udviklingen i fremtiden
Den voksende erkendelse af, at partikler udgør en væsentlig del af sundhedsbelastningen fra luftforurening, har naturligt medført, at der både nationalt og internationalt arbejdes på en reduktion af partikeludslippet.

Den danske regerings og Trafikministeriets redegørelser "Transporthandlingsplanen" fra 1990 og "Trafik 2005" fra 1993 har som mål en 50% reduktion af partikelemissionen fra trafik i byerne frem til 2010 i forhold til referenceåret 1998, og en yderligere reduktion efter 2010 (Trafikministeriet 1990 og 1993). Målet for 2010 forventes opfyldt ved allerede vedtagne skærpede emissionskrav til køretøjer. Dette har resulteret i, at nye benzindrevne personbiler siden 1990 har katalysator. Selvom en katalysator ikke direkte påvirker partikelemissionen, betyder den nødvendige blyfri benzin, at den reduceres. Skærpede emissionskrav til dieselkøretøjer vil fremover reducere partikelemissionen. Den samme virkning har den stigende overgang til dieselolie med lavere svovlindhold samt indførelse af periodisk syn af biler fra 1998.

EU kommissionen har i samarbejde med repræsentanter for den europæisk bil- og olieindustri iværksat det såkaldte Auto-Oil Programme (EU Commission 1996). Dets formål er at fastlægge en strategi for den mest omkostningseffektive metode til overholdelse af fastsatte grænseværdier for luftkvaliteten i byer i 2010, og det skal danne baggrund for fremsættelse af direktiver for emissionsnormer, brændstofskvalitet og ikke-tekniske tiltag. Denne tilgang er ny, idet tidligere regulering har taget udgangspunkt i bedst tilgængelige teknologi. I kommissionens forslag, der endnu ikke er vedtaget, skal partikelemissionen reduceres med omkring 50-65% frem til 2010 i forhold til 1995 for at nå luftkvalitetsmålene. Luftkvalitetsmålene er fastsat for bybaggrunden, og er derfor gennemsnitsmål som ikke inddrager, at eksponeringen er langt højere ved ophold i trafikerede gaderum.

Det er endnu uvist, hvilke konsekvenser Auto-Oil programmets forslag om reduktion af partikelemissionen vil have for luftkvaliteten i danske byer. Det er i denne forbindelse væsentligt at få udviklet pålidelige modeller, som kan forudsige partikelkoncentrationen, men det er også vanskeligt, fordi partikler er komplicerede mht. oprindelse, størrelse, omdannelse, afsætning og udvaskning.

Generelt har man i Danmark og andre industrialiserede lande observeret en faldende partikelforurening. Men om dette også gælder for de fineste partikelfraktioner er usikkert. Dette kan kun afklares gennem detaljerede målinger, da det måske snarere er antallet af partikler i luften end massen af partiklerne, der bestemmer de sundhedsskadelige effekter.

Monitering af de finere fraktioner er foretaget i udlandet, og fremtidig monitering af disse fraktioner herhjemme vil også blive iværksat under Det Landsdækkende Måleprogram ($PM_{2,5}$ og PM_{10}) i forbindelse med implementering af et nyt EU-direktiv, som fastsætter nye grænseværdier for luftforureningen herunder partikler, og som kræver løbende målinger.

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Paper three

Mapping Human Exposure to Traffic Air Pollution using GIS

by

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Mapping human exposure to traffic air pollution using GIS

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Abstract

An ongoing PhD project has the objective to develop a model for population exposure to traffic air pollution in order to improve assessment of health impacts and in support of risk management. A selected urban area is used as a case study. Applying a Geographic Information System (GIS), the model combines calculated air pollution data using the Danish Operational Street Pollution Model (OSPM) and available population data from existing administrative databases. A simple population dynamics model will later on be established to model the number of people which simultaneously are present in a given area at a given time applying simple time-activity patterns. The microenvironments considered are: residences, working places, and streets.

Keywords: Human exposure modelling; Air pollution; Traffic; GIS; Risk management

1. Introduction

In developed countries, road transport has become the dominant source of air pollution especially in larger urban areas where traffic air pollution in busy streets may constitute 80 to 90% of the pollution levels. At Danish ambient levels, the traffic air pollutants identified as health hazards are: fine particles, NO_2, O_3, PAH, benzene, 1,3-butadiene, ethene and propene, and aldehydes (formaldehyde, acrolein, acetaldehyde). Fine particles are believed to be the most critical pollutant and main contributor

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to excess mortality especially among people suffering from respiratory and cardiovascular diseases [1].

Use of ambient fixed monitor stations as indicator for population exposure is a very crude procedure. Personal exposure measurements give detailed information about exposure of individuals but are costly and only limited studies have been carried out. Various exposure models have been developed that combine microenvironment concentrations with individual time–activity patterns and extrapolation to the entire population to give population exposure distributions [2].

The present project takes a microenvironment approach to population exposure modelling but adds a geographic dimension by taking advantage of GIS, digital maps and administrative databases. The objectives of the research project are: (a) to develop an exposure model that combines modelled air pollution data using the Danish Operational Street Pollution Model (OSPM), population data using existing administrative databases, digital maps and a GIS. A simple population dynamics model will be established to model the number of people present in a given area during a given time using simple profiles for time spent in the various areas (at home, at work, and in transit). Additionally, ratios between indoor and outdoor concentrations will be taken into account. The model is tested in a case study for a selected urban area (b) to carry out air quality assessment and (c) to carry out examples of impact assessment of selected urban and traffic planning measures in support of risk management.

2. Methodology of exposure modelling

The method is an indirect exposure determination based on a microenvironment approach relying on presently available data and statistics. The method is illustrated in Fig. 1.

The Danish Municipality of Middelfart has been chosen as the case study area due to easy availability of most data and since a small municipality of about 19,000 inhabitants is sufficient for development and testing of the methodology although air pollution levels are relatively low.

The address point is used as an exposure indicator. Air pollution levels are calculated for each house number and all people living or working at the address are assigned the estimated air pollution. Modelled levels represent ground level concentrations since the receptor point will be located about 1 m in front of the facade and in the height of 2–3 m.

Concentration levels are determined using the Operational Street Pollution Model (OSPM) that calculates ambient hourly concentration levels based on inputs of the street configuration and hourly inputs of traffic, meteorological parameters and urban background concentrations [3]. Pollutants include: CO, NO_2, NO_x ($NO + NO_2$), O_3 and benzene. Traffic emissions are estimated based on emission factors together with average daily traffic (ADT), the percentage of heavy vehicles and the travel speed for each street section applying a default seasonal, weekly and diurnal variation in traffic loads to obtain hourly traffic inputs as well as default values for cold starts [4].

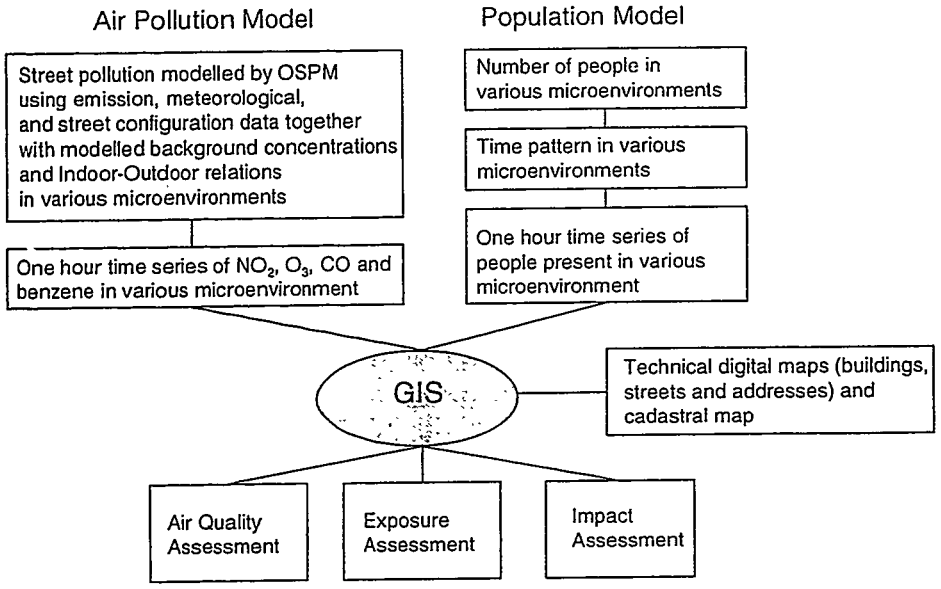

Fig. 1. Outline of the methodology for the exposure modelling in the present study.

Meteorological parameters are taken from a meteorological mast at a nearby larger city (Odense). Since the monitoring programmes only cover a few larger cities in Denmark it has been necessary to develop an extrapolation method to estimate the urban background concentrations in a small city like the case city. The reference for the method is the concentrations observed in Odense 40 km from the case city which has been scaled down applying a method that estimates background concentrations for area sources of known emission density based on the assumption that the concentrations are evenly distributed and that the dispersion depends linearly on the dispersion distance (city diameter) [4]. To determine the resulting pollution levels indoor from ambient levels empirical indoor/outdoor ratios for the various pollutants will be applied for buildings and means of transport (cars and buses).

The urban area will be divided into three general microenvironments: residences, working places, and streets.

Studies on time–activity patterns have not been carried out in Denmark from an exposure point of view. However, some statistical population data is available concerning the use of time [5,6]. Default diurnal variation of time spent in various microenvironments for different population groups will be established.

ArcView is used as the Geographic Information System (GIS) for development of the exposure model. GIS is a software that encompasses storage, retrieval, analysis and display of spatial-geographical data. GIS is a promising tool for exposure modelling due to the increase in coverage and quality of digital maps, developments in administrative databases managed by the authorities and developments of more user-friendly desktop GIS with increasing number of analytic features.

An urban landscape model has been developed using ArcView, the associated program Avenue, and digital maps to automatically generate the street configuration input parameters required by the street pollution model. The type of street configuration data generated from the digital map is shown in Fig. 2. Geocoding of the buildings is a requirement for development of the urban landscape model since the buildings have to be identified in the map in order to retrieve information in the Building and Residence Database about the height of the buildings [7].

The Danish authorities manage a number of comprehensive and detailed national databases for administrative purposes. These databases have two important features: all objects are uniquely identified and it is possible to combine the data from different databases due to common keys. An example is the address which also serves as the most important common key in this project.

The following databases are used to generate input for the exposure model: the Central Person Database (CPR), the Central Business Database (CER), the Building and Residence Database (BBR), the National Address Database, and a local traffic and road database.

