

Environmental research under the Nordic Council of Ministers

# MIL 4

The relative contribution of air pollutants from various sources to man and the environment.

## FINAL REPORT

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## FINAL REPORT - MIL 4

*RELATIVE CONTRIBUTION OF AIR POLLUTANTS FROM  
VARIOUS SOURCES TO MAN AND THE ENVIRONMENT*

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## Preface

THE NORDIC COUNCIL OF MINISTERS DECIDED IN JUNE 1980 TO CARRY OUT SOME OF THE PROJECTS SUGGESTED BY A SPECIALIST GROUP AT A NORDIC SEMINAR ON ENVIRONMENTAL IMPACT OF ENERGY PRODUCTION, AT RØROS IN APRIL 1979. ALL PROJECTS WERE ASSIGNED THE COMMON ABBREVIATION 'MIL' (ENVIRONMENTAL IMPACT OF ENERGY PRODUCTION). THE PROJECT GROUP MIL 4 HAD AS ITS TASK THE QUANTIFICATION OF THE RELATIVE IMPACT OF AIR POLLUTANTS FROM DIFFERENT ENERGY PRODUCTION/CONVERSION PROCESSES, RELATIVE TO OTHER SOURCES, UPON MAN AND THE ENVIRONMENT.

A PROJECT STEERING GROUP WAS APPOINTED, WITH REPRESENTATIVES FROM THE DIFFERENT NORDIC COUNTRIES. THE MEMBERS OF THE PROJECT GROUP FOR MIL 4 SINCE 1980 (UNLESS OTHERWISE NOTED) HAVE BEEN:

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(AIR POLLUTION LABORATORY OF THE NATIONAL AGENCY OF ENVIRONMENTAL PROTECTION) (MSTL)

FINLAND: ALEC ESTLANDER, FINNISH METEOROLOGICAL INSTITUTE (FMI)

NORWAY : GUDMUND GAUPSET, STATENS FORURENSNINGSTILSYN (SFT)  
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FROM 1980: HARALD DOVLAND, SFT

FROM 1981: SIGRID L. BJØRNSTAD, SFT

FROM 1982: LEIF ONGSTAD, SFT

FROM 1984: SIGRID L. BJØRNSTAD, SFT

SWEDEN: LÅRS LINDAU, STATENS NATURVÅRDSVERK (SNV)

(SWEDISH STATE ADMINISTRATION FOR CONSERVATION OF NATURE)

FROM 1984: BJÖRN EJNER, SNV

PROJECT LEADER FOR THE MIL 4 PROJECT WAS BJARNE SIVERTSEN, (NILU) (NORWEGIAN INSTITUTE FOR AIR RESEARCH), WHO ALSO WROTE AND EDITED THIS FINAL REPORT.





## FINAL REPORT - MIL 4

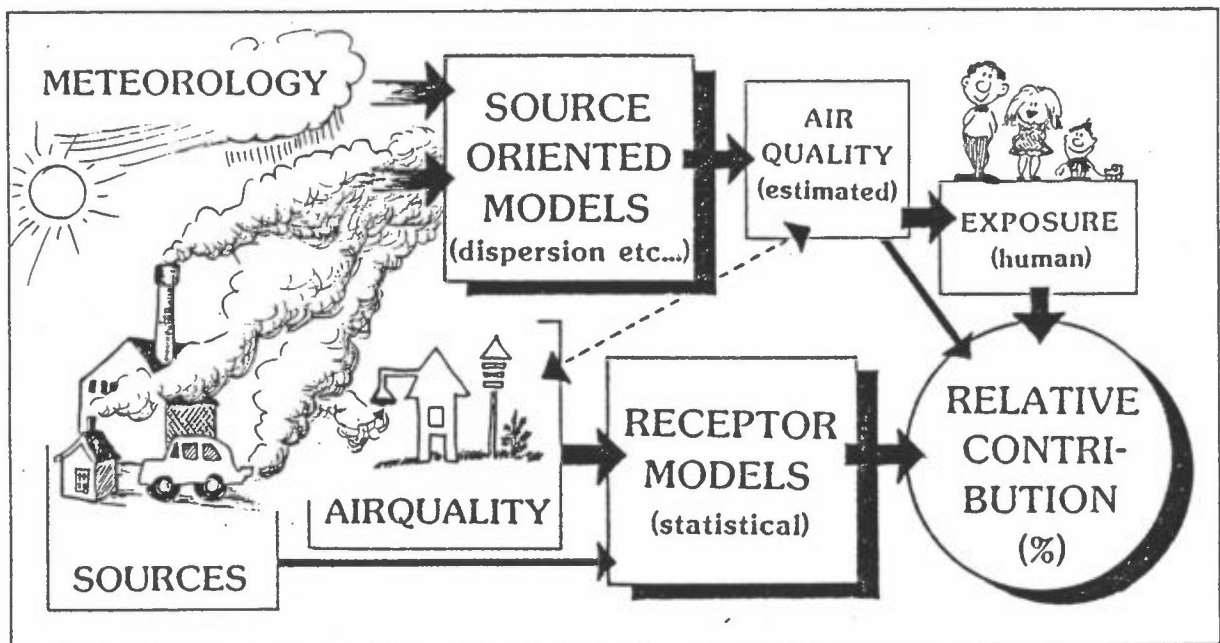
**Summary and conclusions**

THE MAIN OBJECTIVE OF MIL 4 WAS TO QUANTIFY THE ENVIRONMENTAL IMPACT OF AIR POLLUTANTS FROM ENERGY PRODUCTION, RELATIVE TO OTHER AIR POLLUTION SOURCES.

The main conclusions, based upon 15 projects carried out by various institutions in the Nordic countries, may be summarized as follows:

- The SO<sub>2</sub> exposure to the populations of the capital cities of Finland, Norway and Sweden was from 50 to 80% due to energy production (space heating and in Helsinki also electricity production).
- The NO<sub>x</sub> exposure to the population of Helsinki, Oslo and Stockholm was mainly due to traffic emissions (average ~60%). Energy production, regional and long-range transport was sharing most of the remaining contribution of NO<sub>x</sub>.
- The relative contributions to human exposure of toxic metal is very dependent upon the nature of the element:
  - Lead exposure to blood in the population of an area in south-eastern Norway was mainly caused by traffic emissions (~40%) and long-range transport (~26%). Inhalation caused only an average 10-20% of the total exposure.
  - The total exposure to cadmium in the Danish population of Sjælland was due to regional background dry and wet deposition (29%) and the use of fertilizers (27%), both of which caused an accumulation of Cd in soil, uptake by plants and ingestion through foodstuffs. Inhalation represented ~11% of the total exposure, mostly due to cigarette smoking.

- The major contributions to the total suspended particulate matter, measured at various receptor points in central Copenhagen were: traffic (34%), reentrainment of soil dust (26%), and energy production (18%). Outside the city, regional and long-range transport represented ≈40% of the contribution.
- In Oslo the total population exposure to PAH through inhalation was estimated to originate from traffic (24%), energy production, including space heating using wood and coal (28%) and oil (27%), and long-range transport of air pollutants (21%).



The work in MIL 4 was carried out according to two fundamentally different methods:

- 1) Source-oriented models for estimating
  - a) air exposure of the population to  $\text{SO}_2$  and  $\text{NO}_x$
  - b) total exposure through air and food stuffs to lead and cadmium.
- 2) Receptor models for estimating the relative contributions from different sources at receptor points (where air quality measurements were made).

## The models

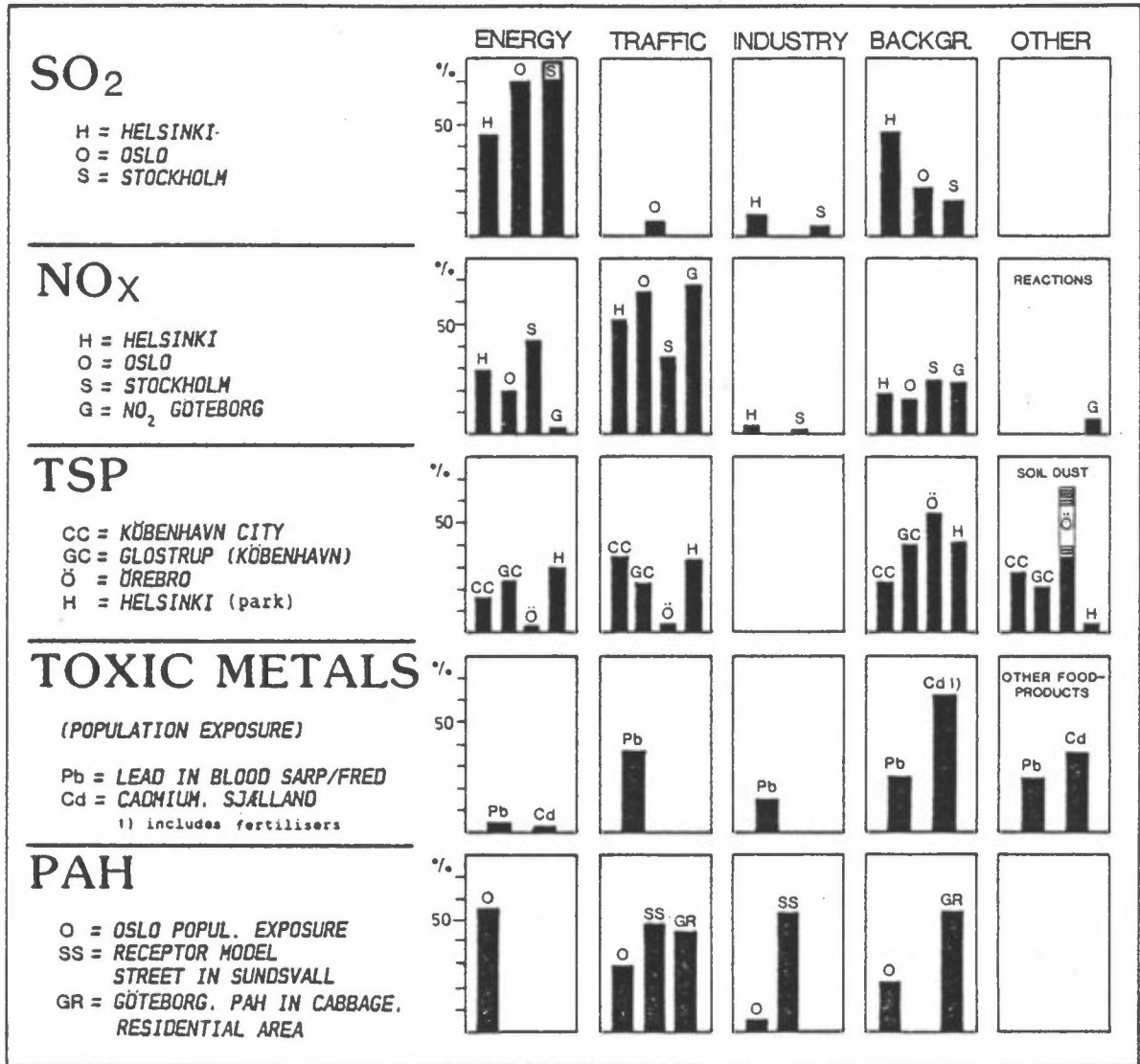
- THE SOURCE-ORIENTED MODELS FOR INHALATION EXPOSURE ( $SO_2$  AND  $NO_x$ ) INCLUDED EMISSION INVENTORIES AND ATMOSPHERIC DISPERSION ESTIMATES. AIR QUALITY WAS COMBINED WITH POPULATION STATISTICS TO GENERATE POPULATION EXPOSURE ESTIMATES. THE MODELS WERE APPLIED SEPARATELY FOR EACH SOURCE CATEGORY TO OBTAIN QUANTITATIVE STATEMENTS ON THE RELATIVE CONTRIBUTIONS TO THE POPULATION EXPOSURE FROM DIFFERENT SOURCES.
  
- FOR ESTIMATES OF THE EXPOSURE TO MAN (WHOLE BODY OR ORGANS) OF TOXIC METALS, INTAKE THROUGH THE CONSUMPTION OF FOODSTUFFS HAD TO BE INCORPORATED. SIMPLE QUASI-STATIONARY COMPARTMENT MODELS WERE ESTABLISHED TO ESTIMATE LEAD AND CADMIUM CONCENTRATIONS IN SOIL AND AGRICULTURAL CROPS, LEADING TO THE CONTAMINATION OF FOODSTUFF.
  
- RECEPTOR MODELS WERE APPLIED TO DIRECTLY ESTIMATE SOURCE CONTRIBUTIONS AT RECEPTOR POINTS FROM STATISTICAL AND MATHEMATICAL ANALYSES. IN CONTRAST TO SOURCE-ORIENTED MODELS, WHICH ESTIMATE AMBIENT AIR QUALITY OR EXPOSURE FROM EMISSIONS, RECEPTOR MODELS START WITH OBSERVED AMBIENT CONCENTRATIONS (PARTICLES, PAH'S...) AT A RECEPTOR AND SEEK TO APPORTION THE OBSERVED CONCENTRATIONS BETWEEN SEVERAL SOURCE TYPES, BASED ON KNOWLEDGE OF THE COMPOSITIONS OF THE SOURCE AND RECEPTOR MATERIALS.

## The projects

| Area                      | Component         | Method   |
|---------------------------|-------------------|--|
| Helsinki                  | $SO_2$ and $NO_x$ | Multiple source dispersion models  |
| Oslo                      | • • •             | Multiple source dispersion models  |
| Stockholm                 | • • •             | Multiple source dispersion models  |
| Göteborg                  | $NO_2$            | Air quality data statistical analysis  |
| Sjælland                  | $Cd^2$            | Population exposure estimates from dispersion/compartiment modeling            |
| Sarpsborg/<br>Fredrikstad | Pb                | Compartment model for population exposure of lead in blood                     |
| Oslo                      | PAH               | Sources, dispersion, population exposure                                       |
| København                 | TSP               | Receptor modeling of 15 elements collected on filter at several stations       |
| Helsinki                  | Elements          | Multi-element analysis of concentrations in moss bags at 164 stations          |
| Ørebro                    | Elements          | Receptor model for 6 elements on filters                                       |
| Southern Norway           | Elements          | Receptor model for 26 elements in live moss                                    |
| Sundsvall                 | PAH               | Receptor models for 30 PAH components at 4 stations around an aluminum smelter |
| Göteborg                  | PAH               | PAH compounds in cabbage   |
| Scandinavia               | Elements          | Analysis of element deposition due to long-range transport to Scandinavia      |
| Urban                     | TSP               | Methodology study of deposition and resuspension of particles in cities.       |



# The results



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## FINAL REPORT - MIL 4

### RELATIVE CONTRIBUTIONS OF AIR POLLUTANTS FROM VARIOUS SOURCES TO MAN AND THE ENVIRONMENT

## 1 Introduction

In June 1980, the Nordic Council of Ministers launched a series of projects to elucidate the effects to the environment of energy production and conversion ("Miljøeffekter ved energiproduksjon" - MIL). One of the projects, MIL 4, managed by the Norwegian Institute for Air Research (NILU), had the aim to quantify the environmental impact of air pollution from energy production, relative to all other sources of air pollution.

MIL 4 consisted of four phases:

1. Development of a project plan, based upon an inventory of ongoing relevant research in the Nordic countries (Sivertsen, 1982a).
2. Development of models and preparation of input data (Sivertsen, 1982b).
3. Testing of proposed computational methods; harmonization of model input data, quantification of the relative source contributions, and presentation of results (Sivertsen, 1983; Sivertsen, 1984a).
4. Final evaluations and reporting.

The entire MIL 4 project was the responsibility of nine different institutions in four Nordic countries. It consisted of 15 sub-projects, divided into three main groups:

- a) Human exposure to the air pollutants  $\text{SO}_2$  and  $\text{NO}_x$ ,
- b) relative contributions of air pollution through inhalation and food consumption to the human exposure of toxic metals and PAH,

- c) receptor modeling for the estimation of the relative contribution from various air pollution source categories.

## 2 Objectives and scope of MIL 4

To evaluate the need for and the extent of abating emissions of air pollutants from energy conversion processes, information is required on the relative contributions to the overall pollution load from all types of sources in the area. Such data will facilitate the adoption of appropriate abatement strategies and the assigning of priorities for environmental protection.

The objective of MIL 4 was to quantify the relative contributions from various types of energy production/conversion facilities in an area, in relation to all other sources (industry, traffic, long-range transport (LRT), agriculture/forestry, etc.). Figure 1 summarizes the objectives and the makeup of MIL 4.

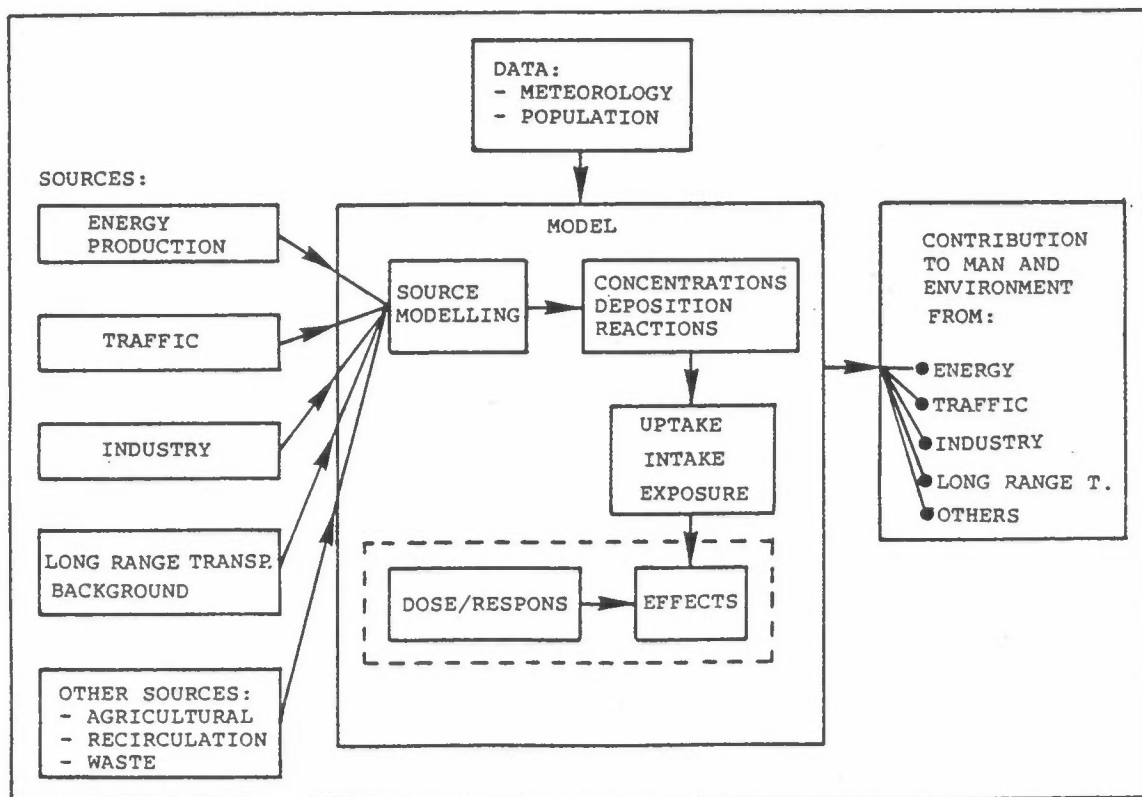


Figure 1: Objectives and elements of the MIL 4 project

Clearly, MIL 4 required expertise in a range of research fields and scientific disciplines related to air pollutant dispersion, deposition, translocation, as well as their effects. It was, therefore, important to limit the scope of MIL 4 by taking into account the realities of:

- limited funds and time available,
- ongoing national and international research, relevant to MIL 4 (i.e., avoiding duplication of effort),
- available information and need for new data,
- the "state-of-art" in the relevant scientific fields.

An important constraint of MIL 4 was the exclusion of impact assessments. The end result of the project was to be the computed estimates of exposures or doses. There has, however, been some contact and exchange of information between MIL 4 and the various Nordic groups working on health effects of air pollution (Clench-Aas, 1983).

### 3 The sub-projects

Due to the limited time and funds, MIL 4 had to a large extent to rely upon available information from ongoing, or already completed relevant studies in the Nordic countries, financed through national funds. It would not have been possible to undertake new measurement programs or develop new models within the scope of MIL 4.

Emission inventories and mathematical dispersion models for several Nordic cities were available for the determination of  $\text{SO}_2$  and  $\text{NO}_x$  concentration fields for some urban areas. In addition, estimates of the number of people in the Oslo metropolitan area, exposed to  $\text{SO}_2$  above certain concentrations, were also available. These then provided a good basis for computation of relative contributions from different source categories to human exposure of  $\text{SO}_2$  and  $\text{NO}_x$ .

In the beginning of MIL 4, it had been planned to use source-oriented dispersion models to calculate man's total exposure to toxic metals from air and food ("compartment" models). Subsequent advances in the development and application of so-called "receptor" models provided an alternative approach. Receptor models make use of chemical characterization of the major air pollution sources and of air quality at the "receptor point" to quantify contributions from the different source categories.

Of the 15 sub-projects within MIL 4, thirteen had as their primary objective to quantify the relative contributions from various source categories, while two sub-projects were dealing with problems related to deposition/resuspension, and contributions from sources outside the Nordic countries.

Table 1 lists all the projects of MIL 4, as well as the person and institution responsible for each sub-project. The locations for selected sub-project areas are indicated in Figure 2.

The results of the projects have been presented in separate reports, referred to in the chapters below describing the different projects.

Table 1: Sub-projects within MIL 4.

| Research topic  | Geographical area   | Responsible person/institution *   | Methodology/component  |
|---|---|--|--|
| 1. Exposure to SO <sub>2</sub> and NO <sub>x</sub>      | a) Helsinki<br>b) Oslo<br>c) Stockholm<br>d) Göteborg                                       | P. Bremer, FMI<br>K.E. Grønskei, NILU<br>S. Laurin, SMHI<br>B. Galle, IVL  | Source-oriented dispersion model<br>.<br>.<br>.<br>.<br>NO <sub>2</sub> analysis, during episodes                |
| 2. Toxic metals, PAH (exposures via air and foodstuffs) | a) Sjælland<br>b) Sarpsborg/<br>Fredrikstad<br>c) Helsinki<br>d) Scandinavia<br>e) Oslo     | O. Edlund, AB Energi-<br>teknikk<br>B. Sivertsen, NILU<br>a. Mäkinen, UiH<br>E. Steinnes, UiT<br>K.E. Grønskei, NILU | Cd exposure model<br>Pb<br>"Moss-bag" technique<br>Deposition of metals<br>PAH dispersion model                  |
| 3. Receptor models                                      | a) Göteborg/<br>Ørebro<br>b) Sundsvall<br>d) Southern Norway<br>e) Göteborg<br>f) København | B. Steen, IVL<br>B. Sivertsen, NILU<br>J. Schaug, NILU<br>E. Brorström-Lunden,<br>IVL<br>F. Palmgren-Jensen,<br>MSTL | V, Cr, Mn, Ni, Pb, Fe<br>PAH (30 compounds)<br>26 elements in mosses<br>PAH in cabbage<br>15 elements on filters |
| 3 c) Deposition and resuspension                        |   | N.O. Jensen, National<br>Lab. Risø   | Particles, mechanisms  |

\*For abbreviations, see Appendix A.