The Central Person Database (CPR) managed by the Ministry of Residence Affairs

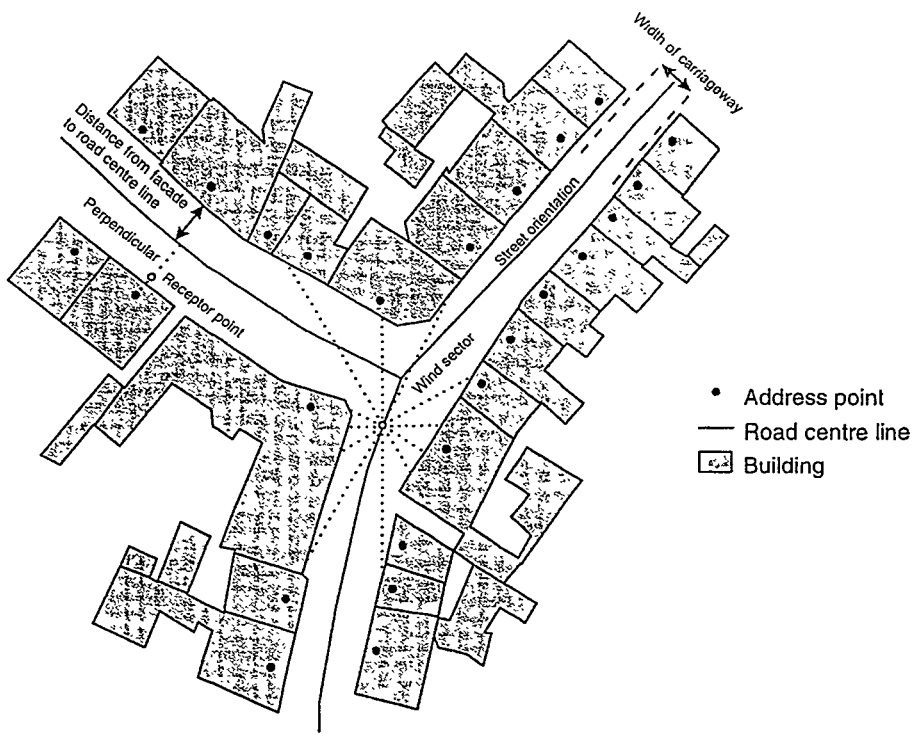

Fig. 2. Street configuration parameters generated by the urban landscape model for use in the OSPM to calculate air pollution levels. The digital map includes building polygons, address points and polylines to represent the middle of roads.

has information on each person in Denmark and the database is used to identify the gender, age and number of people living at their Residence address. Since each person is uniquely identified it is possible to estimate personal exposure for each individual for the residence microenvironment.

The Ministry of Economic Affairs manages the Central Business Database (CER) which contains information on all public and private companies. The database does not identify the individual employees but provides information about the total number of people working at a given working place identified by the address. Based on this information it is only possible to estimate population exposure for the total number of people working at various working places and it is not possible to link the exposure at the working place microenvironment to the residence microenvironment on a person to person basis.

The Building and Residence Database (BBR) managed by the Ministry of Housing contains detailed information on all buildings and residences. The database is used to estimate the height of buildings which is one of the parameters required about the street configuration to calculate air pollution.

Most municipalities manage a road and traffic database for primarily road maintenance purposes. All roads in a municipality have unique names and a unique identification code. Since most streets consist of several street sections each section is normally identified by a from–to kilometric distance. The Average Daily Traffic (ADT), the number of heavy vehicles and the travel speed are usually available from these databases and are the only input parameters needed for the model. Most municipalities including Middelfart have not established a link between their administrative traffic and road database and the digital technical map. Part of the project has therefore been to assign traffic data to the geocoded road network in the digital map. For the street environment it is only possible to estimate population exposure based on the total amount of time people spend in the streets since no information is available on how individuals travel on the network.

The Ministry of Housing has started to establish a new database containing the locations of addresses. This is the only administrative database that has a georeference, that is, the address can be geographically located according to x,y-coordinates.

Two kinds of digital maps are relevant for exposure modelling: technical and cadastral maps. Technical maps may contain many different objects since it is based on photogrammetry. The objects relevant for exposure modelling are streets, buildings and address locations which are represented as lines, polygons and points, respectively. However, a requirement for use of administrative databases is that there is a unique reference between the map and the database called a georeference or geocoding. For technical maps only addresses and streets are geocoded. An address is represented as an x,y-coordinate and located at the front door about 1–2 m inside the building polygon. The address has a unique identification consisting of a municipality code, a street code and a house number. Similarly, a street is represented as a series of x,y-coordinates and has a unique identification consisting of a municipality code and a street code. To a very large extent, the buildings can be automatically geocoded in GIS using cadastral maps and the Building and Residence Database. The cadastral map only serves as a tool for geocoding of the buildings.

3. Application and perspectives

The exposure model is an integrated flexible multi-purpose tool designed for the urban scale with a high geographic resolution primary suitable for planning purposes within population exposure assessment, air quality assessment and impact assessment.

Exploring the visualisation and analytic features of GIS the model gives a geographic dimension to input and output data. Data may be displayed as points (e.g. data related to addresses), polygons (e.g. data related to buildings or contour plots), lines (e.g. data related to streets), and grids (e.g. population density). An example of an application of the model concerning air quality mapping is given in Fig. 3 for a small selected area of the central part of the town of Middelfart which has about 10 000 inhabitants. The selected area has about 1150 inhabitants and 550 addresses.

The model gives population exposure estimates based on estimated hourly time series of ambient air pollution levels for three microenvironments separately: residences, working places and streets. For the residence environment differences in exposures between various population groups presented by gender and age can be estimated since

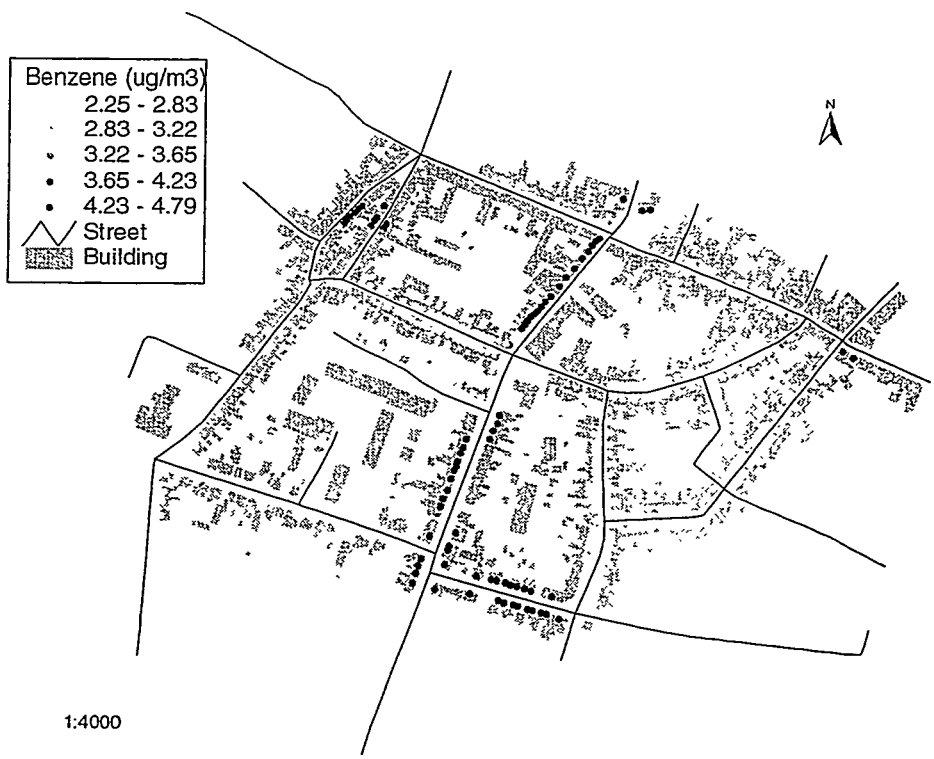

Fig. 3. Annual means of 1-h benzene concentrations ($\mu g/m^3$) in the down town area of a small city using the address point as indicator.

data on individuals is available but for the working place and street environment population exposure estimates cannot be related to individuals. It is also possible to use the model to estimate personal exposures to ambient traffic pollutants provided that detailed time–activity data is collected for the individuals in question.

Rough health risk assessment may be carried out based on the predicted exposures provided that dose–response relations are available. Comparisons of air quality guidelines with estimated air quality levels and population exposures may help assess the health risk. The model may also be used for geographical epidemiology linking exposure estimates to health endpoints in smaller geographic areas [8].

The model may serve decision-support purposes in support of risk management and risk reduction. Air quality and exposure mapping and hot spot identification may be carried out. Impact assessment of alternative urban and traffic control strategies and what-if scenarios can also be carried out provided that the consequences to the input parameters of the model are known (e.g., changes in traffic load and composition, emission, and street data).

Apart from research in issues of exposure assessment, health risk assessment, and risk management the exposure model may be further developed into an Urban Air Quality Information and Management System for use by local authorities for urban and traffic planning purposes.

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Paper four

Human Exposure to Outdoor Air Pollution

by

Hertel, O., Palmgren, F., Leeuw, F.A.A.M, Gee, D., Herbarth, O., Raaschou-Nielsen, O., Pryor, S., Jensen, S.S., Olsen, E.

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IUPAC RECOMMENDATION

HUMAN EXPOSURE TO OUTDOOR AIR POLLUTION

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Introduction

It is well known that air pollution at high levels in some cases leads to acute health effects on human beings. The classic example is the severe London smog (smoke and fog) episode in 1952 where the mortality rate in the city increased dramatically (Wilkins, 1954). In such extreme cases a link between high pollution levels (in this case SO_2 and soot mainly emitted by domestic heating) and human health effects is evident, but even when the link is less clear, air pollution may have considerable health impacts. It has been estimated that 30 to 40% of Europeans live in cities where they are exposed to air pollution levels at or above the guidelines of the World Health Organisation and the European Union (WHO and EEA, 1997). Long term as well as short term exposure to elevated pollution levels may impact on human health; long term exposure to air pollution (especially particulates) may for example increase the risk of chronic respiratory illness (see e.g. Folinsbee, 1992; Schwartz, 1994; Pope et al., 1995) and of developing cancer (see e.g. Hemminki and Pershagen, 1994; Pepelko and Chen, 1993; Knox and Gilman, 1997), and short term exposure to high pollution levels can lead to higher prevalence of bronchitis, asthma and other symptoms (see e.g. Raaschou-Nielsen et al., 1995). For both long and short term effects, the elderly, children and those suffering from respiratory or heart conditions are most at risk. Recent European studies on the externalities of electricity generation and transportation (ExternE) and green accounting exercise (GARP) pointed at health impacts from air pollution to be the single most important damage category (EC DG XII, 1995).Even though mortality dominates the damage costs, a significant part of the costs are due to morbidity (EC DG XII, 1996).

Most of the world's population is situated in cities of varying sizes. Naturally, people living in larger cities are in general exposed to higher pollution levels than those living in smaller villages. During the last decades, due to legislation concerning emissions from power plants and industry together with a steady growth in road traffic, the emission from traffic has become one of the

major sources to air pollution in larger European cities (UNEP and WHO, 1994; EEA, 1997). Traffic exhaust gases contain pollutants such as nitrogen oxides, hydrocarbons, carbon monoxide and particles. When emitted in urban streets with poor dispersion conditions, e.g. inside street canyons, substantial air pollution levels can be reached, especially at low wind speeds (see e.g. Berkowicz et al., 1996; Vignati et al., 1996).

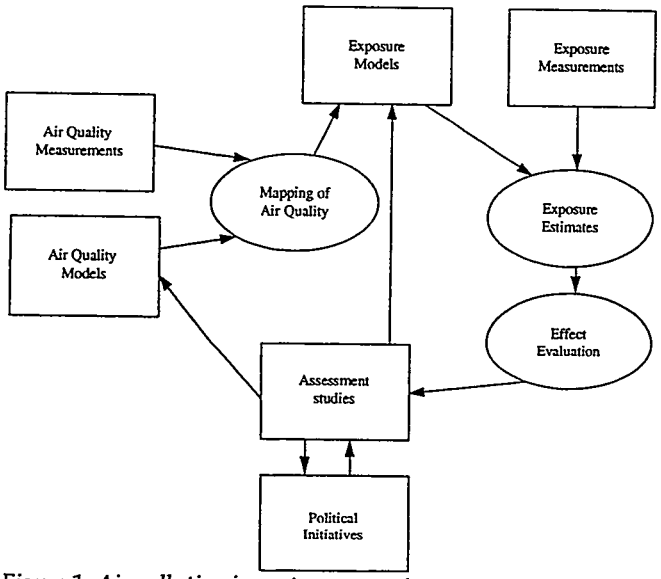

Figure 1. Air pollution impact assessment.

Another important source is residential heating. In particular when domestic heating dominates over district heating as is the case in some third world and Eastern European countries, sulphur and soot emissions are still contributing considerably to the local pollution.Studies of human exposure to air pollution are useful for several purposes, and some examples are discussed in more detail later in this paper. Accurate estimates of the human exposure are essential for a realistic assessment of the possible risk of various health effects. In the past such estimates have often been based on measurements in ambient air obtained from fixed site monitors. However, due to the large local variations in pollution levels these estimates are often associated with high uncertainties. Better estimates are needed using more sophisticated methods, and these will be described in this paper. Exposure to indoor pollution and tobacco smoking are also relevant for total human exposure, but these factors, as well as the health impacts themselves, are beyond the scope of this paper, except for a brief reference to indoor pollution.

Definitions: Concentration, exposure and dose

The concentration of a specific pollutant is the amount of material per unit volume of air. This can be expressed as mass per unit volume (e.g. microgram per cubic meter) or volume per unit volume (e.g. parts per billion (ppb) or parts per million (ppm)). Particles may also be expressed as number per unit volume.

Human exposure refers to an individuals contact with (note: contact not uptake) a pollutant concentration. Therefore it is important to make a distinction between concentration and exposure; concentration is a physical characteristic of the environment at a certain place and time, whereas exposure describes the interaction between the environment and a living subject (Sexton and Ryan, 1988). The pollution pathways may be external (e.g. skin contact) or internal (e.g. consumed or respired, which is usually the main part when air pollution is considered). The term total exposure refers to a persons contact with pollution from all sources and by all pathways, which means any kind of exposure to a given type of pollution (here restricted to air pollution exposure). For the exposure to take place two events need to occur simultaneously; pollution concentration at a particular time and place and the presence of a person at that place and time (Duran, 1982; Ott, 1985).

The concept of defining micro-environments is a common and practical tool in exposure assessment. A micro-environment is defined as a three-dimensional space where the pollution

level at some specified time is uniform or has constant statistical properties (Sexton and Ryan, 1988). The micro-environment can be the interior of a car, inside a house, or urban, suburban and rural areas etc. Integrated exposure (sometimes incorrectly referred to as dose), is the exposure that a specific person experiences over a given period of time. Using the so-called indirect method and introducing the concept of micro-environments, the integrated exposure can be expressed as:

$$E_i = \sum_j^J C_j t_{ij} \qquad \text{(Eq. 1)}$$

where E_i is the integrated exposure for person i over the specified period of time, C_j is the pollutant concentration in micro-environment j, t_{ij} is the residence time of the person i in micro-environment j, and J is the total number of micro-environments.

A constrain for using the indirect methods is that the residence time of the person (termed the time-activity pattern) needs to be known together with the pollution levels in each of the micro-environments.

Distinction between exposure and dose is important; dose is the amount of pollution that crosses one of the body's boundaries and reaches the target tissue (Sexton and Ryan, 1988). Dose and integrated exposure will therefore be identical for conditions where the body boundary does not represent a barrier for the uptake, e.g. in the case of exposure to some forms of radio activity. When the uptake of pollutants takes mainly place in the respiratory system, there is a systematic difference between exposure and dose. For example, consider four persons in a room at a given pollution level, a dead person, a child and two adults, one sitting and one running round the room. Although all four experience the same level of exposure, the dead person does not receibe any dose, the child has small lungs and needs high respiration frequency. Considering the two adults, the physically active person will receive a higher dose than the person in rest, due to difference in the frequency of respiration. An example is the findings of Van Wijnen et al. (1995) which show that car drivers have a higher exposure to certain pollutants than bicyclists in Amsterdam. However, due to the higher rate of respiration, the bicyclists had in some cases higher dose than the car drivers.

Components and types of exposure

Three different aspects are important in connection with estimation of exposure (Sexton and Ryan, 1988):

- magnitude; what is the pollutant concentration?
- duration; for how long does the exposure take place?
- frequency; how often does the exposure to a certain level appear?

It is useful to distinguish between long term and short term exposure due to the differences in effects. For some compounds there may be a health effect of the long term exposure due to accumulation in the body (e.g. lead), whereas the short-term, high level exposure may show little effect. In this case the integrated exposure will provide the needed information for estimating the accumulated amount of material in the body. However, for other compounds effects will only be observed when certain threshold values are exceeded. For some species these thresholds need to be exceeded for a certain period of time before effects are observed, and for these species it is not sufficient to determine the integrated exposure. In this case the exposure has e.g. to be determined as the total period of time with exposure to levels above the threshold value, or as the product of concentration and time for the period in which the exposure was above the threshold value. This

second concept is the basis for the proposed revised O_3 and particulate matter (PM) standards in the United States.

Exposure studies can be carried out with the aim of obtaining estimates of the exposure of the individual (personal exposure) or for a larger population group (population exposure). The exposure can be obtained from direct measurements on individuals, either a total population or selected persons, or it may be determined from model calculations.

Measuring a single persons exposure may be a relatively straight forward procedure. However, in a public health perspective the aggregated exposure of a certain population group (such as a community or an occupational cohort) is of interest, and obtaining this kind of information is a much more complex task, mainly due to the large number of people. One of the advantages of obtaining exposure data for a number of individuals in a specific group is, that it is more straight forward to link exposure to health effects and to determine exposure-response relationships. However, the most common approach is the indirect method of combining pollution levels observed at various micro-environments (often fixed monitoring sites are used to represent some of these) with data logs and diaries about the time spent in specific environments (see e.g. Fugas et al., 1972). The problem of relating fixed site monitoring data to population exposure has been discussed in the literature (Brice and Roesler, 1966; Ott and Elisassen, 1973; Cortese and Spengler, 1976). There are several assumptions implicit in the application of the indirect method (Eq. 1):

- The concentration in the specific micro-environment is assumed constant or having a well determined variation during the time the person is present there. This is often a crude but necessary assumption.
- The presence of a certain concentration in the micro-environment and the presence of the person are assumed to be independent events. This is not always the case.
- The number of micro-environments is limited to a tractably small number, disregarding or simplifying the variations within each micro-environment.
- Often the procedure for exposure estimation is based on hourly or even diurnal mean values, disregarding higher frequency acute effects of shorter term peak exposures.
- Indoor levels are often estimated from outdoor levels using fixed indoor/outdoor ratios which may in reality vary highly from place to place. Furthermore indoor sources will in some cases contribute more to indoor concentration levels than outdoor sources. These two factors are crucial since people in industrialised regions in general spend 80 to 90% of their time indoors (Nitta and Maeda, 1982; Ott, 1988).
- The effect of short-term but extreme exposure conditions are often neglected.

These assumptions need to be taken into account when the exposure estimates are used for the evaluation of possible health effects.

Air Pollution Monitoring

As already stated exposure can be obtained from direct monitoring on a specific person or indirectly by monitoring (or calculating) pollutant concentrations at various environments. A personal monitor can be a small, lightweight device, e.g. a diffusion tube, a badge or a filter with a battery-operated pump, that can be carried or worn by the person during the normal daily routine. Personal monitors make it possible to directly measure exposure for each of the individuals in smaller selected population groups (or cohorts). By combining monitoring with personal diaries or log-books, it is possible to identify areas where the highest concentrations occur and sometimes even to identify the nature of the emission sources.

For a number of pollutants, small portable personal monitors sensitive enough for measuring ambient concentrations are now available, and undoubtedly more will be available in the near future. These monitors can be divided into two types, i.e. integrated samplers that collect pollutants over a specific period of time and then are returned to the laboratory for analysis, and continuous monitors that use a self-contained analytical system to measure on location. Both systems can be passive as well as active monitors. The active monitors use a pump and a power source to move air through a collector or sensor, whereas passive monitors are using diffusion to bring the pollutant in contact with the sensor or collector. Passive diffusion samplers are today available for compounds like Carbon monoxide (CO) (Ott and Elisassen, 1973), nitrogen dioxide (NO_2) (Yanagisawa and Nishimura, 1982; Brown, 1995), nitrogen oxides (NO_x defined as the sum of nitrogen monoxide (NO) and NO_2) (Palmes and Tomczyk, 1979), ozone (O_3) (Zhou and Smith, 1997), Benzene (Brown and Purnell, 1979; Brown et al., 1981). A review of techniques for monitoring NO_2, SO_2 and Benzene has been given by Brown (1993).