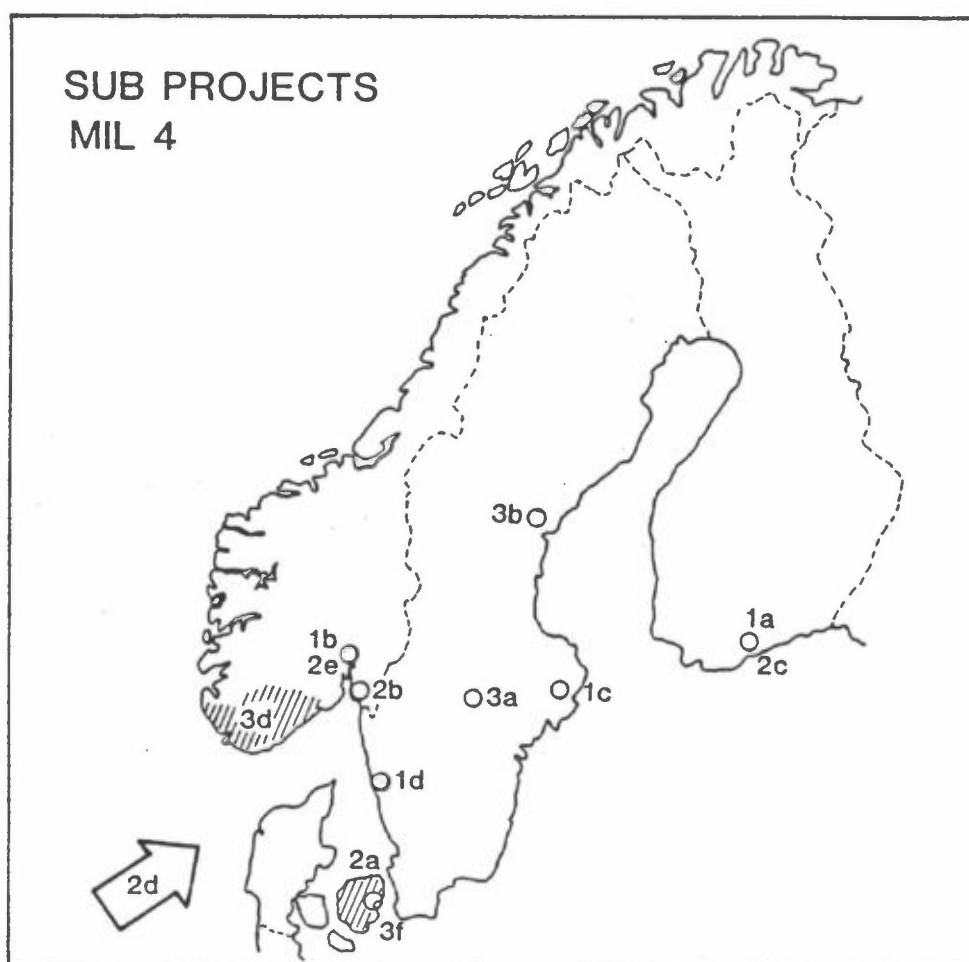


Figure 2: Location of the areas investigated in the various sub-projects of MIL 4.



## 4 Computational methods

To estimate the relative contributions from the different source categories to the overall air pollution load, two different approaches have been used:

- 1 Source-oriented models utilize inputs of information on emissions and meteorological conditions to compute pollutant concentration (or deposition) at a given point.
- 2 Receptor models use input information on the chemical composition of emissions from the various source categories, and of the pollutants collected at the receptor point, to apportion fractional contributions from the sources.

The main differences in the principles, as well as the possible combined use of these two approaches, are illustrated in Figure 3.

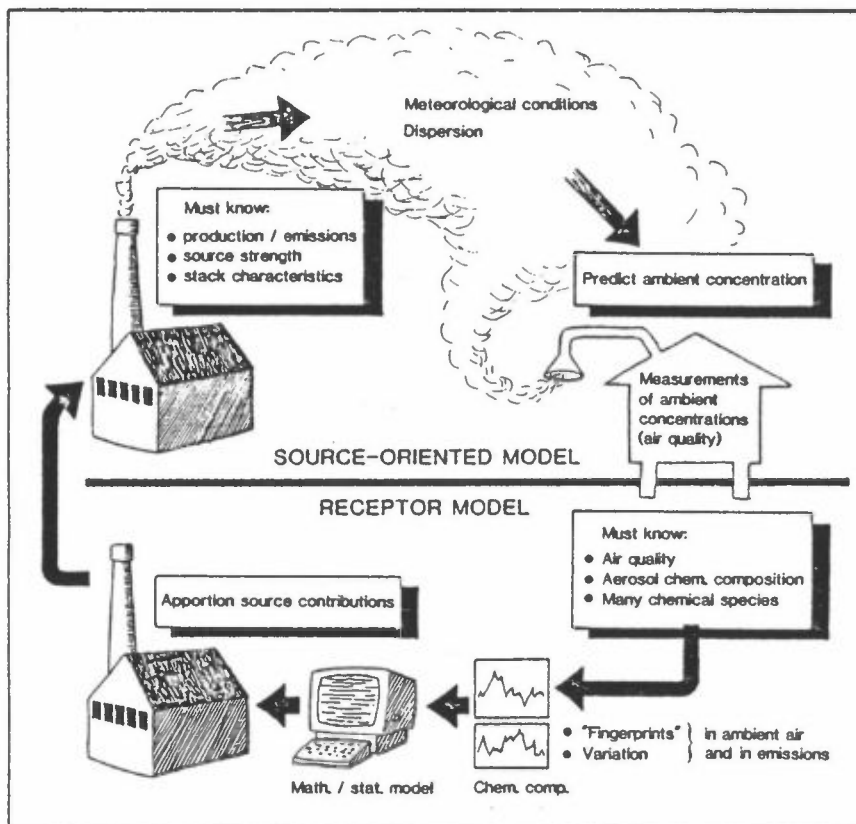


Figure 3: The principal elements of source-oriented and receptor models, and the combined use of these to explain the relative contributions from different sources.

#### 4.1 SOURCE-ORIENTED MODELS

The structure and complexity of source-oriented models depend on factors, such as:

- characteristics of emissions (e.g., emission height, time variation),
- nature of emitted pollutants (e.g., toxicity, half-life),
- pathways of uptake by man/environment,
- size and type of geographical area,
- affected population distributions and activity patterns.

The exposure of man to air pollutants may be estimated by:

- a) combining estimated average air pollutant concentration distributions and the population likely to be exposed to them,
- b) calculating average individual exposure for members of selected population grouping (from information on consumption and activity patterns), and extrapolating to the entire grouping and the population.

In estimating population exposures to  $\text{SO}_2$  and  $\text{NO}_x$  in Nordic cities, the inhalation pathway is considered the most important uptake mode, for which approach (a) above is appropriate. On the other hand, uptake of toxic metals (in a specific organ of the human body) takes largely place via foodstuff consumption, so that individual exposures (as per approach (b) above) must first be estimated.

##### 4.1.1 Dispersion models

Source-oriented dispersion models enable the relating of source emissions to ambient air concentrations, by considering pollutant emission rates, atmospheric dispersion, and possible transformations during the transport.

Ambient air quality must be computed with reasonable accuracy for the various "environments" people may come in contact with during their daily activities:

- in street canyons and near major traffic arteries,
- in off-street urban areas,
- in places directly affected by plumes of large sources (e.g., industrial processes, power plants),
- in long-range transported "background" air.

Models for computing dispersion of air pollutants on various spatial scales in the Nordic countries were already available, and the task for MIL 4 was to devise a way for using these in a combined manner.

The available models included:

- a vehicle exhaust emission model (Boström et al., 1982);
- an urban model for point- and area-sources (with resolution on km-scale) for Stockholm and Helsinki (Bringfelt et al., 1974);
- a multiple-source gaussian model for Oslo (Gram, 1984);
- gaussian point-source models for estimating air pollutant loadings near large point sources (Sivertsen, 1980);
- a numerical model for episodic air pollution (under low wind speed conditions) in Oslo (Grønskei, 1973).

These enabled the apportionment of emission contributions to air quality from energy and heat production, industrial processes, vehicular traffic, and, in some cases, "other area sources".

Population statistics for 1-km<sup>2</sup> grid areas were available based upon:

- the number of people residing in each grid square,
- the number of employees in each grid square,
- traffic density.

In addition, there was information on the living and activity-patterns of Oslo area residents, and the relative distribution of "day" and "night" people in Helsinki (by taking into account the distribution of residences, workers and locations of place of employment). For Stockholm, age distributions of the residents could be used to estimate "average" activity patterns.

#### 4.1.2 Models for exposure to toxic metals

For toxic metals, uptake via foodstuffs is more significant, than atmospheric exposure to such gaseous air pollutants as  $\text{SO}_2$  and  $\text{NO}_x$ . It is, therefore, important to estimate the deposition of toxic metals, translocation in the soil, and eventual uptake by plants and animals. For this, a simple quasi-stationary compartment model was employed. The necessary transfer coefficients, from compartment to compartment (or from medium to medium within a compartment) are usually empirically derived, and thus can be expected to vary from one area to another.

Exposure to Cd on Sjælland, for example, was based upon estimates of intake via inhalation of atmospheric emissions from power plants and refuse incinerators, as well as from food consumption, matter deposited from "background" air (i.e., from LRT), fertilizer applications, and from power plant and refuse disposal emissions.

Individual and total population exposures to Cd from the various source categories on Sjælland were calculated. The spread of Cd in cultivated soil has been studied for two compartments: in an upper tilled layer and a deeper layer (Christensen and Tjell, 1983). Special consideration has also been given to deposition as a function of particle size, and uptakes from air and through the root system have been studied (Edlund and Karlberg, 1984).

Population exposure estimates to Pb in Sarpsborg-Fredrikstad area were based upon estimated average blood lead concentrations of population groups. Each population was split into special population groups according to age, sex, locality, activity patterns, nutritional and smoking habits, etc. Individual exposures for each of these special groups were then first computed.

The modeling areas were also further divided into a number of typical localities or "micro-environments", for which average concentrations due to all or particular source categories had been estimated or measured. The inhalation portion of exposure was computed from available information on activity patterns (i.e., the number of hours spent in a particular micro-environment). Intakes via food for the various groupings were derived from available figures on consumption and the estimated concentrations of the metals in the foodstuffs. Exposures due to inhalation and intakes from various foodstuffs were assumed additive for the individuals in each special population group.

A flow diagram of the procedure for computing individual and population group exposures is shown in Figure 4.

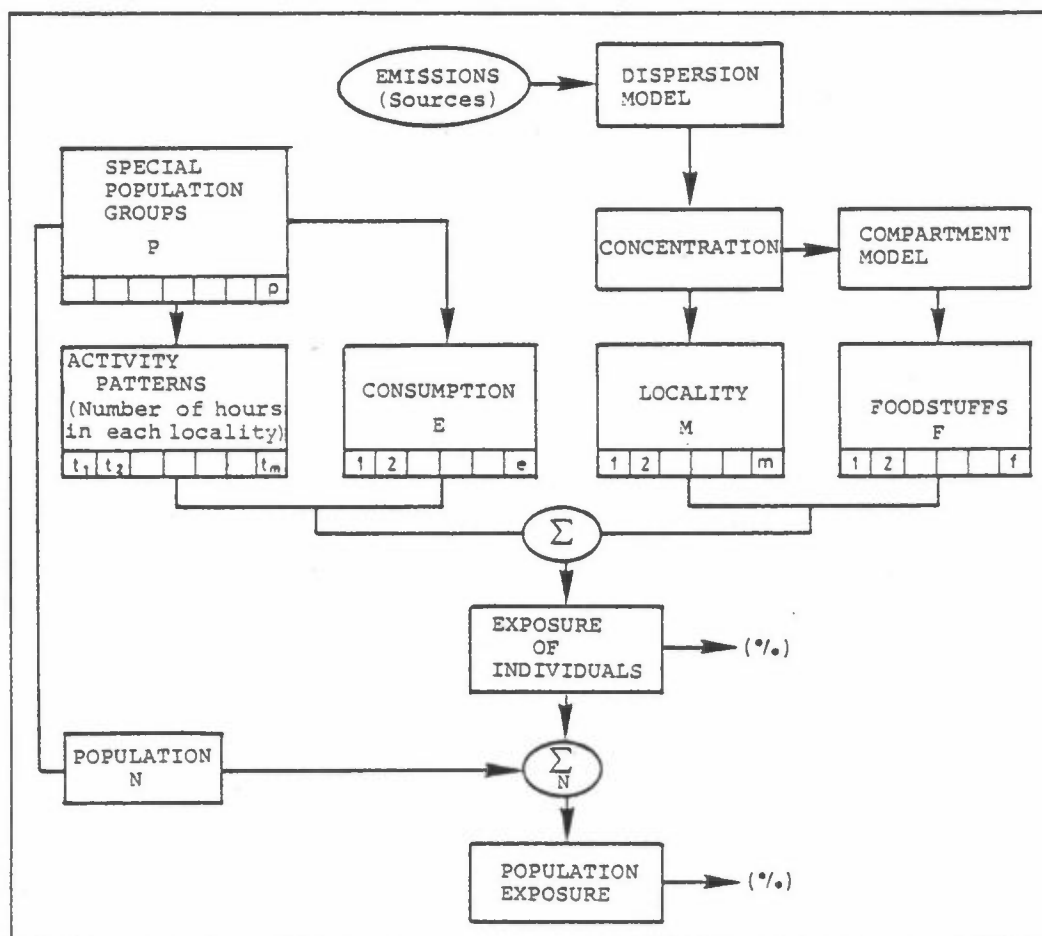


Figure 4: Steps in the calculation of population exposure to toxic metals due to inhalation and food uptake.

#### 4.2 RECEPTOR MODELS

Source-oriented atmospheric dispersion models proceed from known source emissions to predict ambient concentrations or exposures. Receptor models reverse the procedure and, starting with observed concentrations at the "receptor" point, seek to apportion the observed concentrations between several source categories, based on knowledge of the compositions of the source and receptor materials. Receptor models have thus far been mostly applied to the apportionment of aerosols, for which such characteristics, as chemical properties, size distribution, and temporal concentration variations are known (Henry et al., 1984).

Receptor modelling can utilize a number of statistical/mathematical methods to apportion contributions from various source categories:

- principal component analysis
- factor analysis
- chemical mass balance
- cluster analysis
- enrichment factors
- time-series correlation
- multi-variate analysis
- pattern recognition

Generally, a large number of chemical constituents from a reasonable number of sources and ambient samples is required.

The foundation of all receptor models for aerosol source assignment is a simple mass conservation assumption. If a number of sources,  $p$ , exists, and if there is no interaction between their aerosols that causes mass removal or formation, the total airborne particle mass,  $C$ , measured at the receptor, will be a linear sum of the contributions of the individual sources  $S_j$ :

$$C = \sum_{j=1}^p S_j$$

Similarly, the mass concentration of aerosol property  $i$ ,  $C_i$ , will be:

$$C_i = \sum_{j=1}^p a_{ij} S_j$$

where  $a_{ij}$ , is the mass fraction of source contribution  $j$ , possessing property,  $i$ , at the receptor. When the property,  $i$ , is a chemical property, the equation for  $C_i$  represents a chemical mass balance.

The various sub-projects of MIL 4 have applied some, or a combination of the above receptor modelling methods to aerosol particles collected on filters or deposited on vegetation (Schaug, 1984; Brorström-Lunden and Steen, 1984; Kronborg et al., 1984). Use of the chemical mass balance (CMB) method has been most frequently reported. Often, however, the required chemical characterization is not available, or it is specific only for a particular area, or it may be difficult to decide how many source categories should be included. In such cases, factor analysis (FA) and/or principal component analysis (PCA) can supplement CMB. PCA will give an estimate of the number of relevant factors involved, which can be argumented by target transformation factor analysis (TTFA). TTFA compares the composition of emissions from a possible source with each factor explaining a fraction of the variance. By comparing the source composition successively with the factor results (and, in addition, making use of normally available information, e.g., seasonal variations), all sources of any importance can be identified (Schaug, 1983).

## 5 Inhalation exposure of SO<sub>2</sub> and NO<sub>x</sub>

Source-oriented dispersion models, in combination with relevant population data, were used to calculate exposures to sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>) in Helsinki, Stockholm and Oslo. These 3 capital cities were chosen for the modeling because of the availability of appropriate models and input data from earlier studies. Table 2 lists the populations, areas, and emission estimates used in the model computations.

Table 2: Emissions of SO<sub>2</sub> and NO<sub>x</sub> in three Nordic capital cities.

| City      | Model population | Model area km <sup>2</sup> | Winter half-year emission |   |
|-----------|------------------|----------------------------|---------------------------|---|
|           |                  |                            | SO <sub>2</sub> , tonnes  | NO <sub>x</sub> (as NO <sub>2</sub> ), tonnes |
| Helsinki  | 756 900*         | 1000                       | 24 000                    | 7000  |
| Oslo      | 450 400          | 400                        | 3 283                     | 3220  |
| Stockholm | 1 180 000        | 872                        | 24 350                    | 9650  |

\* Helsinki and surrounding areas.

The emission figures for the model inputs were derived from;

- information on oil consumption, sulphur content of oil, and emission factors for NO<sub>x</sub>,
- data on traffic density in each km<sup>2</sup> and emission factors for various types of motor vehicles

The following emission factors for NO<sub>x</sub> (as NO<sub>2</sub>) were applied in the different model computations:

|            |                |                            |
|------------|----------------|----------------------------|
| Helsinki:  | heavy fuel oil | 5.0 kg/m <sup>3</sup> oil  |
|            | light fuel oil | 2.5 " "                    |
|            | anthracite     | 7.8 " coal                 |
| Oslo:      | heavy fuel oil | 5.1 kg/m <sup>3</sup> oil  |
|            | light fuel oil | 2.5 " "                    |
| Stockholm: | point-sources  | 6.47 kg/m <sup>3</sup> oil |
|            | area-sources   | 3.75 " "                   |

Emission factors for the various types of motor vehicles are listed in Table 3.

Table 3: NO<sub>x</sub> emission factors for vehicular traffic, NO<sub>x</sub>/vehicle km (NO<sub>x</sub> as NO<sub>2</sub>)

| Vehicle type           | Helsinki | Oslo | Stockholm |
|------------------------|----------|------|-----------|
| Gasoline-fueled cars   | 2.0      | 2.0  | 2.4       |
| Diesel: Passenger cars | -        | -    | 1.0       |
| Light trucks           | -        | -    | 7.5       |
| Heavy trucks           | -        | -    | 20.0      |
| All diesel vehicles    | 5.0      | 8.2  | -         |



The relative proportions of emissions from the different source categories will vary from city to city. For example, Helsinki has 13 and Stockholm 14 heating/power plants with capacity in excess of 30 MW, while in Oslo there are only 10 plants with greater than 5 MW capacity (none over 30 MW). Centralized space heating is common in downtown Stockholm, while buildings and residences in Oslo rely mostly on smaller and individual heating units. The effect of this is brought out by the percentage emission rates given in Table 4.

Table 4: Share (in %) of SO<sub>2</sub> and NO<sub>x</sub> emissions from the different source categories to total emissions in the three Nordic capital cities during the winter half-year.

| Source category         |                          | SO <sub>2</sub> emissions (%) |      |         | NO <sub>x</sub> (as NO <sub>2</sub> ) (%) |      |                  |
|-------------------------|--------------------------|-------------------------------|------|---------|---|------|------------------|
|                         |                          | Hels.                         | Oslo | Stockh. | Hels.                                     | Oslo | Stockh.          |
| Power+<br>heat          | < 5 MW +<br>area sources | 11                            | 81   | 46      | 8   | 21   | 36               |
|                         | 5-30 MW                  | 9                             | 11   | 13      | 3   | 6    | 11               |
|                         | > 30 MW                  | 71                            | 0    | 33      | 39  | 0    | 28               |
| Industrial<br>processes |                          | 7                             | 3    | 8       | 3   | 1    | 7                |
| Traffic                 |                          | 0                             | 5    | -       | 47  | 72   | 18 <sup>1)</sup> |

1) underestimated

The percentages in the table also confirm that a substantial portion of NO<sub>x</sub> emissions is due to vehicular traffic. The low traffic share in Stockholm is due to missing emission data for traffic in suburban Stockholm. Consequently, the shares of other sources are likely to be overestimated.

The emission figures for the winter half-year are dominated by space heating and power plants. During the summer, however, 80 - 90 % of  $\text{NO}_x$  comes from motor vehicles, which on an annual basis contribute only ca. 60% of the total  $\text{NO}_x$  emissions.

Certain assumptions had to be made about pollutant concentrations in the "background" air. Most of this "background" is due to long-range transport (LRT) from outside the modelling areas, but also regional influences (on a 100 km scale) are included. The values shown in Table 5 are largely based on air quality measurements in the earlier OECD and the EMEP programs (EMEP, 1984).

Table 5: Estimated "background" concentrations,  $\mu\text{g}/\text{m}^3$ , of  $\text{SO}_2$  and  $\text{NO}_x$  (as  $\text{NO}_2$ ).

| City      | $\text{SO}_2$ | $\text{NO}_x$ |
|-----------|---------------|---------------|
| Helsinki  | 14            | 3             |
| Oslo      | 10            | 10            |
| Stockholm | 8             | 8             |

$\text{SO}_2$  and  $\text{NO}_x$  dispersion was modeled for each source category, and concentration maps for winter half-year means and maximum 24-hr average concentrations were prepared. In Appendix B typical calculated winter half-year means are presented for the three cities. The calculated exposures were then derived from concentration and population distributions according to:

- Helsinki: daytime and night-time populations, and a "mobile" portion distributed over all grids,
- Oslo: ambient outdoor concentrations near workplaces in the city, residences, and in public transport,
- Stockholm: stay-home portion and "mobile" portion (who spend 40% or more of the time in commuting and at work) of population.

The relative contributions of the different source categories to the mean ambient concentrations vary considerably, depending on the characteristics of a given location in the model area. For that reason, the locations were classified into 4 or 5 sub-localities, as given in Table 6.

Table 6: Sub-localities of the three Nordic capital cities for estimating relative contributions to mean concentrations of  $\text{SO}_2$  and  $\text{NO}_x$  from various source categories.

| Location                         | Helsinki  | Oslo  | Stockholm  |
|----------------------------------|---|---|--|
| 1 Downtown center                | High population density, traffic, medium and large power plants | Adm. center 6 km <sup>2</sup> , max. $\text{SO}_2$ concentration <sup>2</sup> | Södermalm, densely populated, max. $\text{SO}_2$ conc. |
| 2 Near downtown center           | Bortre Tälö, residential, rush traffic                          | Lysaker-center, traffic, max. $\text{NO}_x$ concentration                     | Adm. center, offices, business traffic                 |
| 3 Suburban                       | Baggböle, traffic arteries, only a few stationary sources       | Groruddalen, highrise housing+ small residences, traffic arteries             | Farsta, 10 km from city center                         |
| 4 Residential, industrial        | Vanda, residential and industrial area                          | Nydalen, residences, industry, businesses                                     | Sollenstuna, 15 km from city center                    |
| 5 Residential, outskirts of town | Gröndal, relatively clean, small point-sources                  | -   | Gustavsberg, suburban, 20 km from city center          |

A judicious choice of sub-localities allows a reasonable allocation of sources for the models.

## 5.1 SO<sub>2</sub> IN THE CITIES

Computations of population exposure to SO<sub>2</sub> were performed for the three Nordic capital cities Helsinki, Oslo and Stockholm and, as shown below, are mainly caused by emissions from heat and power production (50-80%).

### 5.1.1 Air quality - SO<sub>2</sub>

In the central sections of the cities, the large number of smaller heating units and/or area sources (e.g., apartment heating) contribute significantly towards total SO<sub>2</sub> concentrations, as seen in Table 7.

Table 7: Relative % contributions to SO<sub>2</sub> concentrations from various sources in downtown sections of the cities during the winter half-year.

| Source categories | Helsinki | Oslo | Stockholm |
|-------------------|----------|------|-----------|
| Heat/power:       |          |      |           |
| < 5 MW+area       | 16       | 76   | 75        |
| 5 - 30 MW         | 7        | 3    | 9         |
| > 30 MW           | 28       | 0    | 6         |
| Industrial        | 7        | 0    | 1         |
| Traffic           | -        | 5    | -         |
| Background        | 42       | 16   | 9         |

The exception is Helsinki, whose large centrally located powerplants account for 28% of the mean concentrations of SO<sub>2</sub> in downtown areas in winter. In addition, the relatively high "background" concentration of 14 µg/m<sup>3</sup> accounts for as much as 42% of the SO<sub>2</sub> contribution. This contrasts with the ca. 75% contribution of the many small area sources in central Oslo and Stockholm.

Towards the outskirts of the cities, the relative significance of SO<sub>2</sub> contributions from the various source categories vary greatly, as seen in Table 8.

Table 8: Relative % contributions to mean SO<sub>2</sub> concentrations from sources in selected localities (cf. Table 6) outside the 3 city centers.

| Source categories | Helsinki | Oslo  | Stockholm |
|-------------------|----------|-------|-----------|
| Heat/power:       |          |       |           |
| < 5 MW + area     | 10-30    | 63-72 | 35-60     |
| 5 - 30 MW         | 4-16     | 3-4   | 8-10      |
| > 30 MW           | 10-20    | 0     | 7-12      |
| Industrial        | 5-27     | 2-4   | 3-5       |
| Traffic           | -        | 5-7   | -         |
| Background        | 40-50    | 16-28 | 20-40     |

Here, a substantial contribution (20 - 50 %) to SO<sub>2</sub> concentrations apparently comes from regional background concentrations, including long-range transport. Furthermore, in Oslo and Stockholm the small heating units account for half or more of SO<sub>2</sub> mean concentrations in the winter half-year. Industrial emissions of SO<sub>2</sub> appear to be significant (27%) only in the Vanda section of Helsinki. In all other city locations, industry contributes only from 2 to 5 % of the SO<sub>2</sub>.

#### 5.1.2 Population exposure to SO<sub>2</sub>

Population exposure to SO<sub>2</sub> from the various source categories was derived from SO<sub>2</sub> concentrations and population distributions. Figure 5 shows the estimated number of people in the three cities exposed to the mean concentrations of SO<sub>2</sub> (along the abscissa) from all sources, including background.

Large differences between the three cities are obvious. For example, in Helsinki very few persons are exposed to mean SO<sub>2</sub> concentrations higher than ~50 µg SO<sub>2</sub>/m<sup>3</sup> during daytime in winter, while in Oslo and Stockholm around 130000 and 330000 people, respectively, are experiencing winter mean concentrations in excess of 60 µg SO<sub>2</sub>/m<sup>3</sup>. The differences appearing in Figure 5 are mainly due to actual differences in emissions, meteorology and regional background, but might also be caused by differences in the spatial resolution of the different models. Intercomparisons between the cities should therefore be made with care.

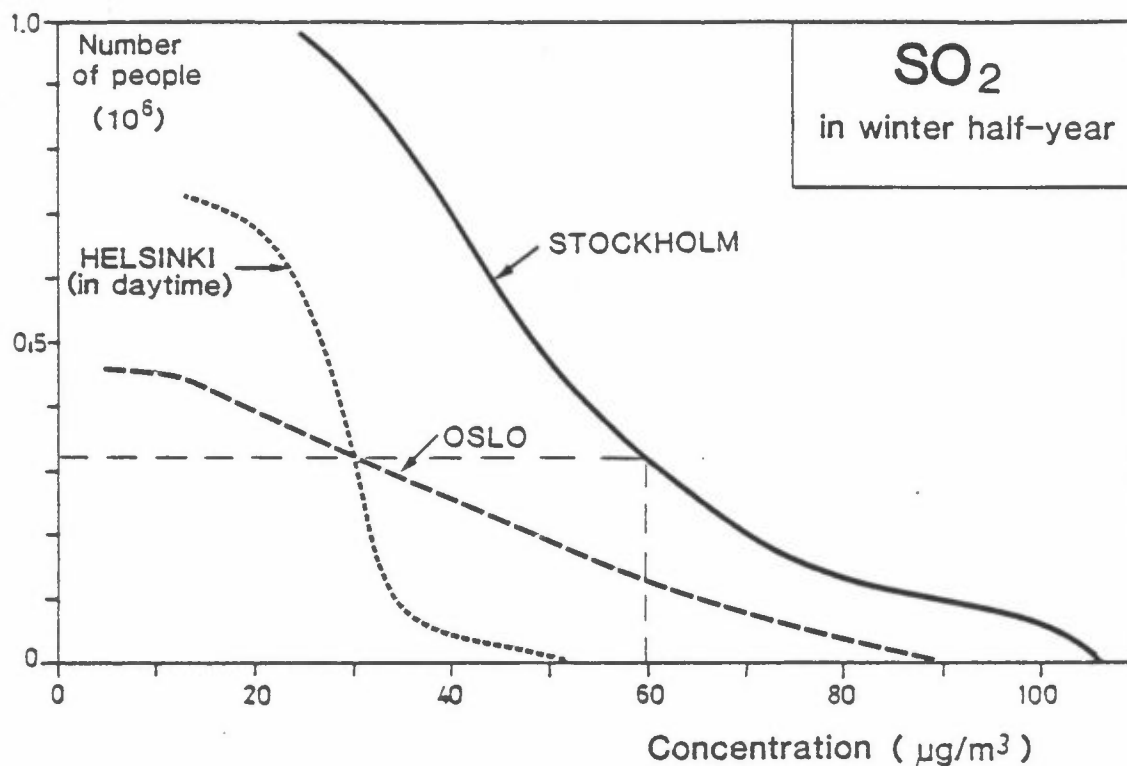


Figure 5: Estimated number of people subjected to mean winter half-year SO<sub>2</sub> concentrations (given on the abscissa) from all SO<sub>2</sub> sources, including background.

The statistics derived from calculations as presented in Figure 5, are summarized in Table 9.

Table 9: Estimated number of people exposed to winter half-year means of over 30 and 60 µg SO<sub>2</sub>/m<sup>3</sup>, and the corresponding % contributions from the various source categories to the 30 µg SO<sub>2</sub>/m<sup>3</sup> mean concentration.

| City      | Number of people exposed to concentrations: |                        | % contribution of sources to 30 µg/m <sup>3</sup> from source category: |       |            |
|-----------|---|------------------------|---|-------|------------|
|           | > 30 µg/m <sup>3</sup>                      | > 60 µg/m <sup>3</sup> | < 5 MW + area   | > 5MW | Background |
| Helsinki  | 360 000                                     | 0                      | 6   | 10    | 14         |
| Oslo      | 300 000                                     | 130 000                | 18  | 2     | 10         |
| Stockholm | 930 000                                     | 330 000                | 17  | 5     | 8          |

It is clear that the elevated emission from the large power plants in Helsinki subject considerably fewer people to high ground-level  $\text{SO}_2$  concentrations in winter, than is the case in Oslo and Stockholm. In the latter two cities, low-level emissions from the numerous small sources result in high ground-level concentrations of  $\text{SO}_2$ , particularly during temperature inversion situations.

Curves as presented in Figure 5 may be estimated for each of the source categories and used to estimate population exposure to  $\text{SO}_2$ . The estimated relative % contributions from the different sources to the population exposure (in  $\mu\text{g}\cdot\text{man}/\text{winter half-year}$ ) are plotted in Figure 6.

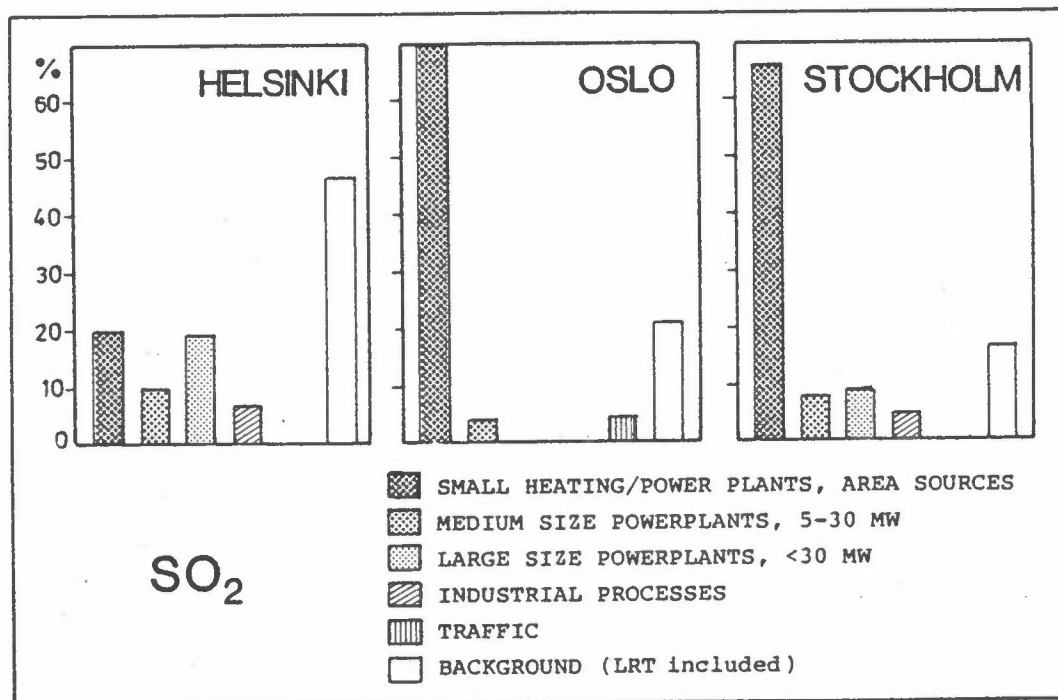


Figure 6: Relative contributions (%) to population exposure by  $\text{SO}_2$  from the various source categories in winter half-year in the three Nordic capital cities.

According to Figure 6, the small heating/power units and residential space heating (area sources) are responsible for up to 70% of the  $\text{SO}_2$  dose to the populations of Oslo and Stockholm. In Helsinki, however, the contributions from the small sources and the large power plants are about even (20% each).

## 5.2 NO<sub>x</sub> IN THE CITIES

The estimated NO<sub>x</sub> exposures to the population of Helsinki, Oslo and Stockholm, as shown below, are mainly due to traffic emissions (~60%). Heat and power production and long-range transport also contribute to the NO<sub>x</sub> exposure.

Although nitrogen in several different oxidation states is found in urban atmospheres, nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>) are the main ones. NO<sub>2</sub> is known to have human health consequences, with inhalation as the predominant uptake. Exposures to NO<sub>2</sub> can be estimated by the same procedure, as described for SO<sub>2</sub> in the earlier sections.

One of the goals of MIL 4 was to obtain reliable NO<sub>2</sub> concentration data. It soon became apparent, however, that the available urban models were not adequate for this task. The reaction mechanisms for the known atmospheric conversion of NO to NO<sub>2</sub> were not well enough understood or documented for simple parameterization. This problem has been examined in a separate sub-project (Galle, 1984) and is reported in Chapter 8.

For purposes of exposure estimation, urban concentrations of total oxides of nitrogen, NO<sub>x</sub>, (assumed to be NO + NO<sub>2</sub>) were used, but expressed as NO<sub>2</sub>.

### 5.2.1 Air quality - NO<sub>x</sub>

NO<sub>x</sub> emissions from motor vehicles result in by far the most significant portion of mean NO<sub>x</sub> concentrations (52 - 69%) in the downtown sections of Helsinki, Oslo and Stockholm, as shown in Table 10.



Table 10: Relative contributions (%) from various source categories to mean  $\text{NO}_x$  concentrations in downtown sections in winter half-year.

| Source categories | Helsinki | Oslo | Stockholm |
|-------------------|----------|------|-----------|
| Heat/power:       |          |      |           |
| < 5 MW + area     | 5        | 18   | 30        |
| 5 - 30 MW         | 2        | 2    | 3         |
| > 30 MW           | 13       | 0    | 4         |
| Industrial        | 2        | 0    | 0         |
| Traffic           | 67       | 69   | 52        |
| Background        | 11       | 11   | 11        |

In the outskirts of the three cities, the relative contributions again vary considerably. The main reasons for this are real differences in geographical distribution of sources, and the lack of information for some localities. Traffic data for all localities outside downtown sections of Stockholm were not available. Table 11 displays the variations in the relative source contributions for the suburban/outskirts localities of the three cities.

Table 11: Relative contribution (%) to mean  $\text{NO}_x$  (as  $\text{NO}_2$ ) concentrations from sources in selected localities (see Table 6) in the outskirts of the three cities.

| Source categories | Helsinki | Oslo    | Stockholm |
|-------------------|----------|---------|-----------|
| Heat/power:       |          |         |           |
| < 5 MW + area     | 7 - 9    | 12 - 17 | 20 - 38   |
| 5 - 30 MW         | 2 - 4    | 1 - 2   | 6 - 8     |
| > 30 MW           | 10 - 17  | 0       | 6 - 8     |
| Industrial        | 1 - 10   | 0       | 6 - 8     |
| Traffic           | 41 - 59  | 69 - 76 | 7 - 14 *) |
| Background        | 13 - 37  | 9 - 16  | 38 - 57   |

\*) underestimated in suburbs

In the suburbs of Helsinki and Oslo, vehicular traffic accounts for 41-76 % of  $\text{NO}_x$  in the winter half-year. The regional background (13-37%) and large power plants (10-17%) in Helsinki rank next in importance, while in Oslo the small area sources and "background" add to the total about 9 - 17% each.

In the suburbs of Stockholm, where traffic emissions were underestimated, the regional background of  $\text{NO}_x$  was estimated to be most important (38-57%). Small sources rank next in importance.

### 5.2.2 Population exposure to $\text{NO}_x$

Figure 7 shows the number of people exposed to mean  $\text{NO}_x$  (as  $\text{NO}_2$ ) concentrations (given along the abscissa) in the three cities in winter half-year.

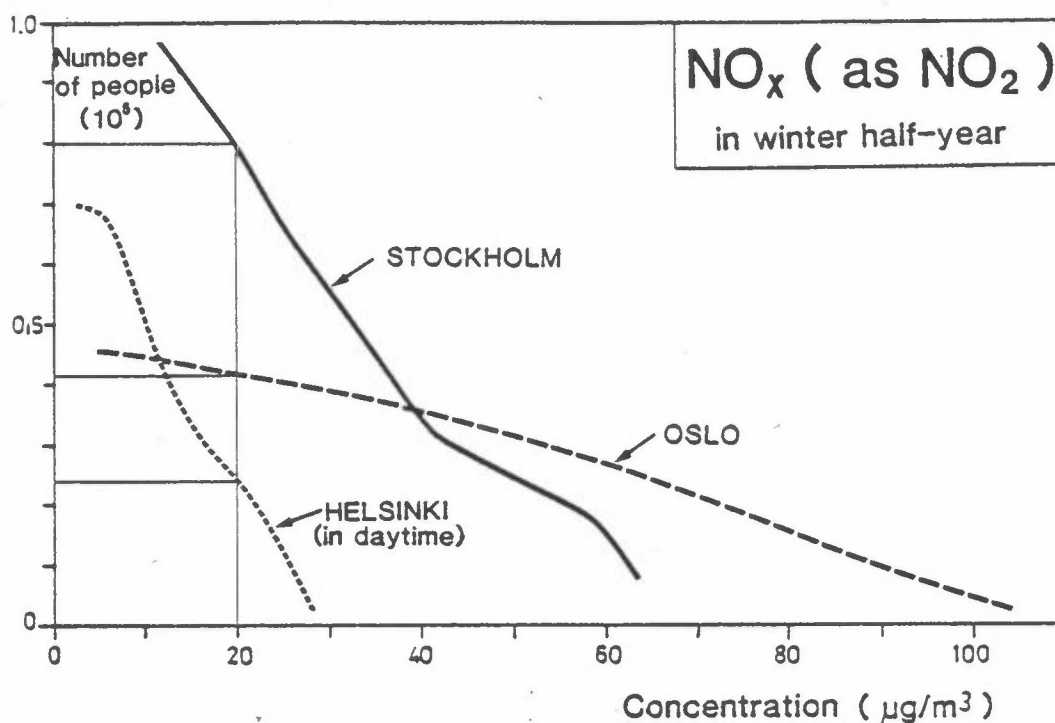


Figure 7: Estimated number of people subjected to the mean winter  $\text{NO}_x$  (as  $\text{NO}_2$ ) concentrations (given on the abscissa) from all  $\text{NO}_x$  sources, including regional background and LRT.

As was the case with  $\text{SO}_2$ , there are again large differences in the population exposures for the three cities. Only the people in Oslo are subjected to mean concentrations above  $70 \mu\text{g NO}_x/\text{m}^3$ . On the other hand, more people in Stockholm experience mean concentrations of  $\sim 30 \mu\text{g NO}_x/\text{m}^3$ , than in Oslo. Although the differences could be real, different ways of modeling vehicular emissions, and dispersion in the streets and along roads in the three cities are probably partly responsible. These type of problems can only be

resolved through continued cooperation and coordination of Nordic modeling efforts in the future.

Table 12 gives the populations exposed in wintertime to mean concentrations greater than 20 and 60  $\mu\text{g NO}_x/\text{m}^3$  in the three cities.

Table 12: Estimated number of people exposed to winter half-year means of over 20 and 60  $\mu\text{g NO}_x/\text{m}^3$ .

| City      | Number of people exposed to concentrations: |                               |
|-----------|---|-------------------------------|
|           | > 20 $\mu\text{g}/\text{m}^3$               | > 60 $\mu\text{g}/\text{m}^3$ |
| Helsinki  | 240 000                                     | 0                             |
| Oslo      | 410 000                                     | 260 000                       |
| Stockholm | 800 000                                     | 150 000                       |

The relative % contributions from the sources to the population doses (in  $\mu\text{g}\cdot\text{man}/\text{winter half-year}$ ) are plotted in Figure 8.

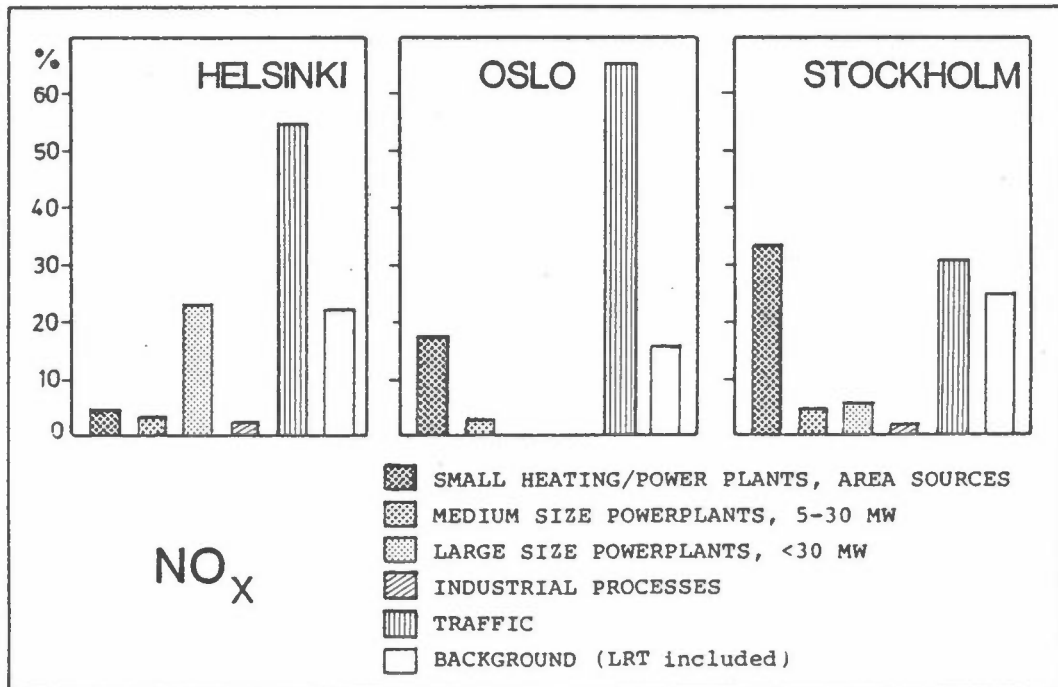


Figure 8: Relative contributions (%) to population exposure by  $\text{NO}_x$  from the various source categories in winter half-year in the three Nordic capital cities.

The total population exposure to  $\text{NO}_x$  is dominated by emissions from motor vehicles, amounting to 55 - 65% of the exposures for Oslo and Helsinki. In Stockholm, the traffic contribution is underestimated (due to lack of suburban traffic data), but nevertheless accounts for more than 30% of the total exposure to  $\text{NO}_x$ .

"Background" accounts for about 20% of the mean  $\text{NO}_x$  concentrations in winter half-year. The relative importance of the small area sources is about the same in Oslo and Stockholm. All Helsinki's heating/power facilities contribute ca. 30%, of which the large power plants alone account for ca. 23 %.

An estimate of daytime exposure during situations with poor dispersion and high emissions (using maximum 24-hour and estimated maximum hourly means) suggests that traffic and small area sources will contribute more and LRT less than indicated in Figure 8 under such conditions.

### 5.3 APPLICATIONS, LIMITATIONS AND UNCERTAINTIES

#### 5.3.1 Applications

The calculated exposures to  $\text{SO}_2$  and  $\text{NO}_x$  point clearly to those sources, which are the most significant for the inhalation intake route. These results can then enable the selection of the most effective abatement strategies to reduce the loads, and thus effects, of these air pollutants. A flow diagram for such a decision process is illustrated in Figure 9.

This type of modelling approach has been earlier applied to evaluate the impacts of different strategies for the abatement of air pollution from energy production in several Nordic towns (Gotaas and Gram, 1984; Olausson et al., 1983).

MIL 4 has extended the procedure by considering all the important source categories separately, by linking air quality estimates and population distributions to estimate the resulting exposures.

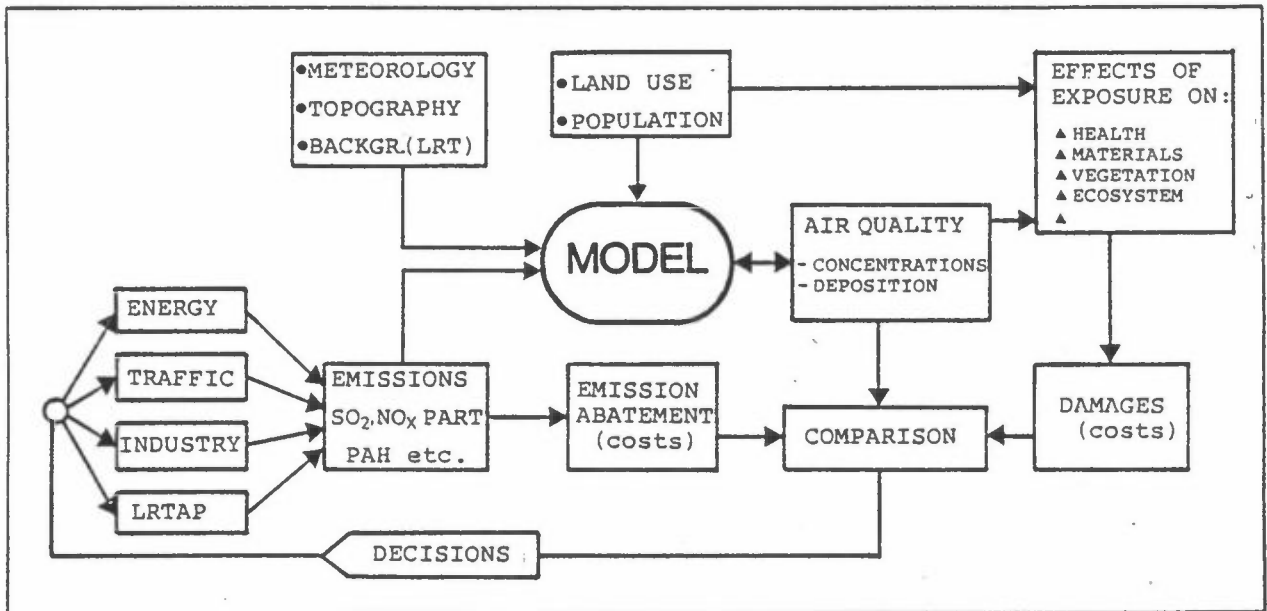


Figure 9: Flow diagram of a procedure for reducing pollutant load, when relative contributions from various source categories to exposures are known.

To give a further illustration of the choosing of a most effective abatement strategy, Table 13 lists numerical ratios, which relate ambient air concentrations to source emissions for the different source categories in selected localities.

A large ratio means that relatively small emission rates can result in poor air quality (i.e., high pollutant concentrations). Atmospheric dilution and natural ventilation for these sources are limited and even the smallest change in source emissions/characteristics might result in a considerably improved air quality.

Table 13 should only be read horizontally as the  $K_i$  ratios were estimated somewhat differently for the different cities. Comparisons of  $K_i$  factors between cities are not relevant.