Most of the personal monitors used today are integrated samplers. Passive diffusion tubes and badges have proved especially useful, since they are inexpensive and easy to operate. By choosing a subgroup from a given population group via a random selection, it is possible to obtain a picture of this total groups exposure. This makes it possible to perform investigations on relatively large population groups. The disadvantage of this type of monitor is clearly the time resolution, and with this type of monitor it is therefore difficult to identify specific sources.

For indirect estimation of exposure, the monitoring networks for ambient air pollution serve as important sources of information about levels and trends of pollution concentrations, mainly at fixed sites. These networks are established with the purpose of providing warnings in connection with pollution episodes; estimating contributions from various sources; conducting process studies; and for following trends in pollution levels. For use in air pollution exposure studies cautious data analysis is needed since these stations represent the conditions for the local site, and at nearby sites the conditions may be significantly different.

The urban monitoring network consists typically of street stations where compounds mainly emitted by traffic (for example, NO, NO_x, carbon monoxide (CO), sulphur dioxide (SO_2), soot, and total suspended particulate matter (TSP)) are measured: More detailed analysis is often also performed for different metals in the particle phase. At a growing number of stations particle measurements are reported as particulate matter in the fraction under 10 μm in diameter (PM_{10}). It is anticipated that $PM_{2.5}$ (particle size less than 2.5 μm) will soon become a standard parameter in urban monitoring networks, since recent research has indicated a clear relation between health effects and the fine fraction particle concentration levels (Dockery et al., 1993; Dockery and Pope, 1994; EC, 1997). Other compounds that are about to become commonly monitored are different kinds of hydrocarbons such as benzene, toluene and xylenes. In addition to the urban stations at "hot spot" sites in streets with heavy traffic, in many cities the networks are now supplemented with urban background stations placed in parks and backyards or (preferably) at roof top level. Besides providing valuable information about the current pollution levels, they also serve as important tools for process studies and model evaluation. Stations outside the city, far away from built-up areas may provide information on rural background levels of air pollution.

Easily measurable 'indicator' components allow estimation of levels of other compounds for which monitoring is less straightforward or highly expensive. Therefore, even though a compound may not be harmful in it self, it may still be very useful, when the compound is used as an indicator for other and more harmful pollutants. An example is the usage of black smoke data

to estimate B(a)P levels (Van Velze, 1996). Data quality requires naturally that the relation between the pollutant and its indicator is checked on a regular basis.

During recent years new measuring techniques have become available, especially a number of new optical methods have been developed for continuous monitoring. These techniques make it possible to measure a considerable number of compounds with high time resolution. With such new and better measuring techniques becoming available, it is important to define criteria for selecting compounds that should be measured. A process called "scouting, scanning and monitoring" has recently been introduced to support describing the graded application of different measuring regimes (see discussion by F. de Leeuw, in Bastrup-Birk et al., 1996). These measuring activities are:

- scouting, the performance of field campaigns with a limited number of exploring measurements in source areas and receptor areas.
- scanning, the performance of systematic measurements of air quality at zone, agglomeration, or country levels
- monitoring, routine measurements performed on multi annual and large area (e.g. country) basis

"Scouting" can give an indication of which compounds may be present at a given site. When these detected compounds are hazardous, or their presence might indicate that other more serious species are present, "scanning" is performed. Scanning involves analysing a variety of compounds with the aim of discovering which compounds may be present at harmful levels, and which should be measured on a routine basis under the monitoring networks. The procedure will identify which compounds are at concentration levels associated with human health risk.

The new EC Council Directive on Air Quality Assessment and Management, often referred to as the "FrameWork Directive" (FWD) aims at better quality and uniformity of air quality monitoring. The aims and objectives are to:

- establish objectives for improving ambient air quality in the EU using a set of exposure/dose limits where harmful effects to the environment as a whole and to human health are minimised
- assess the ambient air quality in Member States in a uniform manner
- make available information on ambient air quality to the public
- maintain good ambient air quality and improve poor ambient air quality

In order to "assess the ambient air quality in Member States in a uniform manner" the network will be designed in similar way in all Members States. Under the FWD "daughter" directives will be prepared for the following pollutants within the next year: SO_2, NO_2, particles (PM_{10} and $PM_{2.5}$), lead, O_3, benzene and CO. Other pollutants will follow. The limit values will be based on Lowest-Observed-Adverse- Effect-Levels (LOAEL). They are mainly based on WHO's recommendations revised in 1996 and to be published in 1997. The target dates for compliance, which is the time when all members states must comply with the limit values, will be based on economic and technical possibilities.

The directives will define the monitoring strategy for every pollutant. Ideally, the monitoring stations should be located in order to be representative for the exposure of the population. However, it is realised that it is necessary to supplement the measurements with other assessment tools, e.g. air quality models. The extent of the monitoring will depend on the level of the

pollution. In zones where the limit values are likely to be exceeded, well defined monitoring must be carried out. In zones with lower pollution levels so-called indicative measurements (or scouting) can be carried out, or measurements supplemented with model estimates. In zones with the lowest levels simpler estimates can be made, based on emission inventories, simple model estimates etc.

An obvious disadvantage of using fixed site monitors or models for outdoor pollutant concentrations in exposure estimation, is the already mentioned fact that people spend a large fraction of their time indoors. Some knowledge about indoor concentrations is therefore vital in assessing exposure. For some pollutants a relatively simple relation between indoor and outdoor levels may be obtained from experimental data. However, for some pollutants and at certain conditions, indoor sources may dominate over outdoor sources and thereby determine the indoor pollution levels. A number of studies have found that for some pollutants indoor levels were frequently higher than outdoor levels (see e.g. Lee et al., 1997; Wallace, 1996). For these cases special surveys need to be carried out. The ratio between indoor and outdoor pollution has been investigated in several studies (see also Yocom, 1982; Braathen, 1990), and it seems that simple ratios can be applied, if the aim is to estimate average population exposure. However, for more detailed studies of the distribution of the cohorts exposure or for exposure of individuals, more specific investigations of indoor sources may be needed.

Air Pollution Modelling

Models may serve as very useful tools for indirectly estimating human exposure. As already stated, it is not possible to perform monitoring in all the various environments that the population meets. Lifetime exposure cannot be measured directly and for this kind of study modelling is necessary. Furthermore, models can supplement the monitoring data for performing mapping of pollution levels in the various micro-environments in which monitoring is not performed. In some cases models have been developed specifically for exposure modelling. Explicitly developed air pollution exposure models are in general use some places. For example, in the United States: NEM (National Air Quality Standards Exposure Model), SHAPE (Simulation of Human Air Pollution Exposure) and HEM II (Human Exposure Model) (Ott, 1985; Sexton and Ryan , 1988) are used to carry out a variety of assessments.

For model tools to be useful in connection with exposure studies, they need to be well tested, and to describe the dominating physical and chemical processes in the atmosphere at the given location (Moussiopoulos, 1997). Further, it is important that comparative studies are performed using harmonised model tools, so that differences due to model parameterizations are avoided. For point source models such work is already initiated (see e.g. Olesen, 1995a,b) and similar work will follow for other model types like e.g. street pollution models and transport-chemistry models on regional and long range transport scales.

Since a significant part of the exposure in many cities is caused by emissions from traffic in urban streets, street pollution models serve as important tools in exposure assessment. Examples of such models are STREET (Johnson et al., 1973), CPBM (Yamartino and Wiegand, 1986), OSPM (Berkowicz et al., 1997), CAR (Boeft et al., 1996). Examples of use of street pollution models are the studies bus drivers exposure during the working day and of postmen along their mail routes, which were shown to be well represented by the OSPM model (Hertel et al., 1996).

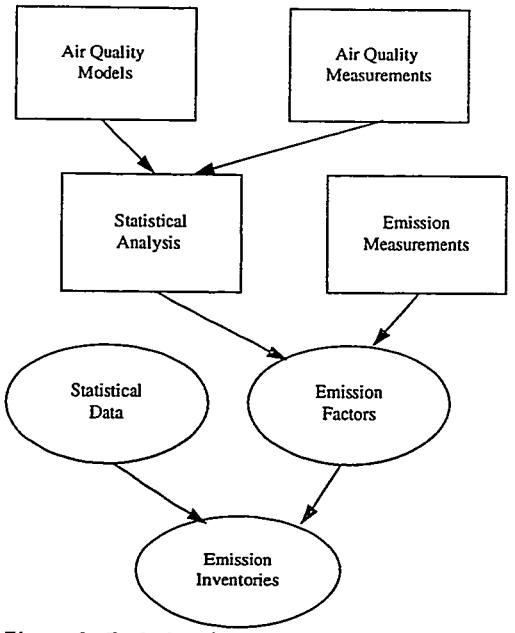

Figure 2. Emission factors may be determined from air quality measurements by use of models.

Essential for the application of dispersion models is reliable information on emission. However, in many cases the emission factors for the different vehicle categories are not well known. In this case a so-called inverse modelling can be performed. This involves a backward calculation procedure, using street pollution models and air quality measurements from a fixed site street and urban background monitor, meteorological data and traffic counts, to estimate the actual emission factors for the specific car fleet (see e.g. Palmgren et al., 1995). The obtained emission factors can then be used for calculations in other streets for which monitoring data are not available.

GIS-systems are being integrated into many monitoring programmes, not only for presentation purposes, but also as useful tools in producing mappings of air pollution levels in urban areas etc. The dispersion of pollution in urban areas is highly influenced by the presence of buildings, and therefore a detailed description of the specific street configuration is needed in order to model the concentration levels from the traffic in a given street (see e.g. Berkowicz et al., 1996). While street configuration data may be obtained manually or from questionnaires filled out by local authorities (see e.g. Vignati et al., 1997), automatic map interpretation programmes may also be designed to provide the street configuration of a given street or even a specific address, which can then serve as a useful tool in exposure modelling (see e.g. Hansen et al., 1997).

Time-activity pattern

One of the simplest ways to perform human exposure investigations is to relate exposure to outdoor pollutant concentrations at the front door, which may be the only feasible approach in many retrospective studies (see e.g. Raaschou-Nielsen et al., 1996). Other types of studies offer the opportunity to measure personal exposure and at the same time collect information about time-activity patterns. In Hagen et al. (1991) 5800 persons in a northern region of Norway were requested to fill in a diary concerning their activities and well being during a campaign period in which a number of air pollution measurements were performed simultaneously. The information concerning their time pattern in different activities was later used as input for an air pollution exposure model.