Table 13: Examples of air quality/source ratios,  $K_i$ , for selected localities in Helsinki, Oslo and Stockholm.  $K_i = C/Q$ , where  $C$  = air quality,  $Q$  = emissions. (Unit for  $K_i = 10^6 \text{ sm}^{-3}$ ).

|               |         | SOURCE CATEGORIES |         |        |                      |         |       |
|---------------|---------|-------------------|---------|--------|----------------------|---------|-------|
|               |         | Heat/power        |         |        | Industrial processes | Traffic |       |
|               |         | < 5 MW + area     | 5-30 MW | >30 MW |                      |         |       |
| $\text{SO}_2$ | City    | Helsinki          | 24-46   | 22     | 9                    | 28      | -     |
|               |         | Oslo              | 11      | 3      | -                    | -       | 19    |
|               |         | Stockholm         | 58      | 11     | 4                    | 4       | -     |
|               | Suburbs | Helsinki          | 20-50   | 10-18  | 5                    | 11-80   | -     |
|               |         | Oslo              | 10      | -      | -                    | -       | 47    |
|               |         | Stockholm         | 68      | 20-25  | 6-11                 | 11-20   | -     |
| $\text{NO}_x$ | City    | Helsinki          | 22-55   | 27     | 23                   | 33      | 92    |
|               |         | Oslo              | 10      | 4      | -                    | -       | 26    |
|               |         | Stockholm         | 68      | 15     | 6                    | -       | 113   |
|               | Suburbs | Helsinki          | 20-44   | 9-18   | 7                    | 22      | 16-31 |
|               |         | Oslo              | 21-69   | -      | -                    | -       | 44-54 |
|               |         | Stockholm         | 30-57   | 30-40  | 12-16                | -       | 20-43 |

The  $K_i$  ratios, estimated in Table 13, indicate that, especially in the suburbs, substantial improvements in air quality could be achieved by reducing  $\text{SO}_2$  emissions from small heating units. In one suburban area of Helsinki, industrial emissions of  $\text{SO}_2$  should also be reduced to improve the air quality.

For  $\text{NO}_x$  the best abatement strategy would be to reduce  $\text{NO}_x$  emissions from traffic, especially in the city center areas. In the suburbs, reductions in  $\text{NO}_x$  emissions from small area sources would also improve air quality in some of the areas.

### 5.3.2 Limitations and uncertainties

The limitations and uncertainties of the  $\text{SO}_2$  and  $\text{NO}_x$  exposure modeling procedure depend largely on the quality of input data on emissions, dis-

persion, and demographic data, as well as on the structure and refinements of the model itself.

It can be pointed out that for all cities uncertainties in  $\text{NO}_x$  emission estimates are mainly due to shortcomings in emission factors and traffic data.  $\text{NO}_2$  transformation modeling was felt inadequate. The "urban dispersion models" were not suited for estimating sub-grid concentrations (in and along streets and roads).

A common shortcoming of the data inputs for all three cities was also the lack of information on  $\text{SO}_2$  and  $\text{NO}_x$  concentration distributions indoors. Most people spend the greater part of time indoors, and the outdoor concentrations, used in the modeling, may not adequately represent indoor exposures. A study of indoor - outdoor air quality relationships is necessary to improve the quality of future exposure modeling efforts.

Further, the definitions of the industrial source category for the three cities were not consistent, and in most cases included oil consumption data only.

The modeling results for the three Nordic cities individually point to the following sources of uncertainties:

- Helsinki: - the mobility/activity patterns of the population are only very rough estimates;  
- the out-lying sections of the modeling area are covered by a coarser grid than downtown.
- Oslo: - contributions from the sources to their own grid are not satisfactorily estimated.
- Stockholm: - divisions between source-types are not strictly done (diurnal variations of emissions were used to define industrial/domestic heating sources);  
- traffic information is lacking for city outskirts and suburbs;  
- diurnal concentration variations are not considered in exposure calculations.

It is very difficult, perhaps impossible, to quantify the uncertainties of these modeling results. The overall uniformity of the computational procedures, however, should allow at least an assessment of the relative importance of the various source categories in a given city.

## 6 Human exposure to toxic metals

Atmospheric dispersion modeling and calculation of inhalation and deposition in respiratory airways are only of minor importance when considering the total intake of toxic elements by humans. The largest contribution to this intake is via deposition on agricultural crops and through consumption of foodstuffs.

To quantify the uptake of pollutants via the food-chain, simple, quasi-stationary, compartment models can be used.

The elements of such a model are shown in Figure 10.

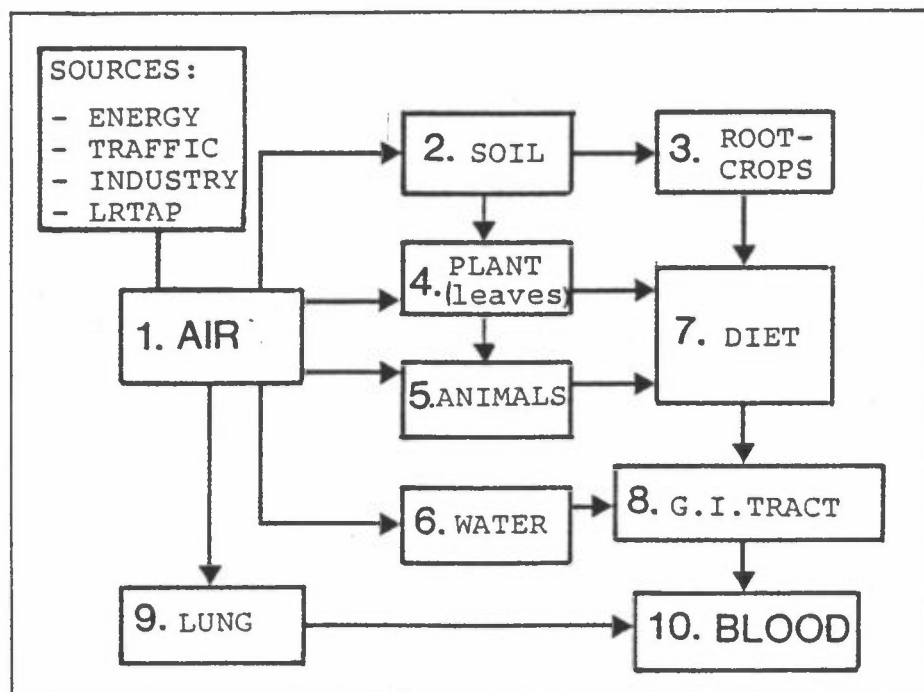


Figure 10: Simple, quasistationary compartment model for calculation of steady-state concentration of lead in blood for selected groups of people, based upon estimated air concentrations from the different source categories.



Similar quasistationary models have been used in radioecology, viz., for estimation of doses by intake of radioisotopes in connection with accidental releases from nuclear power plants. Recently, however, it has been demonstrated that this type of model can also be used to calculate exposures due to ingested toxic metals (Bennett, 1981).

The model assumes that quasistationary equilibria between the concentrations of toxic metals in the various media (air, soil, vegetation, etc.) exist. The resulting transfer factors can be used to calculate human exposure (or the concentration in a given "target" organ), if airborne concentrations due to the various source categories are known, or can be estimated.

It was decided at an early stage in the MIL 4 project to model exposures to cadmium (Cd) and lead (Pb). Cadmium was chosen, because of concern with cadmium contamination of agricultural products, which is due to deposition from the atmosphere, cadmium in phosphate fertilizers, and to application of sewage sludge as a fertilizer. The laboratory at Studsvik had relevant experience from work with radioisotopes, and had also used this method for estimating the impact of benzpyrene emissions from waste incineration. Compartment model calculations have been applied to establish dose-response factors for cadmium, in connection with the planned emission regulations for this element in Sweden (Ramberg 1983). A recent inventory of Cd emissions from various sources in Denmark (Hansen, 1980) led to the selection of Sjælland as one of the study areas.

In Norway the topic of lead exposure had been introduced as a pilot study in the Sarpsborg-Fredrikstad area, as part of an air pollution monitoring program (Hagen et al., 1984). Coordination of this pilot study and MIL 4 resulted in the following goals for the study:

- establishment and validation of models for exposures via air and food intakes;
- estimates of total exposure of individuals to lead in blood;
- evaluation of the significance of inhalation in relation to the food intake;
- assessment of the significance of different emission source categories in relation to exposure of the local population.

The basic data for this study included statistics on land use, population, living habits, and food consumption, in addition to emission estimates and dispersion of airborne lead within the area. Contributions from long range transport of air pollutants were also included.

#### 6.1 CADMIUM EXPOSURE IN SJÆLLAND (ZEALAND).

Cd exposure to the Danish population of Sjælland was estimated to mainly originate from regional background and long-range transport deposition (29%), and the use of fertilizers (27%). Inhalation represented 41% of the total population exposure, mostly due to cigarette smoking.

Cadmium is a widely dispersed element, and it has many potential emission sources. In this survey, the following emission source categories have been considered:

- generation of electricity and heat from combustion of coal and oil,
- waste incineration,
- application of commercial fertilizers,
- "background" deposition (long-range transport of air pollutants from sources outside Sjælland).

Only inhalation of airborne cadmium and intake of cadmium via foodstuffs were considered as modes of exposure. Airborne concentrations were estimated using a Gaussian dispersion model, with one year of meteorological data from Risø's 100 m instrumented mast. Dry- and wet-deposition were calculated for selected values of surface roughness, cadmium-containing aerosol size distributions, and precipitation intensities.

The concentration of cadmium in soil (2 layers) and the root uptake was calculated by means of a simple compartment model, in which the available concentration of cadmium in the tilled layer was related to the pH of surface water (Christensen and Tjell, 1983).

Estimated emissions of cadmium to the atmosphere in Denmark are given in Table 14.

Table 14: Annual emissions of cadmium to the atmosphere in Denmark (tonnes/y).

| Source category  | Denmark    | 19 selected sources on Sjælland |
|--|------------|---------------------------------|
| Energy sector<br>- coal-combustion<br>- oil-combustion | 0.5<br>0.9 | 0.35                            |
| Waste incineration<br>- household waste<br>- used cars | 2.2<br>0.9 |                                 |
| Industrial processes<br>- car painting                 | < 0.2      | -                               |

The locations of the 19 selected single sources of cadmium emissions in Sjælland are indicated in Figure 11.

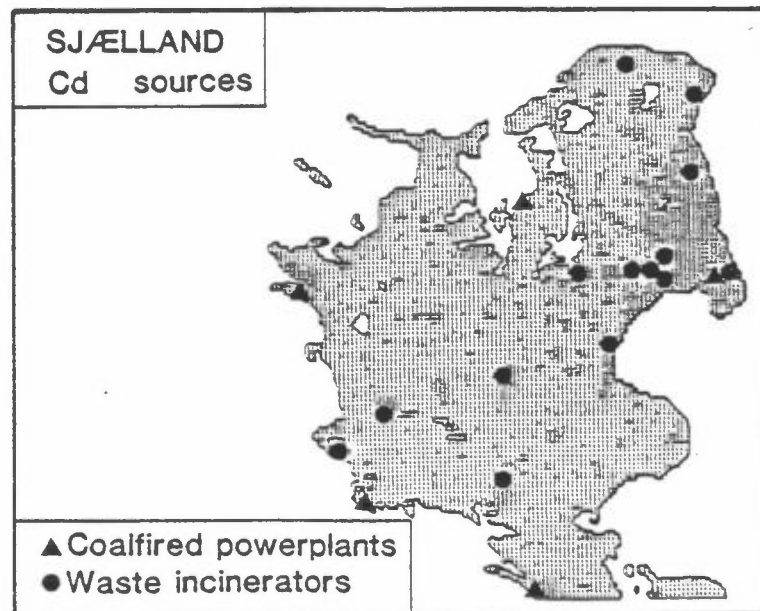


Figure 11: Locations of 19 selected single sources of cadmium emissions in Sjælland. The none shown shading indicates the population distribution.

The percentage contributions to Cd accumulated in the body, calculated from emissions from different sources to individuals and to the total population, is summarized in Table 15.

Table 15: Percent contribution from different source categories to individual and population exposures from cadmium accumulated in the body.

| Source category  | Individual exposure |              | Population exposure |               |
|--|---------------------|--------------|---------------------|---------------|
|  | inhalation          | via food     | inhalation          | via food      |
| Accumulated in soil<br>Annual fertilizer use                                     |                     | 41<br>0.16   |                     | 56<br>0.21    |
| Background concentration<br>- air<br>- crop surfaces<br>- via soil - root/plant  | 0.49                | 4.6<br>0.17  | 0.66                | 6.3<br>0.23   |
| Energy prod./waste incin.<br>- air<br>- crop surfaces<br>- via soil - root/plant | ~0.01               | 0.49<br>0.02 | ~0.01               | 0.66<br>0.024 |
| Other foodstuffs   |                     | 18.9         |                     | 25.8          |
| Smoking  | 34.1                |              | 10.3                |               |
| Total  | 35                  | 65           | 11                  | 89            |

The largest contribution to the total exposure of Cd is due to Cd already accumulated in the soil (56%). Of these 56%, 29% was estimated to originate from long-range transport and regional air pollution deposition, and ~27% from previous use of fertilizers. In addition, Cd content in food, which cannot be explained in terms of root uptake from the soil, contributes to about 26% of the total population exposure. Tobacco smoking is responsible for 10% of the total population exposure, but as much as 34 % for habitual smokers (20 cigarettes/day). Inhalation of ambient air represents less than 1 % of total exposure, while deposition onto vegetation and crops accounts for approximately 6 % for the average population.

Figure 12 summarizes the relative contributions, in percent, from the different source categories to the total population exposure in Sjælland.

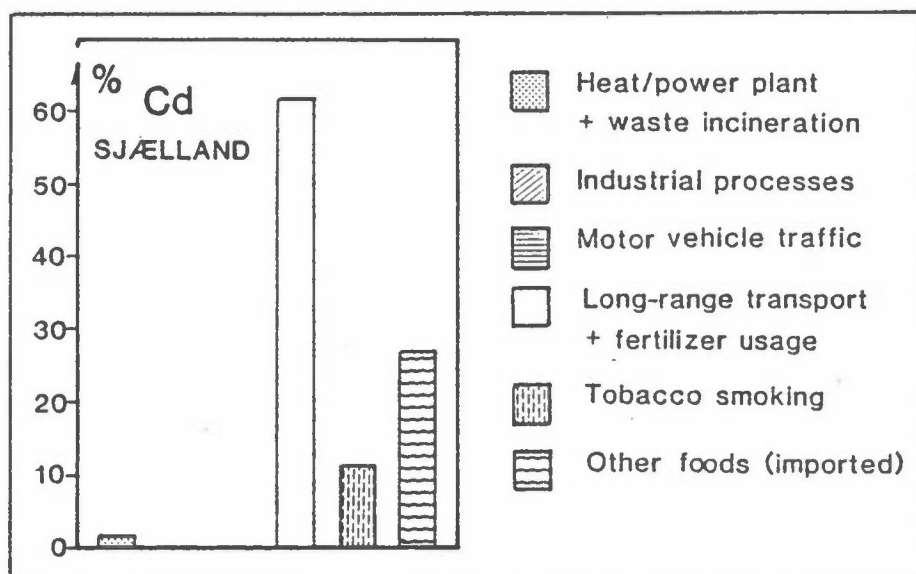


Figure 12: The relative contribution (%) from different source categories to the population exposure of cadmium in Sjælland.

Before drawing final conclusions, however, the following facts should be considered:

- only 14 % of cadmium emissions from fossil fuel combustion and waste incineration on Sjælland is deposited within the area. This represents 0.7 % of the exposure, while deposition of cadmium from sources outside Sjælland (LRT) is ten times higher. Deposition of Cd from the atmosphere is, therefore, an international problem;
- accumulation of cadmium in surface soil is a result of many years of fertilizer application and atmospheric deposition. Together these sources are responsible for about 60 % of the Cd intake;
- contributions from water pollution, and use of cadmium-containing products (plastic, glazed ceramic utensils), were not considered in this study.

Estimated future developments regarding cadmium exposures has already been addressed in a special report (Edlund and Karlberg, 1984). It points out that continuous liming of cultivated soils will be necessary. If this is not done, cadmium exposures can be expected to increase drastically.

## 6.2. LEAD EXPOSURE IN THE SARPSBORG - FREDRIKSTAD AREA

Lead exposure (Pb concentration in blood) to the total population of Sarpsborg-Fredrikstad was estimated to be mainly due to automobile traffic (40%), and long-range transport of air pollutants (~26%). Exposure through direct inhalation only represents 10-20% of the total exposure.

The Sarpsborg-Fredrikstad area (S-F) is located ~75 km south of Oslo on the eastern side of Oslofjord. It contains most of the typical air pollution sources: industries of various kinds, space heating, waste incinerating, and traffic. A map of the area is shown in Figure 13.

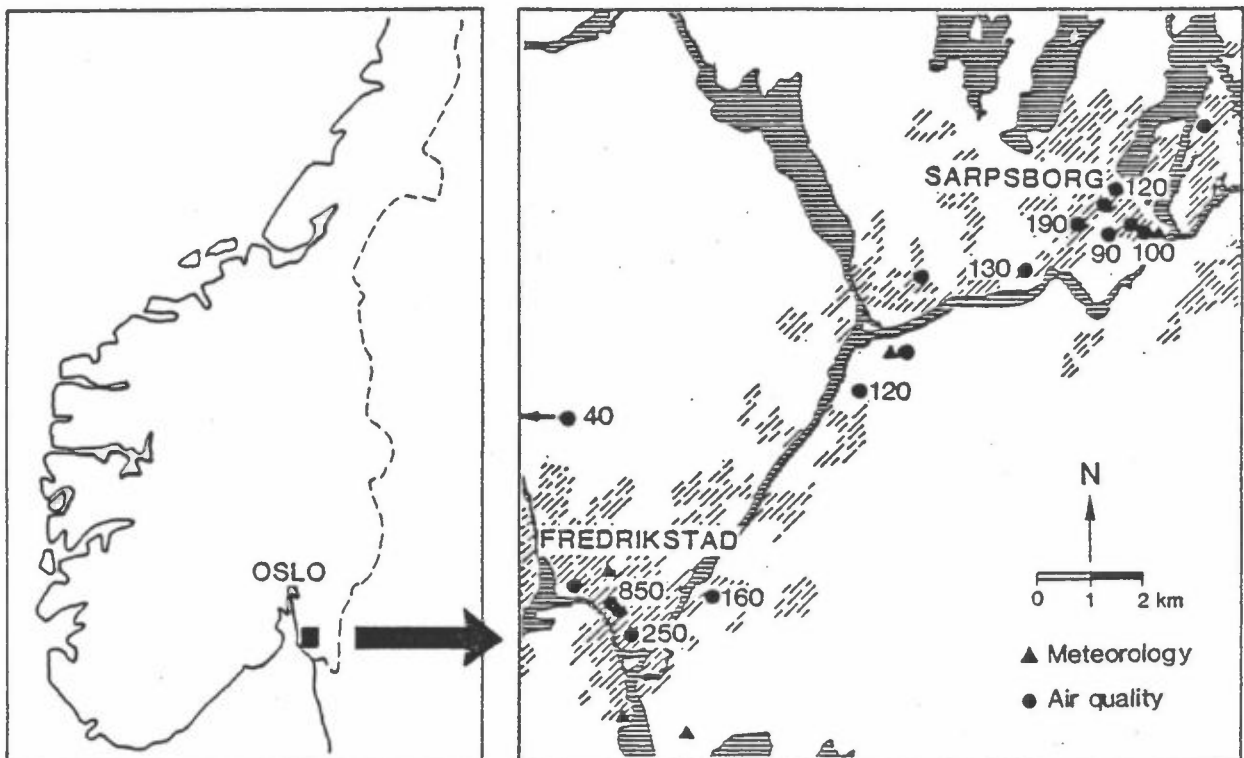


Figure 13: Location of the Sarpsborg-Fredrikstad area with sampling stations indicated. The numbers are average lead concentrations measured during the winter of 1981/82 ( $\text{ng}/\text{m}^3$ ).

A screening of the main sources of lead in human food were performed on the basis of earlier investigations (Mathiesen, 1980). Results of the screening, with data relevant for the study area, are shown in Table 16.

Table 16: Estimated total intake of lead with foodstuffs based on average concentration, and consumption figures.

| Product             | Average concentration<br>mg/kg | Consumption<br>g/day | Intake<br>$\mu\text{gPb/day}$ |
|---------------------|--------------------------------|----------------------|-------------------------------|
| Bread, cereals      | 0.1                            | 214                  | 21.4                          |
| Fruit and berries   | 0.1                            | 127                  | 12.7                          |
| Meat                | 0.1                            | 105                  | 10.2                          |
| Milk/dairy products | 0.02                           | 497                  | 9.9                           |
| Potatoes            | 0.05                           | 182                  | 9.1                           |
| Vegetables          | 0.02-0.05                      | 28                   | 0.9                           |
| Fish                | 0.16                           | 55                   | 8.7                           |

The total intake was calculated to be 86  $\mu\text{gPb/day}$ , of which the listed food products represent about 85 %. Other food products, mainly imported, account for the remaining 15 %.

A schematic representation of the procedure for estimating individual exposure for selected population groups, as well as for the general total population, is shown in Figure 3 (section 4).

Inhalation exposure is calculated from the time spent in outdoor and indoor areas, for which airborne concentrations have been estimated. Food consumption data for different population sub-groups and estimated concentrations of lead in food products are used to calculate lead intake via foodstuffs.

More detailed description of the procedure for calculations of lead intake and exposure is given by Sivertsen (1984b). The following sources of lead exposure within the Sarpsborg-Fredrikstad area have been considered:

- motor vehicle traffic;
- long-range transport of air pollutants (regional background);
- industrial processes;
- space heating;
- intake of "other" food (imported).

Airborne concentrations of lead were calculated from dispersion models, supported by measurements. Detailed documentation is appended to the final report from the pilot surveillance study (Hagen et al., 1984).

The fraction of the lead exposure, which is due to direct inhalation of airborne lead-containing aerosol particles, has been investigated separately for special population groups, in order to demonstrate the significance of this exposure mode, in relation to the exposures from lead intake via foodstuffs.

The calculations represent the annual mean blood lead levels for 78 different population sub-groups (adults, children, males, females, smokers, non-smokers, inhabitants of particular areas, occupational sub-groups, etc.), moving between 30 different habitats (indoor, outdoor, street, urban, rural, commuting, workplace, etc.). The total population of the area was 108 000.

The percent exposures due to inhalation only are summarized in Table 17.

Table 17: Percent fraction of lead in blood from inhalation for selected population sub-groups.

|                           | Urban   | Rural   |
|---------------------------|---------|---------|
| Adult men                 | 11 - 28 | 6 - 15  |
| Adult women               | 16 - 28 | 12 - 15 |
| School children           | 15 - 28 | 6 - 7   |
| Old age pensioners        | 16 - 25 | 5 - 8   |
| High exposure groups:     |         |         |
| industrial workers        | 48 - 56 | 35 - 51 |
| service station personnel | ~ 80    | -       |
| taxi drivers              | ~ 52    | -       |

For adults, not subject to occupational exposure, inhalation represents only 11 - 28 % of the lead exposure to the population sub-groups living in urban areas, and only 6 - 15 % for sub-groups living in the surrounding rural area.



The total population exposure was calculated on the basis of number of individuals in the respective exposure sub-groups. Calculations of relative and absolute contributions were carried out for all pollution sources and source categories. Figure 14 shows the number of persons exposed to levels of lead in blood (in  $\mu\text{g Pb/dl}$  blood) above given values.

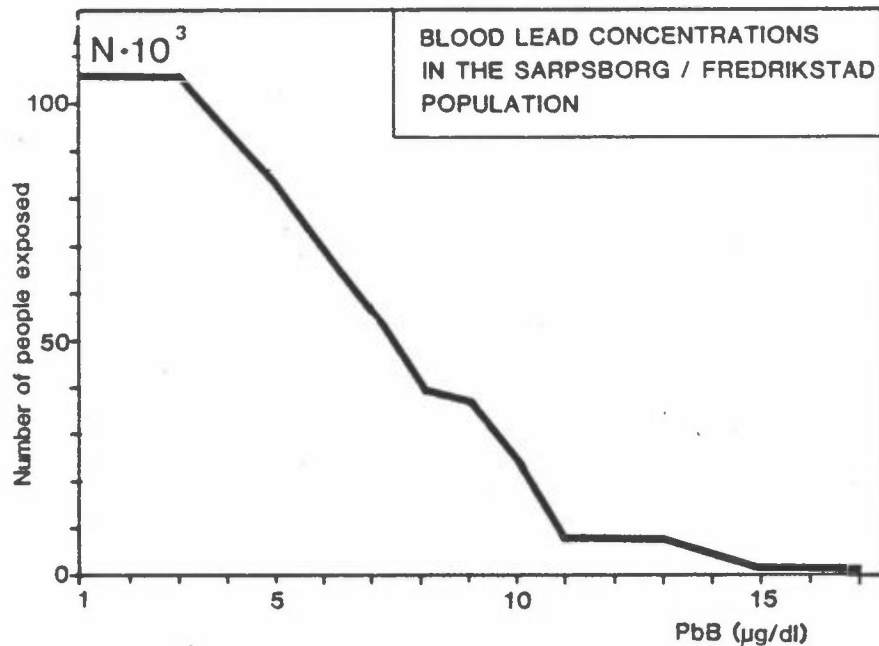


Figure 14: Number of persons in the Sarpsborg-Fredrikstad area with estimated concentrations of lead in blood above given levels.

Similar curves, as the one shown in Figure 14, were estimated for the contributions from single source categories (traffic, energy production, long-range transport, etc.). These estimates showed that in a sub-population of 10 000 individuals, motor vehicles alone contributed  $\sim 6 \mu\text{g/dl}$  of blood lead, out of a total of  $\sim 11 \mu\text{g/dl}$  for this group. Integration of the distribution functions in Figure 11, results in a picture of the relative contributions to the total population exposure of lead in blood in the area, given for the selected source categories. The results of these estimates are shown in Figure 15.

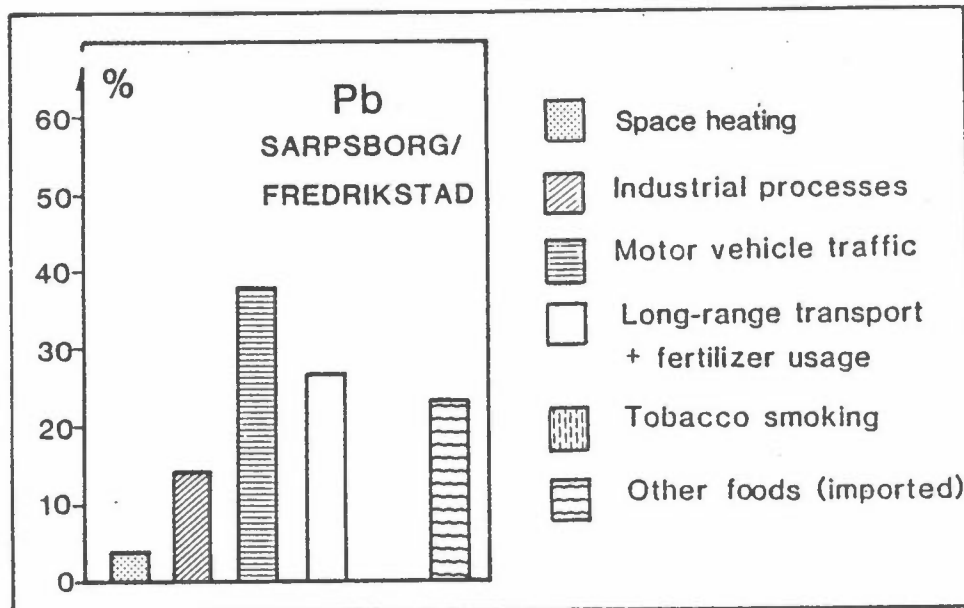


Figure 15: The relative contribution (%) from different source categories to the estimated population lead exposure in the Sarpsborg-Fredrikstad area.

These estimates show that emissions from motor vehicles were responsible for 38% of blood lead levels before the present regulations of lead in gasoline came into effect in 1983. Long-range transport of air pollutants, which is mainly affecting the concentrations of lead in agricultural products, accounted for 26%, and local industry represented 14% of the estimated total exposure.

The estimation of blood lead exposure to the population of Sarpsborg-Fredrikstad represented a first attempt to model the concentrations of a substance in a human organ from the knowledge of its air concentrations. Several uncertainties became apparent during the course of the modeling. These are hard to quantify without performing a detailed sensitivity analysis, based upon more measured concentration data than were available for the area.

In future studies of this kind, better information should be available on:

- physical/biological parameters, such as, retention times, absorption coefficients, etc.;

- measured concentrations of lead in air, soil, vegetables, food products, water and blood (to verify and improve the model parameters).

### 6.3 APPLICATIONS, LIMITATIONS AND UNCERTAINTIES

Even if it is thought that the compartment models for estimating population exposures to toxic metals are still in a stage of continuous development, the results presented within the framework of MIL 4 give a good picture of the relative importance of the different sources. When these models are developed and verified for a given area, component and human target organ, they can easily be applied to quantify effects of changes in source emissions, and to optimize air pollution abatement strategies.

To study the different parameters of importance for estimated changes in the exposure, a sensitivity analysis was performed for the Sjølland cadmium exposure study. The results were found sensitive to the assumption of pH-values in the soil. As pH was changed from 4 to 8, the estimated intakes varied from 0.24 to 10.0  $\mu\text{g}/\text{day}$ . Whole body exposure to cadmium is also sensitive to the estimated ground level concentrations and intake via wheat consumption.

No sensitivity analysis was performed for the Sarpsborg-Fredrikstad area blood lead exposure study. The estimated blood lead levels were, however, compared with measured levels of blood lead in other areas of southern Norway and found comparable.

Recent findings (Clench-Aas et al., 1984) indicate that the model might overestimate blood lead levels, and the following areas of uncertainty are pointed out:

- the contributions from different source categories to air concentrations of lead in the different environments are not specified adequately;
- assumption of concentrations in "other foods" are questionable;
- partitioning of persons in the various sub-groups is too limited and could have been expanded.

## 7 Use of reseptor models

The principles of receptor models are briefly described in Section 4.3 and in the subprojects reports (Lövblad and Steen, 1982; Steen et al., 1984; Schaug, 1984; Brorström-Lunden and Steen, 1984). Table 18 summarizes the use of receptor models as reported in this Section.

Table 18: Receptor models used in MIL 4.

| Area            | Air pollution data            | Medium     | Model type*  | Number of recept. pts. | Instit. |
|-----------------|-------------------------------|------------|--------------|------------------------|---------|
| Ørebro          | V, Cr, Mn, Ni, Pb, Fe         | Filter     | CMB          | 2                      | IVL     |
| København       | 15 elements                   | Filter     | PCA/FA, CMB  | 5                      | MSTL    |
| Sundsvall       | PAH (30 comp.)                | Filter/PUR | PCA, CA, CMB | 4                      | NILU    |
| Helsingfors     | Cd, Cr, Cu, Fe, Ni, Pb, V, Zn | "Mossbags" | CV, PCA, FA  | 164                    | UiH     |
| Southern Norway | 26 elements                   | Moss       | CMB, PCA     | 490                    | NILU    |
| Göteborg        | V, Cu, S, Ni, Pb, C           | Filter     | CMB          | 1                      | IVL     |
|                 | PAH (6 comp.)                 | Cabbage    | CMB          | 4                      | IVL     |

\* CMB = chemical mass balance  
 FA = factor analysis  
 PCA = principal component analysis  
 CA = cluster analysis  
 CV = concentration variability

For measurements of suspended particulate matter, a number of elements have been analyzed and the methods have been used satisfactorily. In cases where data on vegetation and organic compounds (PAH) have been used, problems have arisen in the interpreting of the results. Some of the reasons for this are:

- varying retention time,
- different reaction mechanism,
- varying uptake on different particle fractions.

Common shortcoming for all areas, where a mass balance model has been applied, was that the source composition often was not accurately known.

### 7.1 ELEMENTS ON FILTERS FROM ÖREBRO

CMB estimates on aerosol data from 2 locations in Örebro showed that soil (crustal dust) and background air pollution represented the major sources for TSP, while traffic contributed to 70-98% of Lead, and oil heating to ~85% of the vanadium on filters.

Based on a screening survey of existing air quality data from the Nordic countries (Löwblad and Steen, 1982), IVL chose to perform receptor modeling on high volume filter data of street- and roof-level concentrations in central Örebro. A chemical mass balance model was used.

The measurements were originally not designed for receptor model analysis, and were affected by traffic (16 000 cars per day). Örebro's main pollutant source is an oil-fired heating plant, and only a small copper smelter represents the industrial emissions. Soil dust composition was studied from surface soil samples.

Table 19 summarizes the contributions from 4 source groups to the fine particle fraction in total suspended particulate matter (equiv. aerodynamic diameter < 2.5  $\mu\text{m}$ ).

In winter, at the street-level station, soil dust represented the largest source for total suspended particulate matter. Vanadium originated from oil heating, lead from traffic, and iron from soil dust. During the summer, "background" and long-range transport of air pollutants represented a more significant source of total suspended particulate matter, than during the winter season. Concerning the individual elements, the same picture as during the winter season is evident. Coarse particles, on the other hand, originated largely from soil dust (Steen et al., 1984).

Table 19: The contributions (%) from 4 source categories to the particles in air (<2.5 µm dia.) for vanadium (V), lead (Pb), iron (Fe), and total suspended particulate matter (TSP), based upon samples from street-level in central Örebro.

| Season | Component | SOURCE CATEGORY |         |             |            |
|--------|-----------|-----------------|---------|-------------|------------|
|        |           | Soil dust       | Traffic | Oil heating | Background |
| Winter | V         | 33 (7)*         | 2 (1)   | 85 (20)     | 2 (20)*    |
|        | Pb        | 1               | 98      | <0.1        | 0.1        |
|        | Fe        | 97              | 1       | <1          | <1         |
|        | TSP       | 82              | 4       | 2           | 2          |
| Summer | V         | 10              | 2       | 85          | 3          |
|        | Pb        | 4               | 70      | 0.1         | 25         |
|        | Fe        | 49              | 1       | 2           | 39         |
|        | TSP       | 18              | 2       | 2           | 49         |

\*roof station.

## 7.2 ANALYSIS OF AIRBORNE DUST IN KØBENHAVN

The major contributions to TSP in central København were estimated to originate from traffic (34%), reentrainment of soil dust (26%), and energy production (18%). Outside the city center, regional and long-range transport represented ~40% of the contribution.

In the Danish national air pollution monitoring program, total suspended particles (TSP) and particle elemental composition were determined at 5 stations in greater København area during 1983. By means of factor analysis, the number and types of pertinent sources were determined. TSP was then regressed on the standardized source contributions, establishing the contributions from individual sources to the variation of TSP. Finally, the annual average source contributions to TSP were established by means of the

"tracer" element for each source. Five stations in the greater København area were selected for MIL 4 studies. The elements Al, Si, S, Cl, K, Ca, Ti, V, Fe, Mn, Ni, Cu, Zn, Br and Pb were analysed in the aerosols most of the time. The chemical mass balance model required knowledge about the source composition. Prior to the CMB analysis, a principal component analysis was performed on the existing data. The factors emerging from this analysis defined the main source categories in København:

- soil dust (Al, Si, Ti),
- a traffic factor (Pb, Br),
- fossil fuel, oil (V, Ni),
- long-range transport (S, Mn, Al),
- seasalt (+traffic) (Cl, Br),
- grass burning (K) (uncertain),
- waste incineration (Cu, Zn, Mn).

Following the PCA analysis, a regression analysis of the standard source contributions against the total suspended particulate matter, and the monthly deviations from the annual average source contributions were estimated.

Finally, the annual average source contributions to TSP were established by means of the tracer elements for each sources. The annual average percentage contributions from different source groups to the TSP at the 5 sampling stations in København are shown in Table 20.

Table 20: Annual average contribution (%) from different source categories to the total suspended particulate matter load at 5 measuring points in København.

| Sampling station | SOURCE CATEGORY |                           |       |         |                       |
|------------------|-----------------|---------------------------|-------|---------|-----------------------|
|                  | Soil            | Energy<br>Traffic Heating | LRTAP | Seasalt | Straw<br>burn-<br>ing |
| H.C.A. Boulevard | 20-31           | 38      21                | 24    | 7       | -                     |
| Vanløse          | 24              | 35-38                     | -     | -       | 4                     |
| Tårnby           | 16-17           | 29-36                     | 41    | -       | 5                     |
| Glostrup         | 20              | 38-51                     | 41    | -       | -                     |
| Ballerup         | 23-25           | 40-51                     | -     | -       | 12                    |

Soil (crystalline dust) and emissions from coal-fired energy production were a dominating source for all 5 stations. Traffic and burning of fossil fuels contributed also to all 5 stations. However, only in the city center could these two sources be separated. Finally, long-range transport of sulfur-containing particles was identified at all stations. Less pronounced were the contributions from refuse combustion, straw burning and seasalt, all of which were only seen at some of the stations.

In summary, soil dust accounted for 15-25% of the annual mean TSP. In the center of København, traffic emissions accounted for 40%, and fossil fuel burning for 20% of the annual mean of TSP. At the peripheral stations, however, the combination of traffic and fossil fuel burning made up 32 to 50%. Long-range transport amounted for 41% at the peripheral stations but merely 24% at the city center station. For the rest of the sources no "tracer" element could be found, and thus contributions from these sources could not be assessed. Figure 16 shows that the contributions from soil dust and energy sources vary over the year.

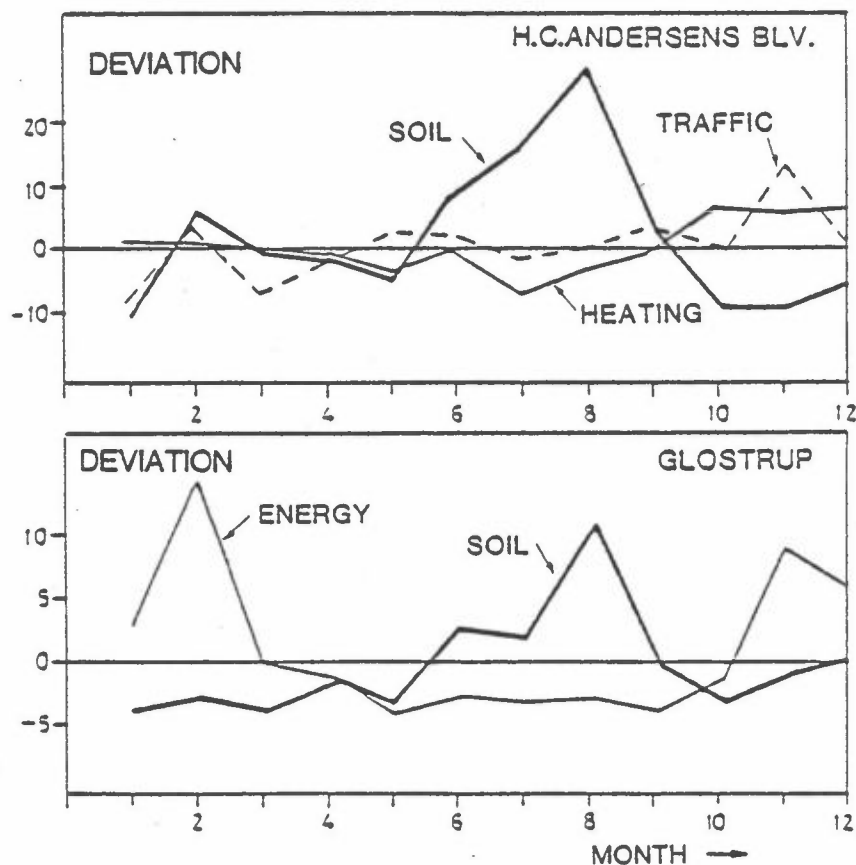


Figure 16. Estimated monthly deviation from annual source contributions in central and suburban København.



Soil dust clearly is the largest contributor in late summer in central and suburban København, while the energy sources are the largest contributors during the winter. Long-range transport seems to contribute mostly during winter and spring (Kronborg et al., 1984).

### 7.3 DEPOSITION OF HEAVY METALS IN HELSINKI

The estimated relative contributions from different sources to the deposition on moss bags in Helsinki varied considerably from one area to another. Ash, which was assumed to be a measure of TSP, originated mainly from energy production (12-53%), traffic (10-77%) and background (LRT) (9-38%).

Heavy metal pollution of the Helsinki air was studied during the winter of 1982/1983 with the moss bag technique. Special attention was paid to the most important emission source categories: refuse burning, traffic and energy production. About 600 moss bags were suspended from twigs of small birches around the study area near the central and eastern part of the city. Accumulations of ash\*, Cd, Cr, Cu, Fe, Pb, Ni, V and Zn in moss bags over a 5 month period (146 days from mid-October to mid-March) were determined, with the elements analyzed by standard flame AAS method.

The greatest accumulations were found near a waste incinerator and near the main streets, with daily traffic densities of 30 000 to 50 000 vehicles. Also the influence of coal-fired power plants in the city center was significant, particularly for ash, Ni and V.

The relative contributions of the different emission sources were estimated by means of factor analysis. Three main factors emerged, as characterized by the chemical parameters:

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\*"ash" = inorganic residue of ashed moss, presumably representative of TSP.

- traffic: Fe, Pb, Cr, and ash
- energy production: V, Ni,
- waste incineration: Cd, Zn.

In addition, long-range transport (LRT) and background air pollution were estimated from measurements at so-called "background stations" outside Helsinki. To estimate the relative influence of the different source categories, 4 representative sub-areas were selected on the basis of factor analysis of metal accumulation. Four measuring station locations were then selected to represent:

1. Clean area (Central Park, Kumpula, away from emission sources).
2. Traffic area (Härneentie).
3. Area around waste incinerator (Kyläsaari).
4. Energy production area (coal-fired power plants, Hanasaari).

The relative contributions from the various source categories to the accumulations in moss bags of ash, lead and cadmium are given in Table 21.

Table 21: The relative contributions (%) from 4 source categories to the accumulation of ash, lead and cadmium in moss bags in 4 areas of Helsinki.

| Component | Area* | Contribution (%) from source categories |                   |         |                 |
|-----------|-------|---|-------------------|---------|-----------------|
|           |       | Energy                                  | Waste incinerator | Traffic | Background(LRT) |
| Ash**     | 1     | 30                                      | 2                 | 30      | 38              |
|           | 2     | 12                                      | 2                 | 77      | 9               |
|           | 3     | 50                                      | 24                | 10      | 16              |
|           | 4     | 53                                      | 2                 | 35      | 10              |
| Pb        | 1     | 20                                      | 37                | 17      | 26              |
|           | 2     | 10                                      | 23                | 57      | 10              |
|           | 3     | 15                                      | 69                | 6       | 10              |
|           | 4     | 35                                      | 33                | 18      | 5               |
| Cd        | 1     | 6                                       | 63                | 6       | 25              |
|           | 2     | 10                                      | 60                | 13      | 17              |
|           | 3     | 5                                       | 86                | 2       | 7               |
|           | 4     | 26                                      | 56                | 3       | 15              |

- \* 1 = Kumpula (park)  
 2 = Härneentie (dense traffic)  
 3 = Kyläsaari (waste incinerator)  
 4 = Hanasaari (coal-fired power plant)  
 \*\* presumably representative of TSP.

The relative contributions vary considerably from one area to the other. In Kumpula, which is not influenced by local sources, the long-range transport and background air quality represent 25 to 38% of the total accumulation. Lead and cadmium originate mainly from waste incineration (37 and 63%, respectively) while the energy production sources contribute 6% cadmium, 20% lead, and 30% ash in the park area.

#### 7.4 ELEMENTS IN LIVE MOSS IN SOUTHERN NORWAY

At the background station BirkeLand, long-range transport was assumed to account for most of the accumulation of elements in living moss. A chemical mass balance estimate indicated that waste incineration and industrial sources could explain 47% of the accumulation of 9 selected elements, traffic 29%, and the burning of coal and heavy oil 24%.

Moss samples (Hylocomium splendens) were collected at 490 different locations in Norway during the summer of 1977. The samples were then analyzed mainly with the neutron activation method (Hanssen et al., 1980), and concentrations of 26 chemical elements obtained. A principal component analysis was conducted to apportion the sources for the chemical elements, and a mass balance estimate was used to estimate the source contributions.

The results from these estimates show that the chemical elements were characterizing a number of factors, which could identify the sources and source categories. The interpretation of the results was difficult due to different retention times of the different chemical elements in moss. Factor scores were used to identify the results.

The lack of quantitative information about the retention times of the different trace elements and other uncertainties limited the possibility of quantitative results. However, an attempt at estimating the contribution of

anthropogenic sources to one sample from Birkeland (southern coast of Norway) was made.

The possible sources were divided into 5 groups. Table 22 shows the contributions from these sources to the air pollution load of 9 elements in the moss (arsenic, cadmium, chromium, lead, selenium, vanadium, zinc, nickel and cobalt), considered the most important tracers of anthropogenic sources.

Table 22: The contribution from 5 source groups to the total concentrations,  $\sum C_i$ , of 9 trace elements in moss, collected at Birkeland in southern Norway.

| Source category      | $\sum C_i$ (ppmw) | % contribution |
|----------------------|-------------------|----------------|
| Heavy oil combustion | 13.6 $\pm$ 5.8    | 6              |
| Coal combustion      | 39.8 $\pm$ 10.0   | 18             |
| Vehicular traffic    | 66.9 $\pm$ 18.4   | 29             |
| Waste disposal       | 95.7 $\pm$ 53.6   | 47             |
| Industrial processes | 11.2 $\pm$ 6.1    |                |

Most of the air pollution load at Birkeland is believed to be due to long-range transport of air pollutants. The analysis showed that long-range transport and local sources of lead, zinc, cadmium, arsenic and selenium explained 34% of the variance in the data. Other anthropogenic sources, releasing nickel and cobalt, explained 7% of the variance. Different types of soil explained 4 to 14% of the variance, and seasalt explained 10% of the variance. Table 22 indicates that, from mass balance analysis, the total load of 9 trace elements in moss at Birkeland is due to waste incineration and industrial sources (47%), traffic (29%), and the use of coal and heavy oil (24%).

## 7.5 CONTRIBUTIONS TO PAH IN PRIMARY ALUMINUM SMELTER SURROUNDINGS

Various receptor model estimates have shown that the contribution to total PAH i air in different primary aluminum smelter surroundings amounts to 50-90%. Even near streets and roads, the smelter contributed to more that half of the total PAH.

A monitoring program of polycyclic aromatic hydrocarbons (PAH) and fluorides in ambient air was carried out in Sundsvall in 1980 and 1981. Samples were collected at 4 stations, once a week on filters and on polyurethane foam (PUR). The samples were analyzed for about 30 compounds of PAH (Thrane, 1982). One aim of this study was to estimate the contribution of PAHs from Gränges Aluminium smelter to the ambient air, relative to other sources of PAHs in the area.

The statistical analyses included computation of average concentrations, medians, frequency distributions and regression analyses of fluorides and PAH for each station, as well as the variation in concentrations with meteorological parameters. An early principal component analysis (PCA) of the data (Henry, 1982) showed that:

- the 22 compounds included were characterizing 3 main sources;
- the lighter PAH compounds had to be excluded due to difficulties in the analysis;
- a good correlation between fluorides and several PAH compounds implicated the aluminum smelter as a central source;
- benzo(a)anthracene seemed to be a good tracer of PAH releases from the aluminum smelter;
- coronene seemed to be a good tracer for vehicular traffic.

Traffic tended to dominate the heavier PAHs, while the aluminum smelter emitted the lighter PAHs. Three PAH compounds (between dibenzothiophene and anthracene) grouped together characterized long-range transport, incineration or oil burning.

A cluster analysis was employed on the data set for evaluation of the relative importance of the different source categories. This method searches for patterns in the data, and divides the data into clusters according to these patterns. The selection of variables for the cluster analysis was based upon the PCA analysis mentioned above. For the identifications of these clusters, the ratios of fluoride and PAH in the different samples, and wind direction were used.

The estimated contributions (%) from the aluminum smelter to the total PAH load at the 4 sampling stations are shown in Table 23.

Table 23: Estimated contributions (%) from the aluminum smelter in Sundsvall to the total PAH load at 4 sampling stations.

| Sampling site        | Area description                          | Contribution from Al smelter |       |        |
|----------------------|---|------------------------------|-------|--------|
|                      |   | Day                          | Night | 24 h * |
| Kubikensborg<br>Haga | <0.5 km from aluminum smelter             | 83                           | 86    | 75     |
|                      | City area, 3 km from<br>aluminum smelter  | 87                           | 84    | 72     |
| Köpmansgata          | City center, traffic                      | 46                           | 54    | 50     |
| Sidsjön              | Background, 4 km from<br>aluminum smelter | 76                           | 57    | 52     |

\* 24 h and day/night are based upon different samples.