In most cases only sparse information about the time budgets, activity pattern and commuting behaviour of the population are available. For example, information about the time in the most critical micro-environments with respect to pollution, can seldom be obtained. It is especially important to investigate whether sensitive groups like elderly people, children and people with different illnesses have time-activity patterns which differ significantly from the general population. There may also be large regional differences, and differences over respective age groups. This suggest that for long term exposure studies different time-activity pattern should be applied over the life time of a person. Furthermore, the residence of a person changes many times during life. Swedish studies indicate that for the Swedish population as a single group, there is an average of about six residential address changes during a lifetime (Svensson et al., 1989), but the number seems to be higher in North America (Letourneau et al., 1995).

There are different ways to obtain this kind of information. A review of time-activity patterns in exposure assessment is given in Ackermann-Liebrich et al. (1995). The information needed in such studies include location of the activity, the period or time when the activity took place (e.g. time of day, phase in life) and the duration of the activity. The information that can be obtained for these kinds of investigations can be divided into two categories: data that are collected by national level institutes such as a national bureau of statistics, building registers etc. (data like age, occupation, socio-economic status, placement of houses, working places etc), and information collected as a specific part of air pollution epidemiological studies (from e.g. questionnaires).

The diary or log-book can be supplemented by an automatic device that is activated each time the person in the experiment has made an entry in the diary. Thereby the time is automatically stored. A totally different concept from diary or log-book is an electronic time-activity monitor or data logger (see e.g. the overview by Jantunen, 1995). Such devices may automatically detect whether the person is outdoor or indoor etc. This kind of device has rarely been used in studies to date, but may be useful tools in the future as the technology becomes more advanced. Related technology to adopt in future systems are likely to include the Global Positioning System (GPS) receiver, based on satellite information, that can compute the location at any time with a precision of about 15m, and related environmental in situ data (e.g. temperature, humidity, light etc.).

An important issue regarding time-activity patterns for exposure studies is to reach for consensus on which parameters are needed. Data may be obtained from more or less automatic devices or from questionnaires. However, it is important that the right information is obtained, stored and made available in a useful form; e.g. the design of questionnaires for determination of time-activity patterns is not an easy task from this perspective.

Biological markers

This kind of investigations involves analyses of bodily material (e.g. urine, saliva, blood, exhaled breath, faeces, hair, nails etc.) in which the concentration of a pollutant, or the metabolite of a pollutant, is determined. Biomarkers give information about the dose that has been received and taken up by the body.

Biological monitoring has three major advantages over environmental monitoring for estimating health risks (Sexton and Ryan, 1988):

1. Only pollutants that cross the boundary and enter the body are included.
2. Biological markers are more directly related to the biological processes from which health consequences arise.
3. Biological monitoring can serve as a basis for total risk estimates from multiple chemicals. It takes into account all exposure from all routes.

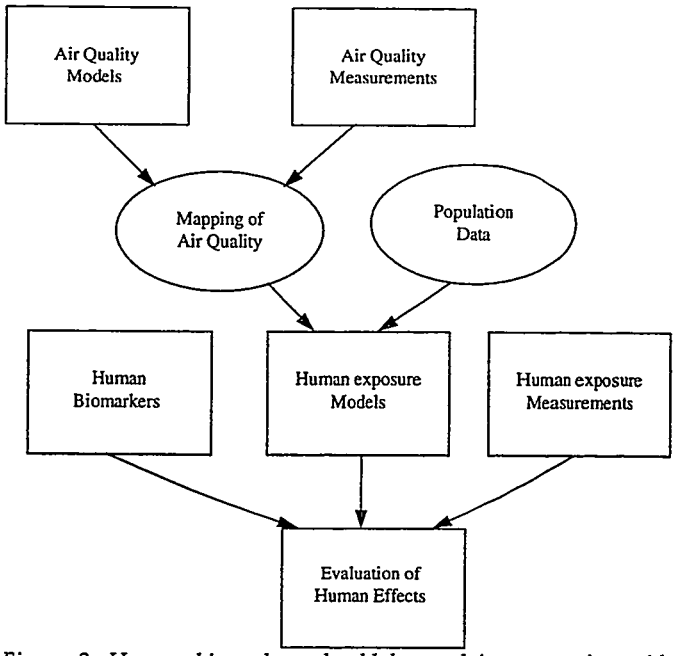

Figure 3. Human biomarkers should be used in connection with exposure data obtained from measurements and modelling.

The greatest disadvantage is that at present specific markers are known for only a few situations, which allow a specific (selective) inference to the air pollution exposure components. For this reason, exposure studies have to investigate simultaneously the internal dose and external (close to human) exposure. This supports the statement, that exposure measurements and biological markers should be used to supplement each other. caution should be taken in the interpretation, when analyses of biomarkers are made and the results are compared with exposure data and estimated dose. Due to the various cleaning processes in the body, material is steadily removed from the target organs, continously changing the concentration in the organ. Description of these processes are beyond the scope of this paper.

Recommendations

Studies of human exposure to air pollution have a number of different uses. It is important to choose the right strategy for the study in order to obtain the optimal accuracy of the exposure estimates. Examples of uses of exposure studies are:

- impact assessment on population exposure, e.g. connection with various types of management i.e. traffic and city planning
- comparison of the exposure of different specific population groups e.g. in connection with epidemiological studies
- estimation of the average or peak exposure levels of the population in connection with e.g. risk analyses
- validation of exposure estimates e.g. in connection with modelling
- identification of the most important sources to pollution exposure

The quality of human exposure data is increasing with improved techniques, and this helps exposure studies to become important tools in city and traffic planning. An example of their use is in optimizing the spatial distribution of pedestrians and bicyclists, compared to urban traffic, with respect to air pollution exposures. The best distribution may be obtained with the help of monitoring studies along various alternative routes, and from modelling of the exposure levels.

For specific population groups (or cohorts) personal exposure monitoring may provide the needed information. For a larger cohort a random selection of a subgroup may provide information that can be extrapolated to the total cohort. New monitoring techniques (active as well as passive) have made it possible to perform personal monitoring for an increasing number of compounds. The new techniques need to be carefully validated against well documented monitoring techniques.

For population exposure of larger cohorts, the above mentioned method with random selection of a subgroup of the cohort may still be applied. However, this may depend on what is technically

and economically possible in the specific study. For long term exposure, it will often be necessary to use indirect methods for estimating human exposure. Furthermore, it cannot be expected that measurements on larger cohorts will be carried out on regular basis. Therefore, when the aim is to follow the development in human exposure, indirect methods to estimate exposure levels will often be the only possible approach. Combinations of monitoring at fixed site locations and use of model tools are useful for obtaining concentration levels at the various outdoor micro-environments. However, there is still a need for more information about the levels and variations in concentrations in various micro-environments. In some cases these are practically unknown.

Most people in the industrialised world live in cities where local sources (especially traffic) contribute significantly to the pollution levels. Inverse modelling (backward calculation methods) using monitoring data and model calculations for the fixed site monitor positions can at specific idealised conditions be used for estimation of emission factors from e.g. traffic when these factors are not directly available. The presence of buildings has a major impact on the local pollution levels in cities. For estimation of urban street pollution levels, local street configuration data are needed. The use of GIS may serve as a tool for automatic generation of such data for large numbers of streets in given urban areas based on present technical digital maps and databases whenever these are available.

For exposure modelling there is still a great need for more detailed databases concerning time activity patterns of the different populations combined with exposure data. Questionnaire investigations combined with the various available statistical data from national bureau's need to be compiled into time-activity databases for use in exposure studies. Personal exposure studies can serve as essential tools for validation of population exposure estimates. Diaries and log-books carried out in connection with such studies should serve as information for general time-activity pattern.

Assessment studies where the effects of different legislation for reduction of emissions are investigated can only be carried out by means of model studies. Detailed parameterization allow one to construct scenario studies and do sector by sector studies. Such parameterizations require accuracy, which in turn require innovative approaches in sampling and analysis. For practical use, in the near term, investigations need to be based on well tested models that describe the dominating physical and chemical processes governing the pollutant concentrations at the various micro-environments. Model harmonisations and careful sensitivity and validation tests are obvious tools in this work.

The identification of the most important sources to human exposure may be compiled from personal exposure measurements with high time resolution combined with detailed diaries/log books. For measuring techniques with long sampling time, statistical methods may be applied for identification of the most important sources. Other types of studies can be measurements at specific sources and use of receptor modelling on detailed data from fixed site monitoring stations. The last two approaches may be used to identify sources to outdoor levels in specific areas.

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A street pollution model for use in epidemiological studies: evaluation with measured levels of nitrogen dioxide and benzene

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An air pollution model for use in epidemiological studies: evaluation with measured levels of nitrogen dioxide and benzene

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Abbreviations: OSPM, operational street pollution model

r, Pearson's correlation coefficient

Key words: air pollution, benzene, epidemiology, modelling, nitrogen dioxide, traffic

ABSTRACT

The aim of the study was to evaluate the predictions derived from the Danish Operational Street Pollution Model (OSPM) when the input data are obtained by simple methods that could be used in large-scale epidemiological studies. The model calculations were thus compared with passive sampler measurements of nitrogen dioxide and benzene at 103 street locations in Copenhagen, Denmark, and at 101 locations in rural areas. Data on traffic and street configuration were collected by means of a simple registration scheme in which forms were filled out by local municipal authorities. Meteorological data were derived from routine measurements at Copenhagen airport, and data on background air pollution were based on a simple empirical model. Differences in air pollution levels between rural areas and Copenhagen and differences in nitrogen dioxide concentrations at various locations in Copenhagen were well reproduced by the OSPM. The correlation coefficients (r) between the measured and the predicted half-year average concentrations of nitrogen dioxide in Copenhagen were between 0.75 and 0.80 for various degrees of precision of the input data for the model. The results indicate that the OSPM used with the presented methods for generation of input data might be useful in assessing long-term exposure to air pollutants in epidemiological studies.

INTRODUCTION

The adverse health effects of air pollution derived from traffic have been a great concern during the last few decades (Ayres *et al.*, 1973; Stern *et al.*, 1981; Gamble *et al.*, 1987; International Agency for Research on Cancer, 1989; Raaschou-Nielsen *et al.*, 1995; Rudell *et al.*, 1996; Hansen *et al.* 1998). Ideally, epidemiological studies require detailed information about the exposure of each participant, but the measurements required are usually impossible, very expensive or time-consuming to perform. Alternatively, traffic pollution models can be used to estimate levels of exposure. So far, such models have been used mainly in the context of town and traffic planning and, despite their great potential, only rarely in air pollution epidemiology (Oosterlee *et al.*, 1996). This maybe due to uncertainty about their validity but also because of the difficulty in obtaining the basic information required for running the models. Exploitation of such models in large-scale epidemiological studies therefore depends on the development of simple methods for collecting input data and verification of model results.

Measurements of nitrogen dioxide and benzene at a wide variety of locations in Denmark were used in order to verify calculations derived from the Danish Operational Street Pollution Model (OSPM) (Hertel and Berkowicz, 1989a). Previous studies showed good agreement between measured concentrations and those predicted by the OSPM when precise input data were used, including detailed traffic counts and measurements of background concentrations and meteorological conditions close to the street location (Berkowicz *et al.*, 1996, 1997a).

This study focuses on the possibility of using OSPM in future epidemiological studies, with use of a simple registration scheme to collect information about traffic and street configuration. The significance of the precision of data on street configuration and meteorological conditions was evaluated, and the performance of the model was studied over different averaging times.

METHODS

Measurements

Locations and sampling periods. Nitrogen dioxide (NO_2) and benzene were measured at 103 street locations in central Copenhagen, Denmark, and 101 locations in rural areas or small towns (denoted "rural areas" in the following), 20-50 km outside Copenhagen. The traffic density on the streets ranged from 10 and up to 57 700 vehicles per day in Copenhagen and from 2 and up to 3000 vehicles per day in rural areas.

Seven measurement campaigns covering about 30 locations each were begin in October-November 1994 and in April-June 1995. During the first week of the six-month campaigns, the mean concentrations of NO_2 were measured at each location, and the mean concentrations of benzene were measured at a subsample of locations during the 1995 campaigns. At each location, monthly mean concentrations of NO_2 were measured for six consecutive months.

Sampling methods and laboratory analyses. Passive diffusion samplers fixed under a cap

of stainless-steel were used for all measurements. In Copenhagen, the samplers were placed about 0.5 m from the front of a building and about 4 m above street level. In rural areas the samplers were placed either in a garden or 0.5 m from a facade and about 1.5 m above the ground.

Benzene was sampled by means of Perkin-Elmer diffusive steel tubes containing 200 mg Tenax TA per tube as the adsorbent. The tubes were sealed with Swage-lock fittings before and after exposure. Samples were analyzed by a thermal desorption system coupled to a gas chromatograph with a split dual-column system: one with a flame ionization detector and one with a mass spectrometric detector. One week of exposure corresponded to a detection limit of 0.5 $\mu g/m^3$ (mean plus 3 x standard deviation for 20 unexposed tubes); eight measurements below the detection limit were assigned the value 0.25 $\mu g/m^3$. On the basis of 14 replicated measurements, the coefficient of variation (standard deviation divided by mean) was estimated to be 26%.

The concentrations of NO_2 during the first week were measured by a badge containing triethanolamine, which absorbs nearly 100% of NO_2 and converts it to nitrite. The nitrite was analyzed on a segmented flow analyzer using Saltzman's reagents, followed by spectrophotometric detection at 540 nm. A one-week exposure corresponded to a detection limit of 0.8 $\mu g/m^3$ and on the basis of six replicated measurements, the coefficient of variation was estimated to be 4%. The method is described by Yanagisawa and Nishimura (1982) and by Raaschou-Nielsen *et al.* (1997).

The mean NO_2 concentrations over one month were measured by Palmes tubes (Palmes, 1981), with three exposed and one unexposed (blank) tubes. The analytical method was identical to that used for the badges. A one-month exposure corresponded to a detection limit of 0.6 $\mu g/m^3$. On the basis of the replicated measurements at all locations, the coefficient of variation was estimated to be 10.5%.

The model

OSPM. Traffic pollution at the sites of measurement was calculated from the OSPM, which has been described by Hertel and Berkowicz (1989a,b,c) and Berkowicz *et al.* (1997a,b). Only a brief introduction is given here.

In the OSPM, the concentration of pollutants (c) in the street is calculated as the sum of the concentration caused by local street traffic (street contribution c_s) and the concentration caused by other sources (background contribution c_b), such that

$$c = c_s + c_b \,. \qquad (1)$$

The concentrations are calculated hour by hour as a function of the actual emissions and meteorological conditions. The principle of simple addition expressed in formula (1) is valid only for chemically unreactive pollutants; a different method is applied for NO_2.

The street contribution (c_s) is calculated as the sum of the direct contribution from

traffic and the contribution from recirculation in the street. A plume model describes the direct contribution that depends on the wind speed at street level, which in turn depends on the wind speed at roof level and the height of the houses along the street. The recirculation is due to the creation of a street canyon vortex (Albrecht, 1933) with a wind direction at street level that is opposite to that above roof level. The concentrations of pollutants due to recirculation of car exhaust are calculated from a simple box model, in which ventilation of the "recirculation box" depends on the wind speed at roof level and the ventilation area; the latter depends on the height of the surrounding buildings and the width of the street.

The formation of a vortex depends on the presence of buildings on the upwind side of the street. Therefore, the combination of orientation of the street, the configuration of the buildings, and the wind direction is important (Berkowicz *et al.*, 1997a). Different building configurations can be accounted for in the model by specifying the height of the buildings along the street in specific wind sectors.

In order to calculate NO_2 concentrations, a chemical sub-model is used which takes into account the reactions that lead to formation of NO_2. Only about 5% of the nitrogen oxides in car exhaust is in the form of NO_2 and most occurs as nitric oxide (NO). The sum of NO and NO_2 is denoted NO_x. The chemical formation of NO_2 in street air is governed by the reaction between NO and ambient ozone (O_3), as follows:

$$NO + O_3 \underset{J}{\overset{k}{\rightleftarrows}} NO_2 + O_2, \qquad (2)$$

where k is a reaction rate coefficient dependent on temperature and J a reaction rate

coefficient dependent on solar radiation. The reaction is reversible because NO_2 photodissociates to form NO and O_3 in the presence of solar radiation. In the OSPM, the concentrations of NO_2 are calculated by taking into account the concentration of NO_x (estimated from the dispersion model), the concentration of O_3 in the background air entering the street, the reaction rate coefficients k and J, and the residence time of car exhaust gases in the street (Hertel and Berkowicz, 1989b; Berkowicz *et al.*, 1997b). The last depends on wind speed and the height of the surrounding buildings. In streets with low or no buildings, the residence time of car exhausts may be short and the reaction between NO and O_3 will produce less NO_2 than in a street with tall buildings. In streets with dense traffic, the amount of O_3 will often limit the formation of NO_2 (Palmgren *et al.*, 1996).

The main contribution to the pollution level in streets with low traffic density may be from emissions in a neighboring street. The contribution that comes from such a neighboring street (c) is calculated from the following simplified line source model,

$$c = \frac{Q}{u(H_s + 0.1x_s)}\ , \qquad (3)$$

where Q is the emission rate per unit distance from traffic in the neighboring street, u is the wind speed at roof level and H_s is the typical height of the buildings in the area. The term $H_s + 0.1x_s$ is the height to which pollution is dispersed at a distance x_s from the street. In this study, only streets within 50 m were taken into account. As a very crude approximation, it was assumed that the distance from a neighboring street to the receptor point was always 50 m, and formula (3) was applied regardless of the

actual wind direction.

OSPM predicts the concentrations on the pavement of the street, by default. When the calculation point was located at distance d from the curb, the following formula was used,

$$c = \frac{c_{edge} * H}{H + 0.1d}, \qquad (4)$$

where c_{edge} is the concentration calculated from the OSPM for the pavement location, and $H + 0.1d$ accounts for additional dilution over the distance d from the street. H is the height of the buildings along the street. It must be emphasized that this method is very simple and may not be appropriate for all distances and building configurations.

Input data for the OSPM

The model requires input data about (1) traffic and emissions in the street, (2) street and building configuration, (3) meteorological data and (4) background pollution. The first two were derived from a registration scheme, in which the relevant municipal office and authorities for each location in this study were asked to provide the required information from traffic counts, maps, local knowledge and judgements. The registration scheme and the methods used for pre-processing the information obtained is described in detail by Vignati *et al.* (1997), and the background pollution modelling by Jensen (1998). Only an outline of the methods is given here.

Traffic and emissions. The registration scheme provided the following information about the traffic at the measurement site: (1) traffic density (average daily traffic); (2) proportion of heavy (greater than 3.5 ton) vehicles, divided into four intervals; (3) average driving speed, divided into five intervals and (4) traffic density on cross or parallel streets within 50 m of the site, with five intervals. The last question was omitted if the traffic density at the site exceeded the traffic density at cross or parallel streets.

As the OSPM requires hourly data on traffic, average daily traffic was distributed according to default diurnal traffic profiles (Jensen, 1997). Standard diurnal variation in the percentage of cold starts was also included.

Hourly traffic emissions of NO_x and benzene were derived from traffic flow (vehicles per hour) and vehicle-specific emission factors (g/km). The emission factors for NO_x were based on the results of a Danish study (Miljøstyrelsen, 1991), whereas those for benzene were deduced from analyses of measurements at a monitoring station in Copenhagen (Palmgren *et al.*, 1995). The introduction of catalytic converters in the Danish car fleet has influenced emission factors; therefore, the proportion of cars with catalytic converters was derived from car sale statistics (1994: 25%; 1995: 30%). The effect of traffic speed was taken from the Norwegian traffic model VLUFT (Torp *et al.*, 1995). Data on historical changes in emission factors (1960-1995), to facilitate calculations for the past, were provided by the Danish Technical University (S. Sorensen, personal communication).

Street configuration. The site at which the concentration of pollution was estimated in the OSPM was defined as outside the front door of a particular address. The street was characterized on the basis of information about (1) the type of street: municipal or private, or county or state and (2) the street and building configuration, with five street categories, ranging from a few houses on an open street to tall buildings on both sides of the street (Table 1), based on the categories in the Dutch street pollution model CAR (Eerens *et al.*, 1993). The registration scheme provided information about the distance between the facade of the house and the opposite curb or, when there were buildings on both sides, the distance between the building facades. For street canyons, the street width was considered to be the distance between the building facades given in the registration scheme. For the other street categories, the street width was estimated from the information about traffic density (Jensen, 1997). The registration scheme also provided information about the height of the building at the address. We used assumptions about the height and the width of the other buildings and used geometrical considerations to calculate wind sectors for different building heights in various types of street categories (Vignati *et al.*, 1997) .

le 1

For all addresses, it was arbitrarily assumed that the street was orientated north-south and that the calculation point was on the west side of the street. Furthermore, for all addresses, only 50 m of the street in both directions from the calculation point was taken into account.