The estimated contribution from the aluminum smelter in the city center, where street traffic also contributes, amounted to about 50%. Away from the streets, the contribution from the aluminum plant was estimated to be more than 50%, and at the station closest to the smelter between 75 and 86%.

A mass balance model was also tried, but the source emissions were too poorly characterized. Results from the city center station during the summer period indicated, however, that the contributions given in Table 23 were reproduced.

Estimates, similar to those presented for Sundsvall, have been performed for other areas with aluminum industry in Norway (Thrane et al., 1983). Results of these estimates are presented in Table 24.

Table 24: Estimated contributions of PAH from primary aluminum production (%) of total measured PAH in air at 4 sampling stations.

| Sampling site | Contributing source         | Contribution (%) |        |
|---------------|-----------------------------|------------------|--------|
|               |                             | winter           | summer |
| Høyanger      | Aluminum smelter            | 75               | 85     |
| Mosjøen       | " "                         | 46               | 64     |
| Øvre Årdal    | " "                         | 76               | 83     |
| Ardalstangen  | Anode plant                 | 44               | 58     |
| "             | Aluminum smelter Øvre Årdal | 46               | 32     |

Primary aluminum production in Høyanger, Mosjøen and Øvre Årdal, accounts for 50 to 85% of the total PAH concentration at the given sampling locations. In Mosjøen, traffic and space heating account for about half of the contribution, while these sources at Ardalstangen represent only about 10%.

#### 7.6 PAH IN CABBAGE IN GÖTEBORG

A simple mass balance estimate of different source contributions to PAH in cabbage grown in the Göteborg area indicated that diesel vehicles are the most important source in the city, while long-range transport and diesel traffic account for equal amounts in the remote location outside Göteborg.

An investigation of the deposition of PAH on cabbage in the Göteborg area was carried out in 1981-1982 (Brorström-Lunden et al., 1982). The results showed that PAH content in cabbage was high in central Göteborg, compared to a reference area 35 kilometers north of Göteborg. PAH concentrations decreased quickly with distance from major traffic routes.

A mass balance model was applied to 6 selected compounds of PAH to estimate the relative contribution from 3 source groups at 4 locations in the area:

- gasoline-fueled vehicle emissions,
- diesel engine emissions,
- long-range transport of air pollutants (LRT).

Three locations were representative of central Göteborg: along the street (Polhemsplassen), 10 meters away from the street (Skånegatan 10), and 25 m away from the street (Skånegatan 25). One location was 10 km away from a freeway 20 km outside Göteborg (Sandsjöbacka).

The source composition of the vehicular emissions was assumed similar to that measured from automobiles in Sweden and USA, while background air quality measurements of PAHs at the sampling station Rörvik (south of Göteborg) were taken to be representative of LRT. One difficulty in applying the chemical mass balance model was that the source composition varied little from one source group to another. This resulted in a negative contribution of long-range transport at one of the street stations. The results, presented in Table 25, therefore should be used with caution.

Table 25: The contributions to the total deposition in cabbage of 6 PAH compounds from 3 source categories at 4 localities in the Göteborg area.

| Sampling station | SOURCE CATEGORIES        |                 |                      |
|------------------|--------------------------|-----------------|----------------------|
|                  | Gasoline-fueled vehicles | Diesel vehicles | Long-range transport |
| Polhemsplassen   | 0.36                     | 0.64            | 0.06                 |
| Skånegatan 10    | 0.41                     | 0.60            | 0.04                 |
| Skånegatan 25    | 0.49                     | 0.59            | -0.04                |
| Sandsjöbacka     | 0.11                     | 0.48            | 0.57                 |

Diesel traffic accounts for most of the PAH deposition to cabbage at the sampling locations in central Göteborg. At Sandsjöbacka (outside Göteborg) long-range transport of air pollutants accounts for more than half of the PAH deposition, while diesel traffic accounts for slightly less than half.



One source of error in the analysis of PAHs in vegetation might be the fact that reactive compounds undergo changes. PAH profiles identifying the sources may thus not be representative of those found in vegetation samples. A study of PAH profiles in different cabbage, with different exposure times shows, however, that the selected PAH compounds were stable after deposition onto the cabbage.

## 7.7 APPLICATIONS, LIMITATIONS AND UNCERTAINTIES IN RECEPTOR MODELING

### 7.7.1 Applications

Receptor models have been demonstrated to be useful in apportioning the relative contributions from different sources, where information on several air pollutants is available on ambient air or vegetation samples. The most often used type of receptor model used in MIL 4 is the chemical mass balance model (CMB). CMB has been mainly used to analyse aerosol data, but can also be used on other types of data. Receptor models should not be considered an alternative to source-oriented dispersion models, but rather complementary to these. The chemical mass balance model can identify and quantify the source contributions of particles and non-reactive gases, and thus improve existing emission inventories, used as inputs to dispersion models (see Figure 3 in section 4). Receptor models cannot, however, be used to estimate the effects of changes in emissions.

Some typical applications of receptor models were demonstrated through examples from the sub-projects, reported above in this chapter. Another way of presenting some of the results is shown in Figure 17.

As can be seen from Figure 17, the relative contributions from energy and heat production depend largely upon the selection of components, and sampling locations. For the selected data sets in Figure 17, the contribution from energy varies from 2% in Ørebro (which also has a large fraction of unexplained contributions) to 50% in Kyläsaari, which is located close to coal-fired power plants. A typical contribution from energy sources seems to be between 25 and 50%.

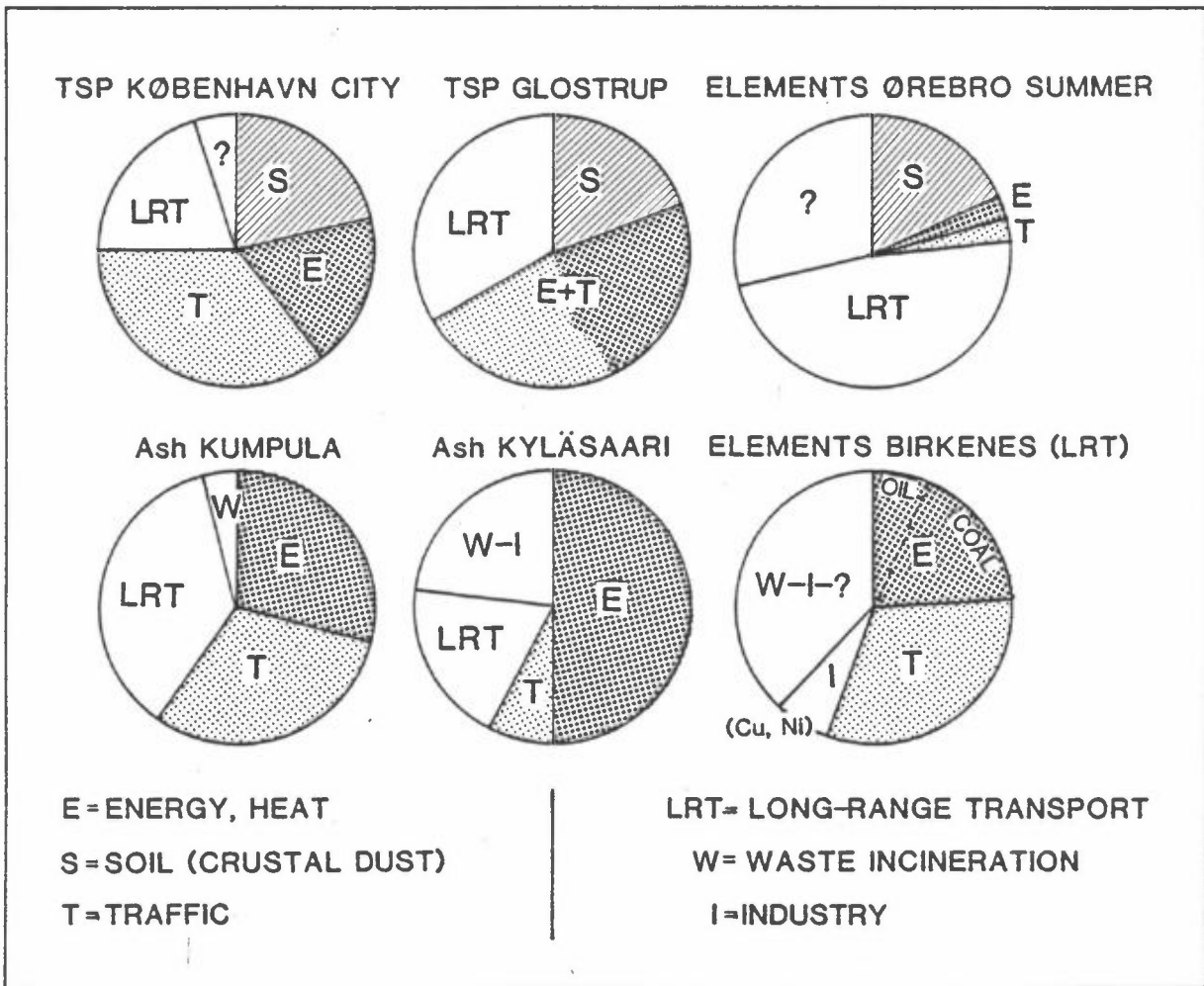


Figure 17: Receptor model results.

Examples of estimated relative contributions from different source groups to the concentrations of total particulate matter on filters (TSP), ash (in moss bags), or selected elements (on filters).

### 7.7.2 Limitations and uncertainties

Reliable apportionment of the different source contribution to the total air pollution load requires information on as many as possible key compounds. Aerosols are usually analyzed by x-ray fluorescence or proton-induced X-ray emissions (PIXE), for which the filter media have to be selected with care. Aerosols are often sampled in two size fractions, separating particles of less or larger than about 2  $\mu\text{m}$  aerodynamic diameter, to avoid that chemical

reactions change the nature of the samples collected on the filters, and to obtain better information about the sources of the aerosols.

Limitations and uncertainties in the use of receptor models, especially where CMB models have been applied to apportion the contributions of the different source groups, include:

- uncertainties in the receptor point measurements,
- uncertainties in estimated source composition,
- systematic errors in measurements or in source composition data,
- chemical reactions and deposition of some of the compounds or elements in transport between the source and receptor point,
- changes in the importance of sources due to dependence on wind speed and wind direction,
- several emission sources with similar composition.

An attempt to quantify such uncertainties was performed in March 1982 at an EPA workshop (Stevens and Pace, 1984). Several participating experts were asked to independently apply receptor models to apportion ambient aerosol mass to emission sources in Houston, Texas, from measured data, and on a simulated data set, estimated from dispersion modeling. Some of the conclusions from this exercise (Schaug, 1984) were:

- the chemical mass balance model, combined with multivariate methods, gave results within 10 to 15% from the "true" values of the simulated data set;
- using the Houston data set, the different investigators obtained results within 20 to 50% of the "true" values.

## 8 Other sub-projects in MIL 4

### 8.1 POPULATION EXPOSURE TO PAHs IN OSLO

Estimates of the total population exposure to PAHs through inhalation in Oslo have shown that traffic contributes about 25-30%, space heating and combustion of fuel oil and coal contribute with 35-55%, while background and long-range transport represent about 20%.

The total exposure to PAHs during the winter half-year has been estimated for Oslo, based upon the same methodology, as employed for SO<sub>2</sub> and NO<sub>x</sub> and described in the previous chapters. The exposures thus only represent that part of the total exposure which is due to direct inhalation. The calculations were based upon information on the total traffic load in Oslo, the consumption of fuel oil, and combustion of coal and wood in different part of the city. Total releases of PAHs were estimated from emission factors partly obtained in the MIL 2 project (Ramdahl et al., 1983). The total PAHs include 22 compounds from acenaphthene to benzo(ghi)perylene. The emission factors and the total average winter half-year emission rates are given in Table 26.

Table 26: Estimated emissions of total PAHs (22 compounds from acenaphthylene to benzo(ghi)perylene) in Oslo during the winter half-year.

| Fuel                       | Source               | Emission factors           | Average winter emission rate (gPAH/h) |
|----------------------------|----------------------|----------------------------|---------------------------------------|
| Coal/coke                  | Space heating        | 60 g/tonnes                | 89                                    |
|                            | Industrial processes | " "                        |                                       |
| Wood                       | Space heating        | 40 g/tonnes                | 343                                   |
| Fuel oil                   | Space heating        | 1-10 <sup>1</sup> g/tonnes | 60-590 <sup>1</sup>                   |
|                            | Medium size boilers  | 0.15 "                     |                                       |
|                            | Industrial processes | 0.02 "                     |                                       |
|                            | Large power plants   | 0.01 "                     |                                       |
| Gasoline<br>Diesel<br>fuel | Vehicular<br>traffic | 1.84 mg/km <sup>2</sup>    | 1456                                  |
|                            |                      | 4.0 " <sup>2</sup>         | 300                                   |

<sup>1</sup> assumed high PAH content in heavy duty oil

<sup>2</sup> emission factors for total PAHs were assumed high for winter season (Larsen, 1984).

The background concentrations, due to long-range transport and regional influences, were assumed during the winter half-year to be about  $20 \text{ ng/m}^3$ .

The relative contributions from the selected source groups depend largely upon the assumptions of PAH content in the fuel oil used for space heating. To demonstrate the variation, the estimates were carried out for both light ( $1 \text{ g PAH/tonne}$ ) and for heavy fuel oil ( $10 \text{ g PAH/tonne}$ ). The winter average concentration distribution with heavy oil for space heating is shown in Appendix B, and the number of people subjected to given concentrations for both alternatives is given in Figure 18.

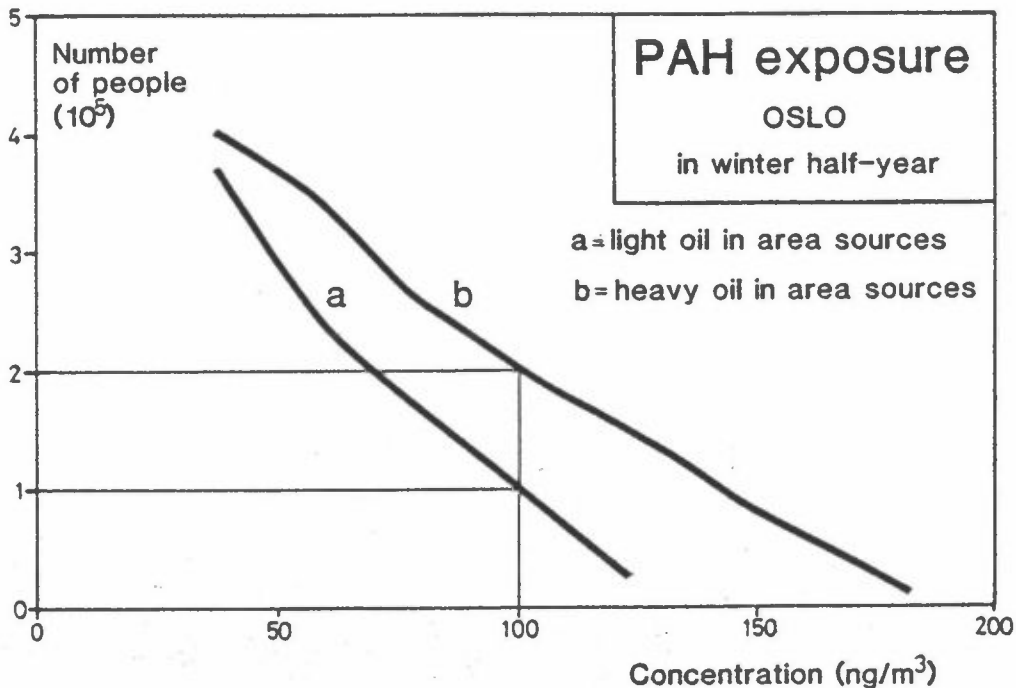


Figure 18: Number of people subjected to mean winter half-year PAH concentrations (given along the abscissa) from all PAH sources, including background.

The estimates, including 22 compounds of gaseous and particulate PAHs, show that assuming light fuel oil usage by "area sources" and space heating, 100 000 people were exposed to concentrations in excess of  $100 \text{ ng/m}^3$ . Heavy fuel oil use by "area sources" and space heating (Figure 18) exposes 200 000 people to concentrations exceeding  $100 \text{ ng/m}^3$ . Measured PAH concentrations in Oslo are closer to the latter alternative.

Integration of the distribution functions (as shown in Figure 18) for selected source groups, gives the relative source contributions to total population exposure in Oslo. The result of such estimates for heavy fuel oil for space heating (case b in Figure 18) is shown in Figure 19.

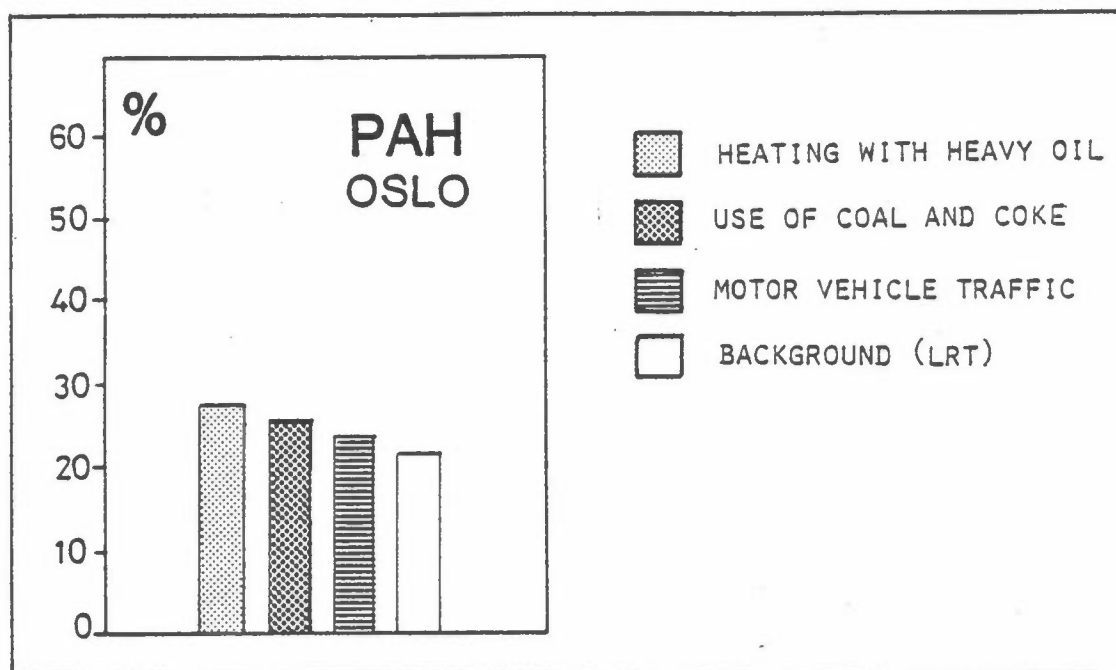


Figure 19: The relative contributions (%) from selected source categories to the "inhalation" exposure of 22 PAH compounds of the Oslo population (winter half-year, assuming 10 g PAH/tonne emission from space heating).

Assuming that heavy fuel oil, with an emission factor of 10 g PAH/tonnes, is used for space heating, the contributions from the 4 selected source categories are of about the same magnitude. For this alternative, space heating with heavy fuel oil give the largest contribution, 28%, while background and long-range transport amount for about 21%. If only light fuel oil (with emission rates of 1 g PAH/tonne) is assumed to be used for space heating, the contribution from these sources would only be about 4%. This shows how sensitive the modeling results are to the assumptions of emission rates.

Earlier measurements of PAHs in Oslo, used for simple receptor model estimates indicated that the contribution from the traffic to the total PAH load in a street location during daytime was more than 85%, and during nighttime 48 to 72%. At a sampling station not directly influenced by traffic, the regional contribution from traffic was estimated during daytime to be 29 to 43%. During low nighttime temperatures in winter, the contribution from space heating increased considerably also at the street location. These findings confirm the estimates performed for MIL 4, that traffic contributions on a kilometer scale amounts to about 25 to 30% of the total PAH load, while the contributions of space heating and the combustion of fuel oil and coal vary considerably, depending on the assumed emission rates (total contribution from 35 to 55%).

Several uncertainties appears in the population exposure estimates to PAHs in these first estimates performed for Oslo. The selection of PAH compounds in the total PAH estimate is important, and has been based upon definitions given in the MIL 2 project (Ramdahl et al., 1984). The PAH content in air also varies considerably with the ambient air temperatures. These variations cannot be explained by dispersion conditions alone, but must also reflect variations in emission factors. As long as emission factors have to be applied in estimates of this kind, there is a need for improving the accuracy and reliability of these factors in the future.

## 8.2 SOURCES OF NO<sub>2</sub> DURING EPISODES IN GÖTEBORG

Traffic was estimated to represent the dominating source for NO<sub>2</sub> concentrations, measured in central Göteborg during air pollution episodes. The total contribution from traffic amounted to ~68% and Long-range transport and background NO<sub>2</sub> to ~23%.

Measurements of air quality in central parts of Göteborg, from 1975 to 1983, formed the basis for estimating the contributions of nitrogen oxides from

different sources, and the importance of episodes to the annual mean  $\text{NO}_2$  concentrations.

Typical time variations of  $\text{NO}_x$  concentrations have been analysed, and correlation analyses between  $\text{NO}_x$ ,  $\text{NO}_2$  and  $\text{SO}_2$  performed. It was found that air pollution episodes contribute about 2.7% to the total annual exposure of  $\text{NO}_2$  in central Göteborg. To estimate which specific sources contributed to the  $\text{NO}_2$  concentrations, 35 days were selected. During these days the  $\text{NO}_x$  concentrations exceeded about  $1200 \mu\text{g}/\text{m}^3$  (as  $\text{NO}_2$ ). The episodes occurred mostly during the winter half-year, and mainly around the rush hours in the morning and in the afternoon. The  $\text{NO}_2$  concentrations were assumed to originate from primary emissions of  $\text{NO}_2$ , reaction mechanisms and long-range transport of  $\text{NO}_2$  from sources outside Göteborg. The following expression was studied:

$$[\text{NO}_2] = A + B[\text{NO}] + E$$

where: A = long-range transport of  $\text{NO}_2$  and background

B = proportion of primary emitted  $\text{NO}_2$ , relative to NO

E =  $\text{NO}_2$  produced by reactions in the atmosphere

The contribution from atmospheric reactions, E, is insignificant during the morning hours, but has a maximum in the afternoon and in the evening. By adding the contributions for each hour when  $\text{NO}_x$  is larger than  $1200 \mu\text{g}/\text{m}^3$  (as  $\text{NO}_2$ ), the total contribution of  $\text{NO}_2$  from atmospheric reactions is obtained. This contribution varies from day to day, and averages to about 7%.

The contribution from background pollution and long-range transport, A, varies from 11% to 48% for each day. The rest of the  $\text{NO}_2$  was assumed to come from traffic and heat/power production sources, and estimated to be between 30 and 90%. To estimate the contribution from traffic, relative to heat and energy sources, a receptor model approach was applied. The ratio  $\text{SO}_2/\text{NO}_x$  for each hour was used. It was assumed that the proportion of primary emitted  $\text{NO}_2$ , B, is the same for traffic as for other combustion sources.



By estimating the average values of A, B and E for all episode days, it is possible to establish the relative contributions from the different source categories, as shown in Figure 20.