In order to test the sensitivity of model predictions to the precision of data on street configuration, a more precise but time-consuming method (in the following denoted

"detailed method") was used to gain information about the street configuration at the sites of measurement in Copenhagen. Maps and observations about the locations were used to obtain information about the actual orientation of the street, the location of the measurement point, the length of the street in both directions from the receptor point, the street width, sectors with no buildings, and the distance to proximate streets (if any) with dense traffic. Subsequently, input data based on the registration scheme was replaced by this information and the OSPM calculations were repeated. In the rural areas, in which the street contribution was small compared to the background contribution, only the method based on the registration scheme was used.

Meteorological conditions. Hourly observations of wind speed, wind direction, global radiation and temperature for 1994 and 1995 were obtained from Copenhagen Airport, located about 8 km from the center of Copenhagen, and used in the model calculations. Global radiation, which was used to calculate photodissociation of NO_2, was inferred from routine cloud cover data using the relationships given by Nielsen *et al.* (1981).

Meteorological data for the relevant years will not be available for most retrospective epidemiological studies. Data for another year used as a standard would be roughly correct with regard to seasonal changes, but might be wrong for day-to-day variation. In the present study, we repeated the OSPM calculations using meteorological data for 1994 as a substitute for 1995 and vice versa, in order to test the effect of using meteorological data from another year.

Background pollution. Measurements of background pollution were not available near all the addresses in this study. Therefore, an empirical background model was developed in which NO_x and carbon monoxide (CO) concentrations measured during 1994 and 1995 at a monitoring station in Copenhagen (roof level) and NO_2 and O_3 concentrations measured during the same period at monitoring stations in rural areas were used (Jensen, 1998). The rural background concentrations of NO_2 and O_3 (hour by hour) were calculated from the measured two-year average concentrations multiplied by predefined factors for seasonal and diurnal variation; urban NO_x concentrations (hour by hour) were calculated in a similar way. The two-year average NO_x concentration for urban locations other than Copenhagen was calculated from the value measured in Copenhagen multiplied by a reduction factor that corresponded to the population size of the urban location in question (Jensen, 1998). Background concentrations of CO in urban areas were estimated by the same method as applied for NO_x whereas rural background concentrations of CO were assumed to be 50% of the background level in Copenhagen.

For rural locations, the hourly concentrations of NO_x were calculated on the assumption of a photochemical equilibrium between NO, NO_2 and O_3 (reaction (2)). The background concentrations of NO_2 and O_3 in urban areas were calculated using a procedure similar to that for calculating NO_2 in streets (Hertel and Berkowicz, 1989b) but with the residence time determined by the size of the city area and the wind speed (Hertel and Berkowicz, 1990). The driving parameters in this process are the rural concentrations of O_3 and NO_2 drifting over the city and reacting with urban background NO_x. The background concentrations of benzene at rural and urban

locations were calculated from the empirical relationship between benzene and CO found in a busy street in Copenhagen.

The measurements of this study indicated that the contribution of a city to the urban background concentration declines with distance from the city center (Figure 1). An exponential fit to the data reveals a dependency that corresponds to the equation: concentration ($\mu g/m^3$) = 25.4 exp(-0.39 x distance(km)). Although the function was found for NO_2, the same dependency is assumed to be valid for NO_x. The following formula was used to calculate the urban background concentration with respect to distance from the city center:

$$[NO_x]_{ub} = [NO_x]_c \exp(-1.6(d/R)) + [NO_x]_{rb} , \quad (5)$$

where $[NO_x]_c$ is the contribution of the city to the urban background in the center of the city and $[NO_x]_{rb}$ is the rural contribution, d is the distance to the city center and R is the radius of the city (which is approximately 4 km for Copenhagen). The introduction of R into the exponential decay factor is based on the assumption that the decline in background concentrations with distance from the center is faster for smaller than for larger urban areas; however, no experimental justification for this assumption can be given presently.

Figure 1

Calculations

Average concentrations corresponding to the period of measurements were computed with the OSPM for each location. The half-year average concentrations for each

location were calculated as the time-weighted average of six consecutive monthly mean measurements. Scatterplots and Pearson's correlation coefficients (r) were used to evaluate the associations between calculated and measured concentrations. Correlation coefficients were calculated separately for urban and rural locations and were based on values transformed by the natural logarithm.

RESULTS

We present the results of our assessments of the performance and sensitivity of the model with respect to the length of the averaging period, the precision of input data on street and building configuration and the meteorological data used for the calculations. Moreover, model performance for NO_2 and benzene are compared. Finally, traffic density as an indicator of air quality is compared with the results of the model.

Length of the averaging period. For locations in Copenhagen, the correlation between predicted and measured concentrations was much stronger for half-year values (r between 0.75 and 0.80) than for monthly mean values (r between 0.61 and 0.68), whereas in the rural areas the correlation was weaker for the half-year values (r = 0.70) than for monthly mean values (r = 0.76) (Table 2). Moreover, Figure 2 shows much smaller variation in measurements for a given predicted half-year value than for a given predicted monthly mean value, thus, indicating more precise model predictions for long-term averages.

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Precision of input data on street configuration and meteorological conditions. Tables 2-3 show a consistent pattern in Copenhagen of stronger correlations when the input data on street configuration were obtained by the detailed method and when the actual meteorological data were used in the calculations. In rural areas, the use of meteorological data from another year had no effect on the correlations.

One-week mean concentrations of NO_2 and benzene. Figure 3 shows plots of modelled versus measured concentrations for NO_2 (N=203) and benzene (N=128). Table 3 shows a similar degree of correlation between predicted and measured values for NO_2 and benzene for locations in Copenhagen.

In rural areas, when all measurements were considered, the correlations for NO_2 were much stronger than for benzene. However, NO_2 was measured during seven different weeks in October, November, April, May and June, whereas benzene was measured only during April, May and June. When only data from locations where simultaneous measurements were made of NO_2 and benzene (N=127) were considered, the range of the background NO_2 concentrations was reduced significantly, thus lowering the correlation coefficient dramatically in rural areas. As a result, the correlations were similar for NO_2 and benzene in rural areas as well: the predicted and measured concentrations were almost uncorrelated for both pollutants.

Figure 3
Table 3

Traffic density as an indicator of air pollution levels. Table 2-3 show that the measurements of NO_2 and benzene in Copenhagen were stronger correlated with predicted concentrations than with traffic density. This was true even when the least precise

input data were used. Moreover, the one-week measurements (Table 3) indicate that the correlation with traffic is stronger for benzene than for NO_2.

In rural areas, where local traffic makes a negligible contribution to air pollution, the measured concentrations were not correlated with traffic density.

Systematic differences between predicted and measured values. We have hitherto focused on the correlation between predicted and measured concentrations, without considering any systematic differences between the two variables. Figures 2-3 indicate that the predicted values in general tended to be higher than the measured ones. This is confirmed in Table 4, which shows higher predicted mean values.

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DISCUSSION

Length of the averaging period. There may be several explanations for the stronger correlation for half-year averages in Copenhagen. Any model uncertainty or random measurement error would tend to weaken the correlation between predicted and measured values. Model uncertainties could be due to either limitations in the descriptions of various physical and chemical processes or to imprecise input data on e.g. street configuration, traffic density or meteorological conditions. Such uncertainties will draw the predictions in different directions under different conditions (of e.g. wind direction and wind speed), and over- and underestimates of short-term values will tend to equal out when longer averaging periods are considered. Vignati

et al. (1997) demonstrated that model results for short-term mean concentrations were much more greatly affected by improper input data than were long-term predictions. Similarly, random measurement errors diminish when several measurements are averaged. Systematic errors associated with the model or the measurements will not diminish similarly.

The weaker correlation for half-year values for the rural locations is probably due to the significantly lower variation for the half-year than for the monthly mean concentrations. The absence of traffic and other major local sources of NO_2 reduces the variation between locations to a minimum and makes seasonal (month-to-month) changes in background concentrations the major source of variation. Calculating half-year average concentrations from six consecutive monthly mean values will therefore tend to smooth out the variation. It is noteworthy that the weaker correlation does not imply less precise model predictions.

Information about the significance of the averaging period extracted from comparisons with the one-week mean measurements should be treated with caution, because those measurements were performed during other periods than the monthly mean measurements.

One-week mean concentrations of NO_2 and benzene. Despite the similar correlations between the measured and predicted concentrations of benzene and NO_2, the model performance for the two pollutants is not necessarily similar in general. First, vehicle

emissions of benzene depend much more than emissions of NO_x (the precursor for NO_2) on driving conditions, consequently greater uncertainty can be expected in benzene emissions when rough estimates of driving speed are used, as in this study. Secondly, owing to the non-linear relationship between NO_2 and NO_x concentrations (Berkowicz *et al.*, 1997a), the uncertainties related to dispersion modelling decrease in importance at high concentrations of NO_x, whereas those related to estimated O_3 concentrations increase in importance. Factors related to chemical reactions are irrelevant for the relatively inactive benzene. Thirdly, contributions from other sources than traffic may be different for benzene and for NO_2. In urban areas with dense traffic, other sources are normally of limited importance for both pollutants, whereas in rural areas local sources can contribute significantly. Finally, it should be noted that the measurement uncertainty was substantially higher for benzene than for NO_2 and that measuring benzene with a similarly low uncertainty would tend to increase the observed correlation between the measured and predicted concentrations of benzene.

Traffic density as an indicator of pollution levels. Several epidemiological studies have used traffic density as a surrogate measure for traffic-related air pollution at residential addresses (Savitz and Feingold, 1989; Wjst *et al.*, 1993; Duhme *et al.*, 1996; Brunekreef *et al.*, 1997). Use of this measure is certainly a simpler, less time-consuming method for assessing exposure than performing calculations with street pollution models. However, the results of this study showed that calculated concentrations correlated stronger than traffic density with measurements of NO_2 and benzene. Therefore, we would expect that the use of calculated concentrations in stead of traffic density would reduce the misclassification of the exposure.

Traffic density correlated stronger with measurements of benzene than with those of NO_2. The main reason for this difference is that benzene is a primary air pollutant from the traffic and NO_2 mainly a secondary one, and primary air pollutants can be expected to correlate better with traffic (their direct source) than secondary pollutants.

Systematic differences between predicted and measured values. The systematic differences between predicted and measured values could be due to deficiencies in either the model parameterization, the measurements (e.g. the calibration) or both. Although limited agreement between the predicted and measured concentrations would be crucial if the predictions were used to evaluate violations of threshold values, the strength of the correlation (not the agreement) would be the major concern in an epidemiological context.

CONCLUSIONS

This study shows that the differences in air pollution concentrations between urban areas with dense traffic and rural areas without dense traffic were well reproduced in the model. The model reproduced variation in measurements within rural areas due to seasonal changes in background concentrations but could not reproduce minor differences between locations. In urban areas, where traffic is the major source of ambient air pollution, the predicted concentrations showed quite strong correlations with measured concentrations for long-term average concentrations, even when imprecise input data were used.

Moreover, the study shows that the model predictions for NO_2 and benzene, even when the crudest input data are used, correlate stronger than traffic density with measurements.

Altogether, the results indicate that the OSPM could be useful in assessing exposure in epidemiological studies of air pollution. The association between predicted street concentrations and the personal exposures of individuals living inside the dwellings on the street have not yet been addressed.

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Legends to figures

Figure 1. Dependency of the contribution of the city to background NO_2 concentrations ($\mu g/m^3$) in Copenhagen on distance from the city center. The figure is based on monthly mean measurements at locations with a daily average traffic of 1000 vehicles or fewer, and no streets with more than 5000 vehicles per day within 50 m. Rural background concentrations measured simultaneously were subtracted.

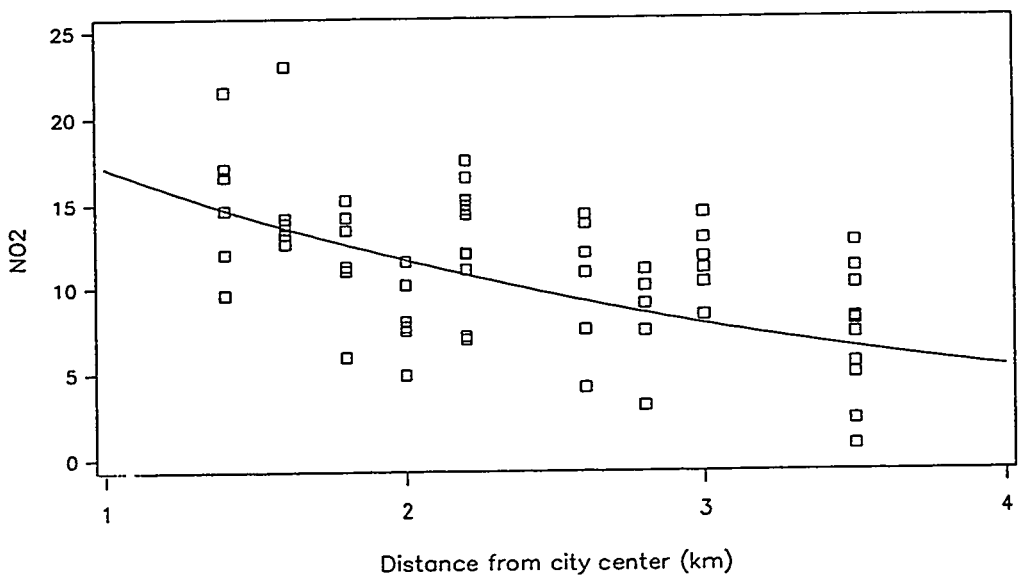
Figure 2.

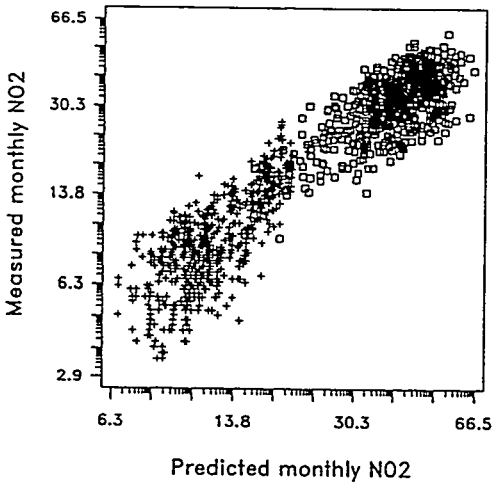
Measured versus predicted monthly (left panel) and half-year (right panel) mean NO_2 concentrations. Data for Copenhagen are indicated by squares and data for rural areas by plus signs. Concentrations are given in $\mu g/m^3$; logarithmic axes. Input data for the OSPM derived from the registration scheme and meteorological information for the same year.

Figure 3.

Measured one-week mean concentrations versus predicted values of NO_2 (left panel) and benzene (right panel). Data for Copenhagen are indicated by squares and data for rural areas by plus signs. Concentrations are given in $\mu g/m^3$; logarithmic axes. Input data for the OSPM derived from the registration scheme and meteorological information for the same year.

F. 1

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Measured monthly NO2
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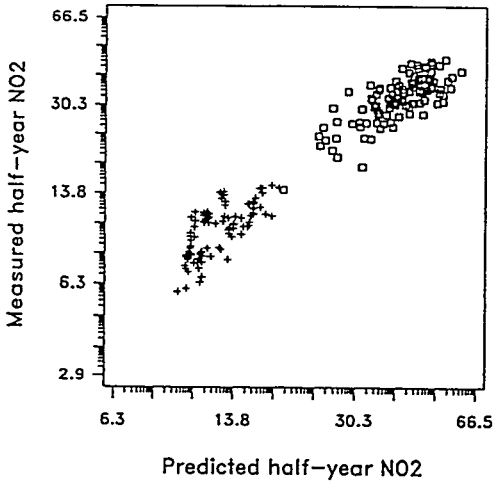
Predicted monthly NO2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2.9
Measured half-year NO2
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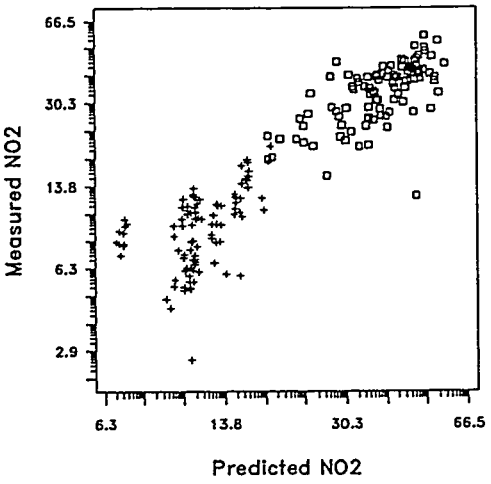
30.3
66.5
Predicted half-year NO2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Measured NO2
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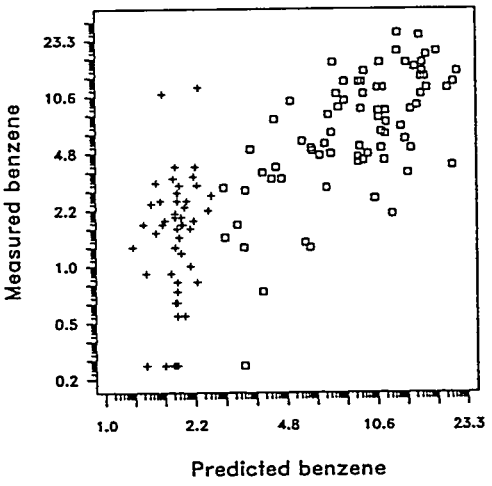
Predicted NO2


23.3
10.6
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1.0
0.5
0.2
Measured benzene
1.0
2.2
4.8
10.6
23.3
Predicted benzene

Table 1. Number of measured locations by street category

Street category[a]	Number of locations	
	Copenhagen	Rural areas
A	0	33
B	3	65
C	3	3
D	83	0
E	14	0

[a] A: single house; B: low, scattered houses; C: street canyon, various building heights; D: street canyon, equal building heights; E: tall buildings on one side of the street and no buildings on the other

Table 2. Pearson's correlation coefficients (r) for associations between measured NO_2 concentrations and concentrations predicted by the OSPM and between measured concentrations and traffic density. The correlation coefficients are based on log-transformed values.

Length of measurement of NO_2 (months)	No. of observations		Method for street configuration	Copenhagen			Rural areas		
	Copenhagen	Rural areas		Model		Traffic density	Model		Traffic density
				Meteorological data from			Meteorological data from		
				Another year	Actual year		Another year	Actual year	
6	103	101	Registration scheme	0.75	0.78	0.66	0.70	0.70	0.16
			Detailed	0.79	0.80				
1	618	606	Registration scheme	0.61	0.66	0.53	0.76	0.76	0.08
			Detailed	0.62	0.68				

Table 3. Pearson's correlation coefficients (r) for associations between measured one-week air pollution concentrations and concentrations predicted by the OSPM and between measured concentrations and traffic density. Simultaneous measurements of benzene and NO_2; in parentheses, calculations based on all measurements. The correlation coefficients are based on log-transformed values.

One-week measurement	No. of observations		Method for street configuration	Copenhagen			Rural areas		
	Copenhagen	Rural areas		Model		Traffic density	Model		Traffic density
				Meteorological data from			Meteorological data from		
				Another year	Actual year		Another year	Actual year	
NO_2	76 (102)	51 (101)	Registration scheme	0.57 (0.62)	0.61 (0.66)	0.43 (0.43)	0.08 (0.48)	0.08 (0.47)	-0.02 (-0.05)
			Detailed	0.60 (0.64)	0.65 (0.68)				
Benzene	76 (77)	51 (51)	Registration scheme	0.62 (0.61)	0.66 (0.66)	0.55 (0.54)	0.15 (0.15)	0.19 (0.19)	0.03 (0.03)
			Detailed	0.68 (0.68)	0.68 (0.68)				

Table 4. Mean concentrations ($\mu g/m^3$) and standard deviations (SD) in model predictions and measurements in Copenhagen and rural areas, respectively. Model predictions were based on the registration scheme and actual meteorological data

Measurement	Copenhagen					Rural areas				
	N	Model		Measurements		N	Model		Measurements	
		Mean	SD	Mean	SD		Mean	SD	Mean	SD
½-year NO_2	103	40.6	8.5	32.6	6.5	101	12.7	2.2	10.2	2.2
1-month NO_2	618	40.6	8.9	32.7	8.3	606	12.7	3.4	10.2	4.2
1-week NO_2	102	39.1	9.5	34.4	9.1	101	12.0	2.7	9.7	3.4
1-week benzene	77	10.1	4.7	8.7	5.8	49[a]	1.88	0.25	1.66	1.1

[a] Two outstandingly high concentrations were excluded. When those observations are included the measured mean concentration is increased to 2.04 $\mu g/m^3$ (SD = 2.2). The model results were affected only marginally