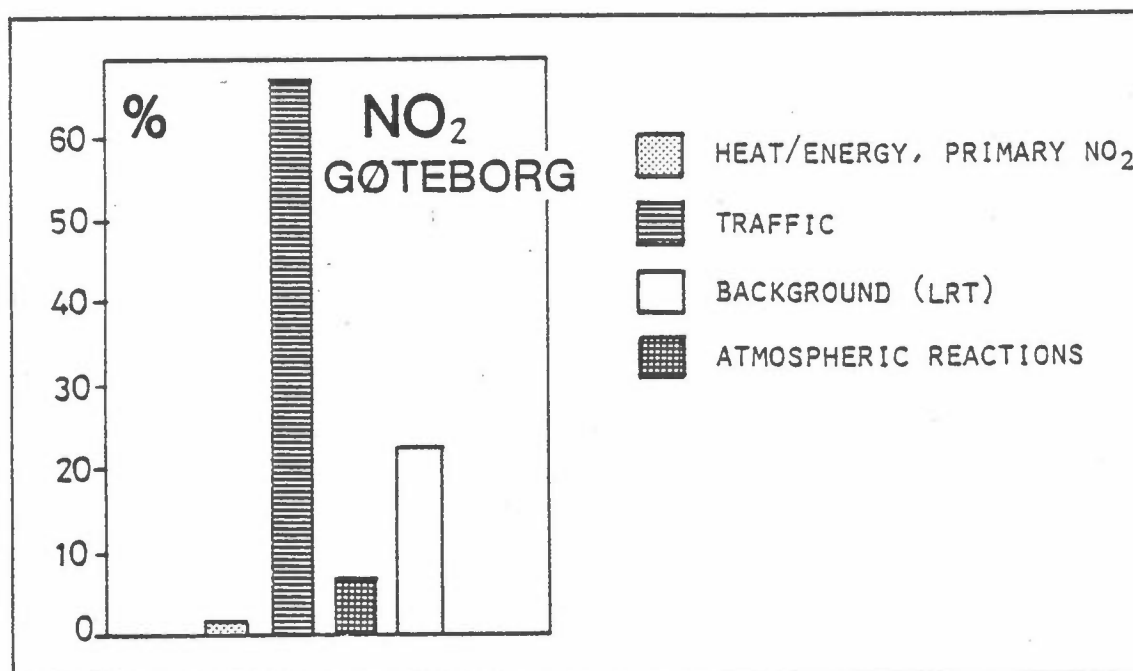


Figure 20: Estimated relative contributions (%) from selected source categories to the NO<sub>2</sub> concentrations in central Göteborg during episodes (when  $[\text{NO}_x]^2 > 1200 \mu\text{g}/\text{m}^3$ , as NO<sub>2</sub>).

Traffic is the dominating source of NO<sub>2</sub> concentrations in central Göteborg. This accounts for both primary emitted NO<sub>2</sub>, and NO<sub>2</sub> formed by chemical reactions. The total contribution from traffic amounts to 68%. Long-range transport and background NO<sub>2</sub> account for 23% of the total NO<sub>2</sub> concentrations during episodes (Galle, 1984).

### 8.3 ATMOSPHERIC TRANSPORT OF METALS TO SCANDINAVIA

With transport of air masses from sectors to the south and south east of background stations in southern Scandinavia, long range transport seems to account for 70-80% of the accumulation of compounds such as Pb, Cd, and As in moss samples. Annual deposition at typical locations is presented.

The deposition of metals (V, Cr, Mn, Fe, Co, Ni, As, Cd, Se, Sb, Pb) to background areas in southern Scandinavia was studied for 1980 (Steinnes, 1984). Samples of live mosses were collected at different sites in Scandinavia and their content of the metals analyzed. Precipitation data were used to estimate wet deposition contributions. The Swedish data presented some problems, as the measurements were made by different institutions over short periods. In Norway, most of the work was carried out by Steinnes and NILU. The Norwegian samples were collected mostly during 1977, and spatial concentration distributions of several elements are available (Rambeck and Steinnes, 1980). Similar investigations were performed in 1975 and in 1980 in Sweden (Rühling, 1982), and in 1980 in Denmark (Gydesen et al., 1983).

An attempt was made to quantify the total contribution of long-range atmospheric transport to the deposition of trace metals in southern Scandinavia, to assess the importance of the background component in relation to the other air pollution sources. The results reflect the situation around 1980. Typical values for the deposition in southern Norway (Sørlandet), southern Sweden (Båhuslän), and Denmark (eastern Jylland) are presented in Table 28.

Table 28: Typical deposition of trace elements at background stations in the Nordic countries ( $\text{mg m}^{-2} \text{y}^{-1}$ ).

| Element | Norway<br>(Sørlandet) | Sweden<br>(Båhuslän) | Denmark<br>(Jylland) |
|---------|-----------------------|----------------------|----------------------|
| Pb      | 10                    | 6                    | 7                    |
| Cd      | 0.3                   | 0.2                  | 0.2                  |
| As      | 0.5                   | 0.3                  | -                    |
| Ni      | 0.6                   | 0.4                  | 0.7                  |

A sector analysis of data from Birkenes in southern Norway shows that long-range transport of lead from the south and southeast accounts for 70-80% of the deposition. The Norwegian and the Swedish data on vanadium in moss are not consistent. The Norwegian data show a typical long-range transport pattern (about 80%), while the Swedish data indicate a regional influence. The same can also be said for the nickel deposition. For cadmium and lead, all the Scandinavian data are consistent, and there exist enough precipitation data to verify the general pattern. A sector analysis, performed on data from Välen (Shaw 1981), indicates that in the southeastern sector the long-range transport contributed up to 10 times more than the local sources. Lannefors et al. (1983) found that S, Cu, Zn, and Pb levels increased substantially in air masses having traversed Great Britain or the European continent, when compared with those in northerly air masses. The "foreign" contributions to S and Pb were estimated to about 75%, while those of V and Ni were on the order of 25 and 50%, respectively, of the total deposition.

In the future, it is desirable to obtain more data on trace metals in precipitation. There is also a need for establishing better knowledge of conversion factors between trace element concentrations in moss and deposition values.

#### 8.4 DRY DEPOSITION IN URBAN ENVIRONMENTS

When modeling exposures to heavy metals, it is necessary to have a suitable parameterization of the deposition processes. The knowledge of the movements of aerosols in urban environments is limited. Most data on deposition concern vegetated surfaces. There is, however, some experimental evidence that deposition to smooth surfaces in the city might be somewhat smaller than in vegetated areas. On the other hand, resuspension of deposited material during windy conditions might cause increased airborne aerosol concentrations. To obtain information about these matters, a special sub-project of MIL 4 was established (Jensen 1984). The aims of this sub-project were to:

- give an overview of existing knowledge on this topic,
- recommend methods to improve the knowledge (e.g., can deposition/resuspension be parameterized?)
- recommend how experimental data can be improved and experiments conducted to investigate this problem in the Nordic countries.

A literature study revealed that very little, if any, data exists on dry deposition of fine particles to city surfaces. The classical paper by Sehmel (1980) for example, gives no references on deposition to smooth surfaces. A study of deposition velocities of cesium-137 to building surfaces (Roed, 1983) indicated that the deposition velocity was smaller than the values usually found in the literature (deposition velocity,  $v_d = 0.03$  cm/s).

Resuspension of deposited material can be described by a resuspension factor,  $K = C_a / C_0$ , which is the ratio between air concentrations ( $C_a$ ) and the amount deposited ( $C_0$ ).  $K$  is dependent upon wind velocity and size of the surface roughness elements. Resuspension can be caused by the wind and by the air movement induced by traffic. There is a lower limit for wind speeds that can cause particles to be resuspended, which depends upon particle size and surface roughness (threshold drag velocity). The effect of traffic on resuspension is dependent upon the driving speeds. Measurements indicate that the resuspension rate can increase by a factor of 10, as the speed of the vehicle increase from 15 to 80 km/h.

Methods for the study of the importances of the resuspension in urban environments have been suggested. Size-fractionated high-volume particle samples should be analysed for a number of elements to give quantitative information about resuspension as a function of wind velocity ( $u$ ). For low wind velocities, the ambient air concentration of particulate matter might be proportional to  $1/u$  (no resuspension, only dispersion). For high wind velocities, the ambient air concentrations might be proportional to  $u^2$  (flux of the suspended matter increase thus with  $u^3$ , and the dispersion with  $1/u$ ). The fraction of the flux actually going into suspension is sometimes simply assumed to be some fixed percentage, but in general it must depend on the relative fractions of small and large particles on the surface. The expected

air concentrations of particulate matter, as a function of wind speed, are illustrated in figure 21.

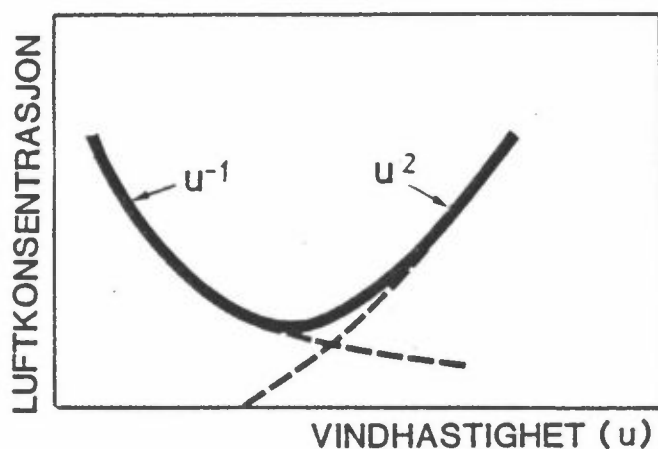


Figure 21: Expected ambient aerosol concentrations in a typical urban environment, as a function of wind speed.

Particle resuspension by moving vehicles is caused by air turbulence in their wake, and the direct mechanical forces exerted by the tires. Both mechanisms can be assumed to contribute to the measured air concentrations. The relative contributions can not be sorted out directly. The sub-project report (Jensen 1984) gives a general ideas of how a suitable, full-scale experiment should be designed to generate some data on the problems of dry deposition to typical surfaces in the city.

## 9 Future Nordic cooperation based upon MIL 4

During the work of the different sub-projects and in discussions at MIL 4 seminars, a number of unsolved problems have surfaced. Several of these may be resolved by future cooperative Nordic projects. Such projects might include the following assessments:

- 1) Uncertainties in input data, parameters and methods.
- 2) Gaps in knowledge about physical and biological constants, indoor/outdoor concentration ratios and parameterization of processes.
- 3) Limitations in existing methods, such as combination of different models and consequences of inadequate data inputs.
- 4) Dissemination and application of results.

The limitations and uncertainties in the different sub-projects of MIL 4 have already been mentioned in each chapter of this report. The following is, therefore, only a brief summary of some of the problem areas that the project group in MIL 4 feels can be solved through future Nordic co-operation.

#### 9.1 UNCERTAINTIES

The possibility of quantifying the effects of variability in input/output data and the uncertainties in model estimates is today lacking in most investigations. Only in one sub-project of MIL 4 was a statistical analysis performed to study the consequences of the variability in input and output data. The other sub-projects, without resorting to quantitative assessments, have nevertheless pointed to uncertainties in:

- emission data (suburban traffic in Stockholm, PAH emissions in Oslo, source composition);
- exposure estimates in urban areas (inadequate spatial resolution);
- outdoor/indoor concentration ratios;
- physical and biological parameters in exposure models;
- the effect of deposition and resuspension (when filter analyses are performed for urban areas);
- estimates of NO<sub>2</sub> formation by chemical reactions in the atmosphere.

## 9.2 GAPS IN KNOWLEDGE

At the beginning of the MIL 4 project, some possible sub-projects had to be deleted because of a lack of necessary knowledge in particular areas. Some of these tasks could not be solved properly within the time and economical resource constraints of MIL 4. This was the case for nitrate deposition, photochemical oxidants, and mercury exposures. Research in these fields is being conducted under other projects in the Nordic countries. The lack of basic information in several other fields was also revealed during some of the MIL 4 project. Some of these represented a limiting factor in the performance of the sub-projects, as for instance:

- emission factors for different components from various sources during different climatological conditions;
- population data on typical movement patterns and consumption of foodstuffs;
- exposure in cars and buses on busy city streets;
- data on toxic metal concentrations in different environments (e.g., air, water, soil, foodstuffs).
- better source composition data ("fingerprints").
- $\text{NO}_x$  to  $\text{NO}_2$  transformation in source emissions and in the atmosphere;
- deposition and resuspension of aerosols in urban environments;
- quantitative information about uptake mechanisms and retention times for toxic metals and PAHs in biological material;
- better information about dose-response relationships and health effects related to exposure time;
- reactions between PAH compounds and other air pollutants.

In exposure modeling, only human subjects have been considered as targets. Exposure of plants and materials to  $\text{SO}_2$  and  $\text{NO}_x$  were not included, because the necessary inventories were not available. It would, however, be possible to make exposure estimates for receptors other than humans, if knowledge on the distributions of materials or vegetation were available.

### 9.3 LIMITATIONS OF METHODS

In some of the exposure estimates and source apportionments, the quality and consistency of the results were limited by the available methods and the models. For the source-oriented exposure models these limitations included:

- a combination of kilometer-scale grid modeling and modeling in street canyons (the spatial resolution of the models);
- modeling the movement and distribution of people with the same spatial resolution as for air quality modeling;
- modeling of indoor exposure;
- the simplicity of quasi-stationary box models for modeling exposures. (In the future these models should be further developed, and the transfer factors should be based upon actual data from the Nordic areas.)

For the receptor models the following limitations included:

- the ambient air concentration measurements were originally not designed for use in receptor models. Consistency in sampling and analysis is therefore inadequate and important parameters are lacking;
- meteorological variations and chemical reactions are not included in the receptor models;
- in some of the projects, the source characterizations were based upon literature studies.
- Combined use of factor analysis, principal component analysis, and mass balance studies has been lacking.

### 9.4 POSSIBLE FUTURE TASKS FOR NORDIC COOPERATION

In view of the above mentioned uncertainties, gaps in knowledge and limitations, the project group of MIL 4 can suggest a number of tasks suitable for future Nordic cooperation.



The following list gives the suggested research topics in descending order of priorities (as perceived by the MIL 4 group):

- (1) Development of better exposure models, where indoor/outdoor air quality estimates can be linked to population distributions and living patterns on all scales:
  - a) indoor/outdoor air exposures
  - b) better data on living and movement patterns
  - c) information about the importance of  $\text{NO}_x$  exposures in streets and along roads, as compared to the kilometer-scale grids.
- (2) Better data for establishing transfer factors in exposure models for toxic metals.
- (3) Parameterization of  $\text{NO}_2$  formation in urban areas (including  $\text{NO}_x/\text{NO}_2$  relationships in source emissions).
- (4) Deposition/resuspension on/from smooth surfaces in urban environments of:
  - a) size-segregated aerosols,
  - b) gaseous air pollutants ( $\text{SO}_2$ ,  $\text{NO}_x$ ).
- (5) Improved quality of PAH exposure estimates for selected cities from:
  - a) better emission data for PAHs,
  - b) inclusion of chemical reactions between PAHs and other air pollutants,
  - c) as in items 1a), 1b), and 1c) above.
- (6) Development of a Nordic "catalogue" for chemical composition ("source fingerprints") of aerosols and PAHs from different source categories in the Nordic countries.

- (7) Further development of exposure modeling for materials (atmospheric corrosion) and vegetation.
- (8) Further studies of concentrations of toxic metals and PAHs in air, soil, and in vegetation, to obtain information on:
  - a) retention times,
  - b) effects of pH and climatic variables,
  - c) effective biological half-lives (scavenging, growth, harvesting).

## 10 Summary results

This report contains the main results from the work carried out in 15 sub-projects within MIL 4. The methods and the models have been briefly presented in this report. More details and background references for all the sub-projects can be obtained from reports listed under References.

In an earlier introduction to the project in 1979, it was pointed out that the available basis for establishing priorities in air pollution abatement and environmental legislation was inadequate. To establish emission reduction strategies for the in energy-producing sector, it is necessary to have knowledge about the contributions from other source categories in a given area.

One of the main objectives of MIL 4 was to quantify the environmental impact of different forms of energy production, in relation to the impacts from other sources.

During the development and conduct of the project, the different sub-projects of MIL 4 have endeavored to give a clear picture of the relative importance of the different source categories. Keeping in mind the reservations, uncertainties and limitations discussed in Chapter 9, Table 29 summarizes the estimated relative contributions from the different energy production categories.

Table 29: A summary of estimated relative contributions (in %) from different selected source categories to air pollution exposures in selected areas in the Nordic countries. (The same results are also summarized in Figure 21.)

| Component          | Region                                     | Source categories           |                    |                                   |                 |  |            |
|--------------------|--|-----------------------------|--------------------|-----------------------------------|-----------------|--|------------|
|                    |  | Energy/<br>space<br>heating | Vehicle<br>traffic | Indust-<br>rial<br>pro-<br>cesses | LRT<br>)        | Others<br>)                              | Model<br>) |
| SO <sub>2</sub>    | Helsinki                                   | 48                          | 0                  | 6                                 | 46              | -  | S          |
|                    | Oslo                                       | 74                          | 5                  | 0                                 | 21              | -  | S          |
|                    | Stockholm                                  | 81                          | -                  | 3                                 | 16              | -  | S          |
| NO <sub>x</sub>    | Helsinki                                   | 25                          | 54                 | 1                                 | 20              | -  | S          |
|                    | Oslo                                       | 20                          | 65                 | 0                                 | 16              | -  | S          |
|                    | Stockholm                                  | 43                          | 31                 | 1                                 | 25              | -  | S          |
| NO <sub>2</sub>    | Göteborg<br>(episodes)                     | 2                           | 68                 | -                                 | 23              | 7  | R          |
| Metals             | Pb Sarsborg-<br>Fredrikstad                | 3                           | 38                 | 14                                | 26 <sup>2</sup> | 24 <sup>4</sup>                          | S          |
|                    | Cd Sjælland                                | 1                           | -                  | -                                 | 61 <sup>2</sup> | 37 <sup>4</sup>                          | S          |
| Pb<br>Cd<br>Ash+   | Helsinki park                              | 20                          | 17                 | -                                 | 26              | 37 <sup>1</sup>                          | R          |
|                    | " "  | 6                           | 17                 | -                                 | 26              | 37 <sup>1</sup>                          | R          |
|                    | " "  | 30                          | 30                 | -                                 | 38              | 2 <sup>1</sup>                           | R          |
| TSP                | København,<br>City center                  | 18                          | 34                 | -                                 | 22              | 26 <sup>3</sup>                          | R          |
|                    | " København:<br>Glostrup                   | 38                          | 51                 | -                                 | 41              | 20 <sup>3</sup>                          | R          |
|                    | " Southern Norway                          | 24                          | 29                 | 5                                 | -               | 42 <sup>1</sup>                          | R          |
|                    | Örebro, winter<br>" , summer               | 2<br>2                      | 4<br>2             | -<br>-                            | 60<br>~75       | 34-92 <sup>3</sup><br>14-26 <sup>3</sup> | R<br>R     |
| PAHs in air        | Oslo                                       | 40-55                       | ~23                | 2                                 | 21              | -  | S          |
| PAHs in air        | Sundsvall                                  | -                           | -                  | 46-85                             | -               | -  | R          |
|                    | Ardal                                      | -                           | -                  | 76-90                             | -               | -  | R          |
|                    | Mosjøen                                    | -                           | -                  | 46-64                             | -               | -  | R          |
| PAHs in<br>cabbage | Göteborg:<br>street<br>residential<br>area | -                           | 95                 | -                                 | 5               | -  | R          |
|                    |  | -                           | 49                 | -                                 | 51              | -  | R          |

\*) <sup>1</sup> = Waste inciner.  
<sup>2</sup> = + fertilizers  
<sup>3</sup> = Soil  
<sup>4</sup> = Other food

\*\*) S = Source-oriented model  
R = Receptor model  
+ = Presumably representative of TSP

Different air pollutants, different areas, variations in the quality of input data and computational methods might result in different estimates of the relative contributions. Thus, the results from one area and for one set of component should be applied with care to another area, where different set of components exists.

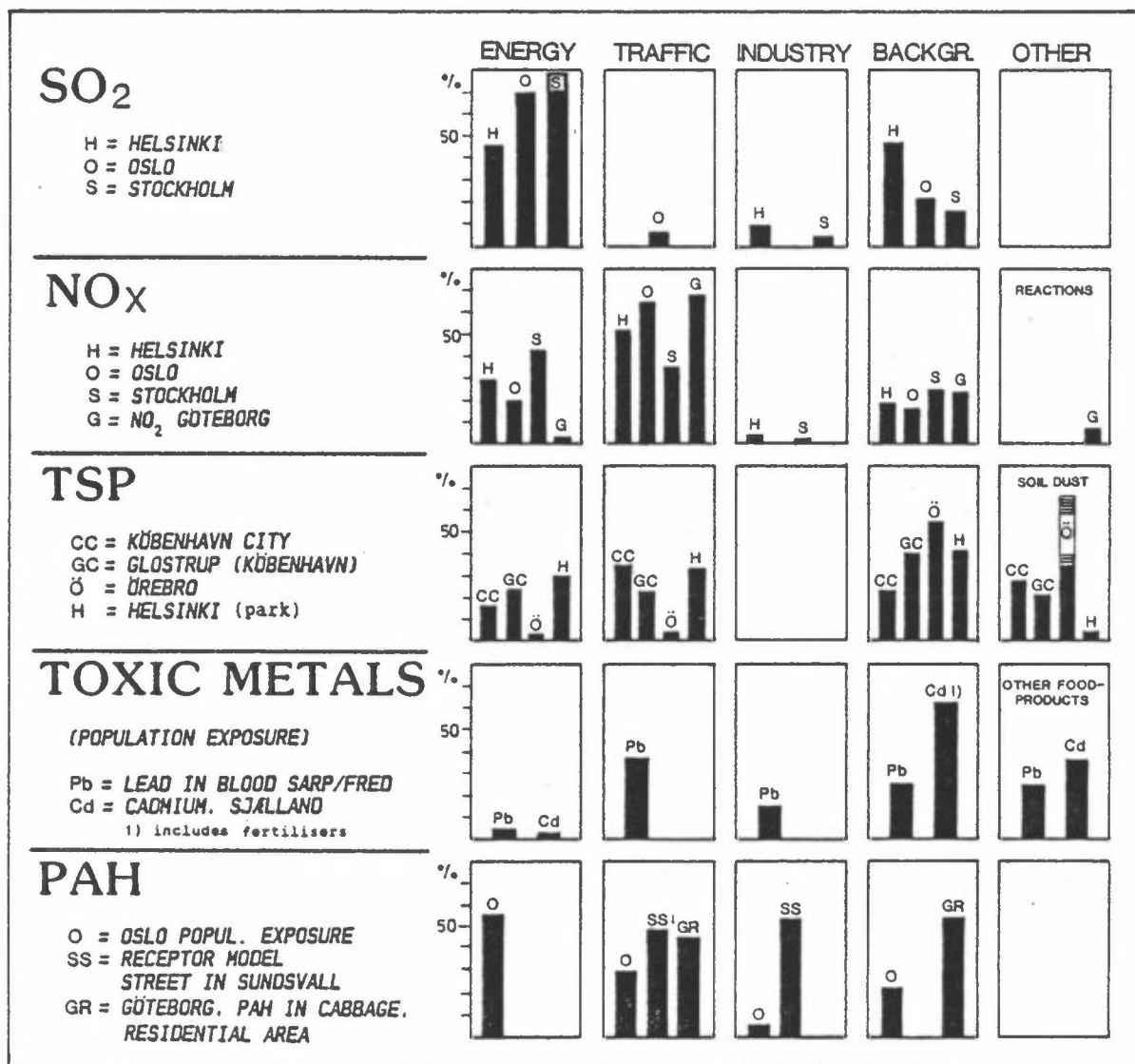


Figure 21: A summary of estimated relative contributions (%) from selected source categories for different areas of the Nordic countries selected in MIL 4.

In summary, then the work within MIL 4 has shown that:

- SO<sub>2</sub> exposures to the populations of the Nordic capitals Helsinki, Oslo, and Stockholm are mainly due to heat and energy production (48-81%).
- NO<sub>x</sub> exposures of the populations of Helsinki, Oslo, Stockholm, and Göteborg are mainly due to vehicular traffic (about 31-80%).
- the relative contribution to the exposures of toxic metals is dependent on the metal and the area considered; for example in the Sarpsborg-Fredrikstad area, traffic is causing about 40%, and long-range transport about 26% of the population exposure to lead; in Sjælland 60% of cadmium exposure is due to long-range transport and the use of fertilizers.
- total suspended particulate matter, collected on filters, originates from energy sources (about 20%), traffic (dependent upon sampling location, but about 30% on average), and long-range transport and background air pollution (about 40% average).
- the estimated population exposure to PAHs in Oslo is attributed to coal and fuel oil combustion (50%), traffic emissions (24%) and "background" or long-range transport (21%).

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## APPENDIX A

Participating institutions



## PARTICIPATING INSTITUTIONS

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## APPENDIX B

Average concentration distributions during the winter half-year  
in Helsinki, Oslo and Stockholm of:

B1: SO<sub>2</sub> (µg/m<sup>3</sup>)

B2: NO<sub>x</sub> (as NO<sub>2</sub>) (µg/m<sup>3</sup>)

B3: PAHs in Oslo (ng/m<sup>3</sup>)





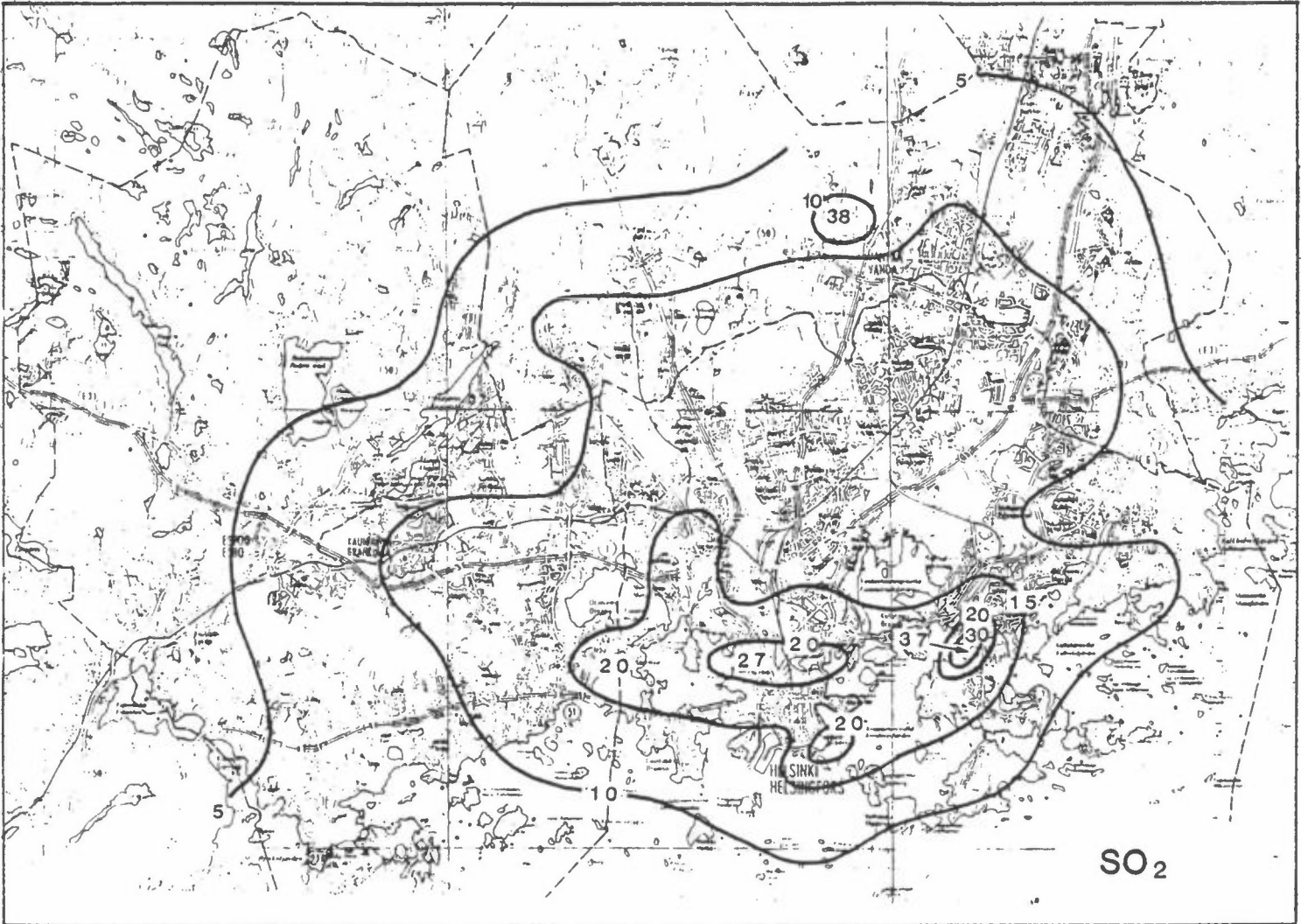


Figure B1a: Calculated winter half-year means of SO<sub>2</sub> concentrations in Helsinki (µg/m<sup>3</sup>) (background concentrations are not added).

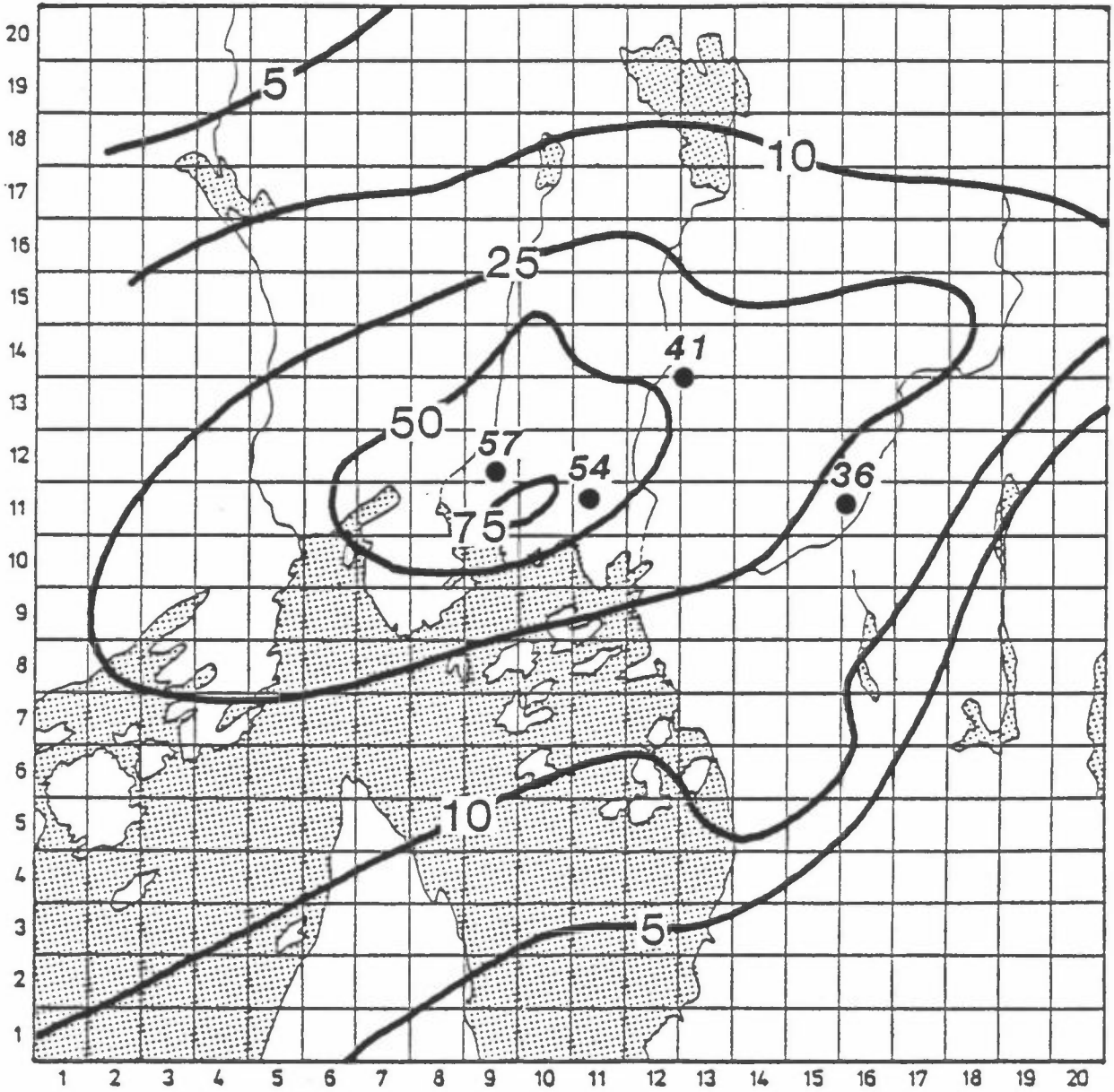


Figure B1b: Calculated winter half-year means of SO<sub>2</sub> concentrations in Oslo (µg/m<sup>3</sup>). (The background level of 10 µg/m<sup>3</sup> is not added.)  
 ● = observations.

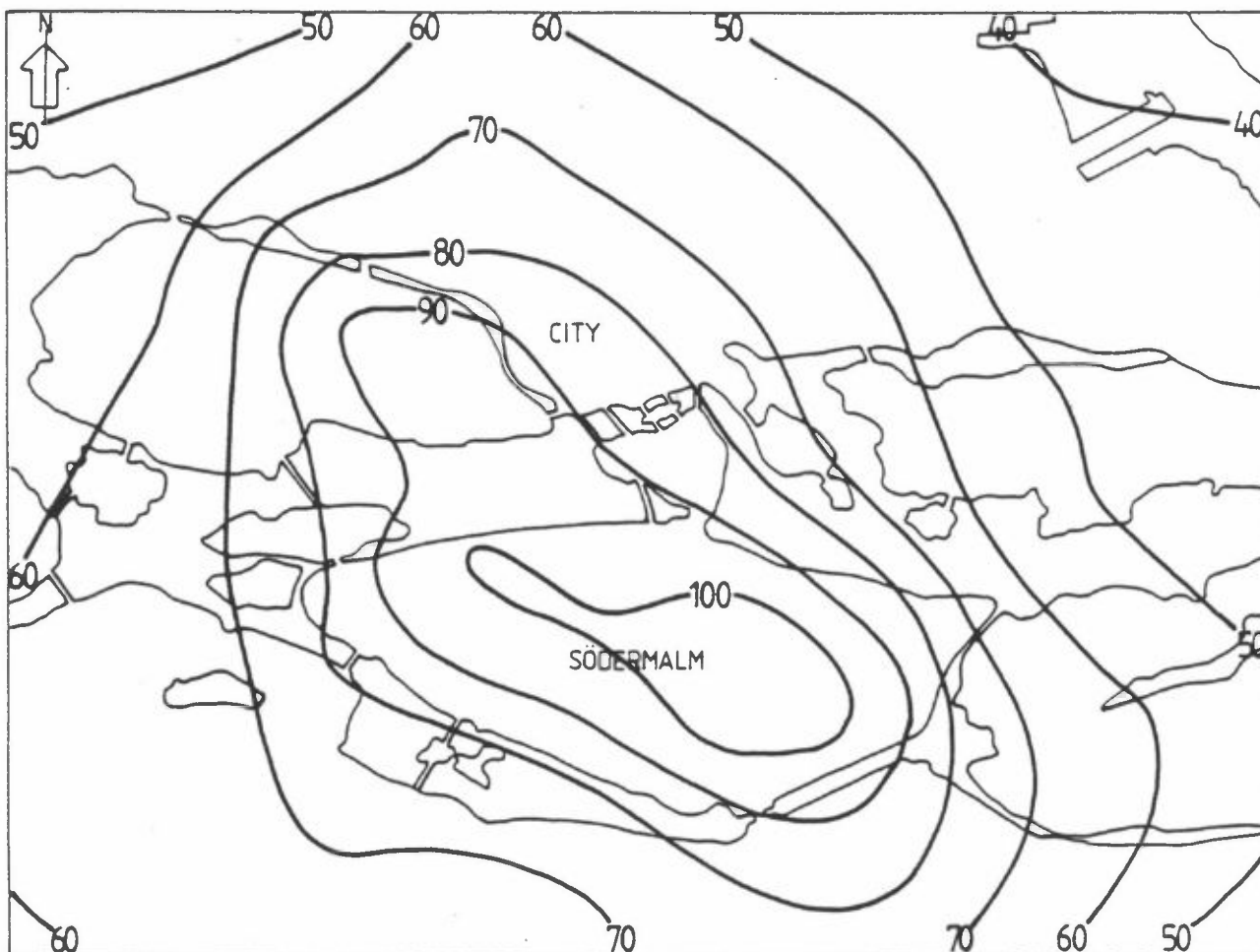


Figure B1c: Calculated winter half-year means of SO<sub>2</sub> concentrations in Stockholm ( $\mu\text{g}/\text{m}^3$ ).

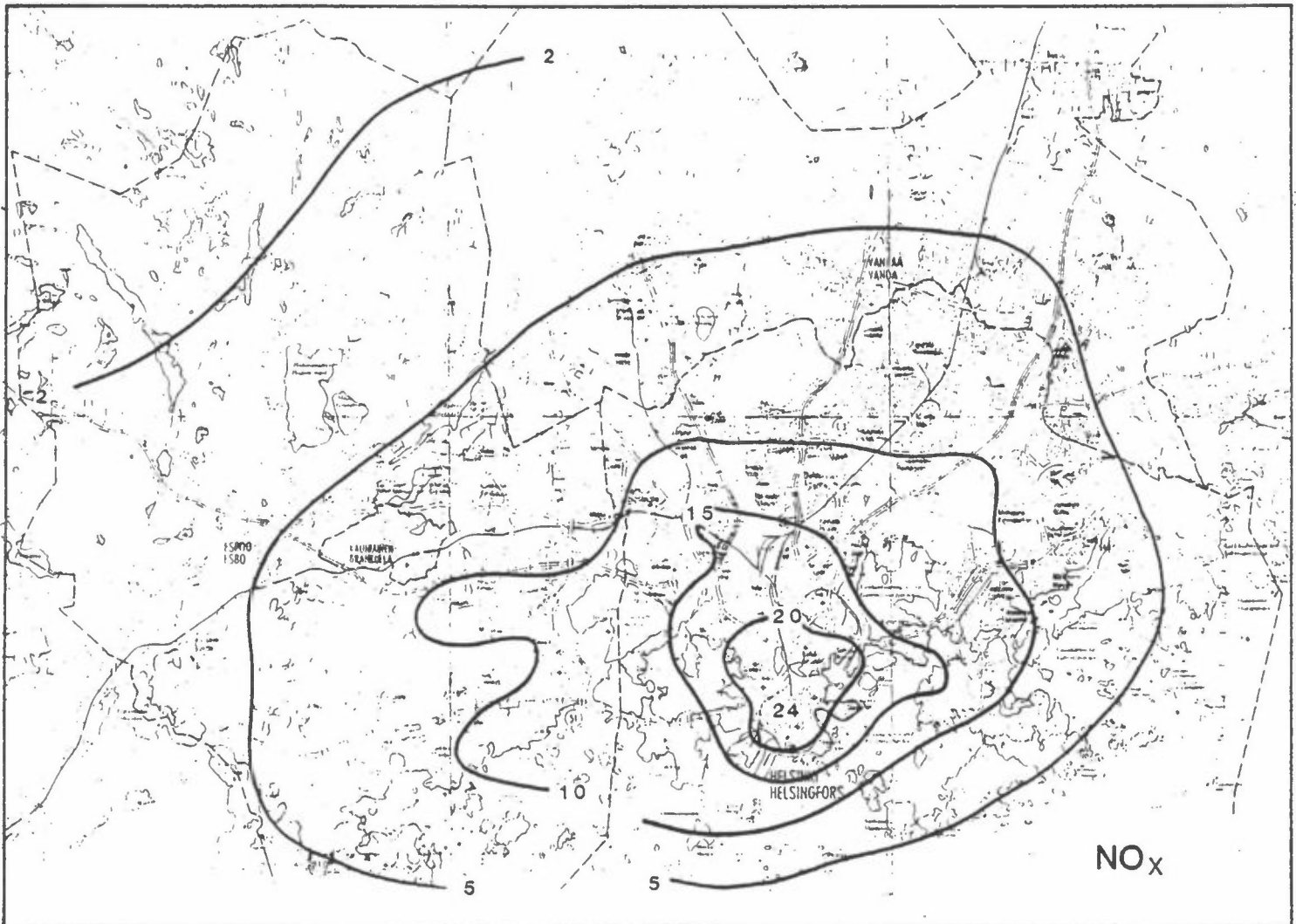


Figure B2a: Calculated winter half-year means of NO<sub>x</sub> as NO<sub>2</sub> concentrations in Helsinki (µg/m<sup>3</sup>).



Figure B2b: Calculated winter half-year means of NO<sub>x</sub> (as NO<sub>2</sub>) concentrations in Oslo (μg/m<sup>3</sup>).

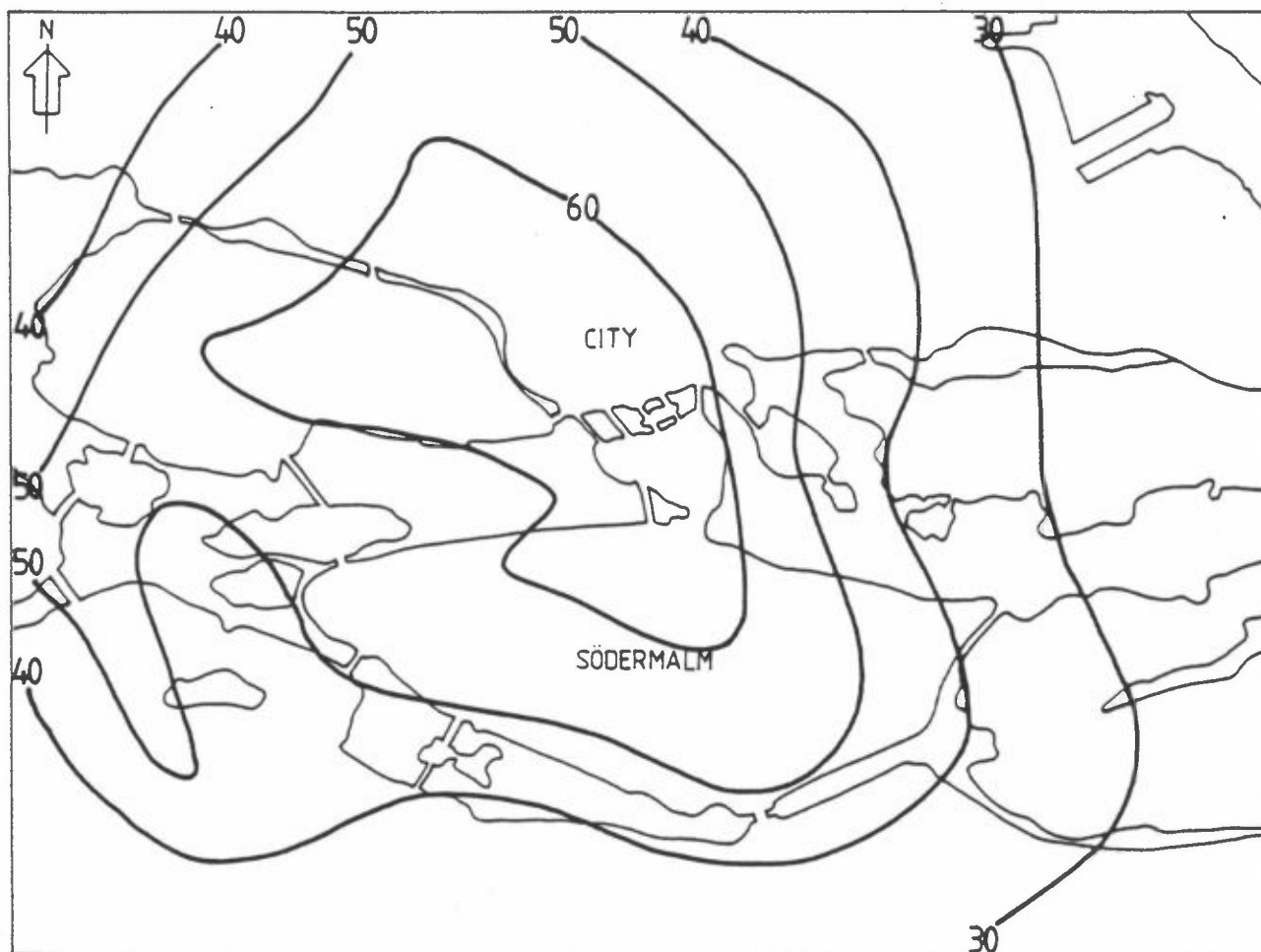


Figure B2c: Calculated winter half-year means of NO<sub>x</sub> as NO<sub>2</sub> concentrations in Stockholm (µg/m<sup>3</sup>).

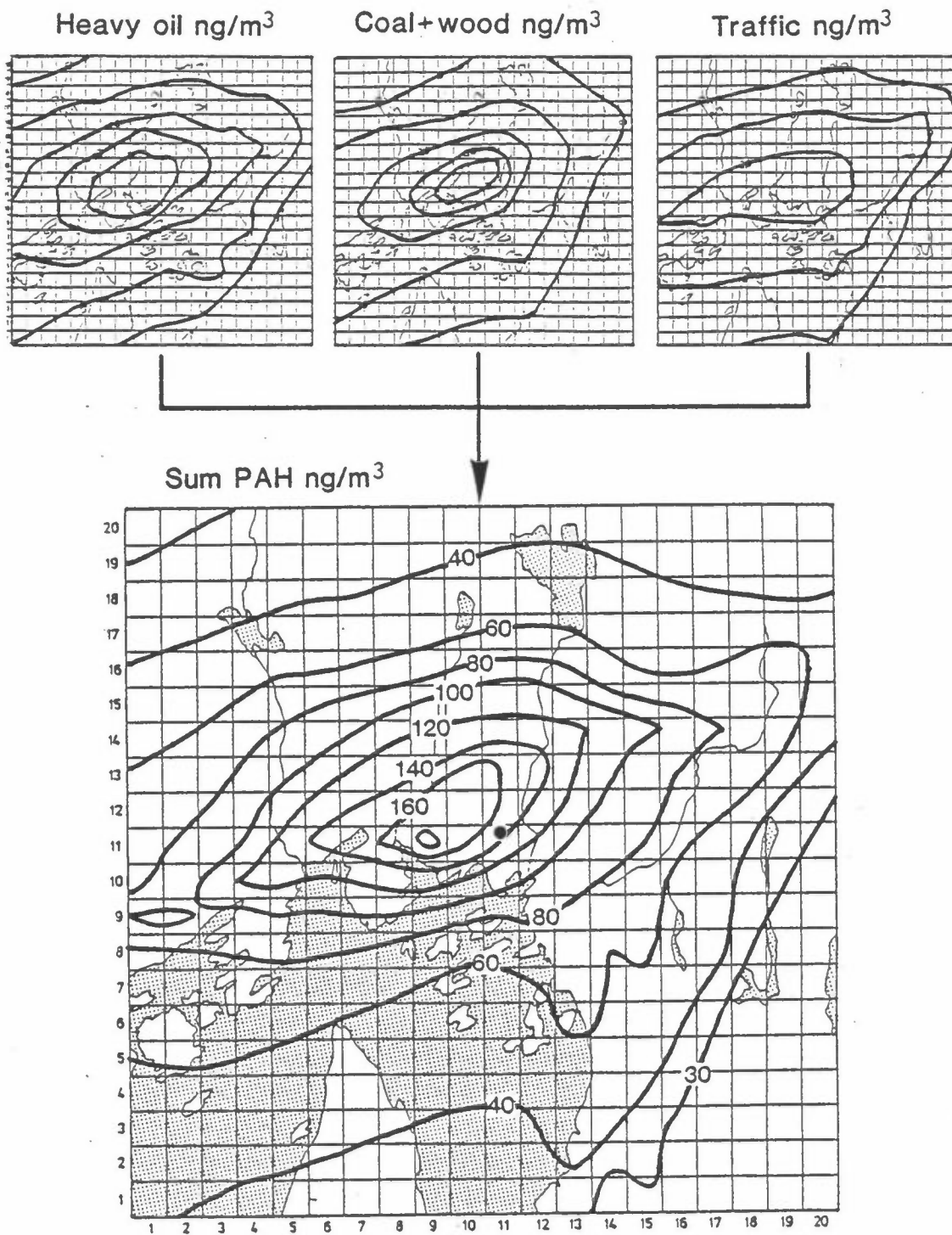


Figure B3: Calculated, winter half-year means of total PAH concentrations in Oslo ( $\text{ng}/\text{m}^3$ ), assuming that heavy duty oil was used for space heating (area sources).

● = observed concentrations  $\sim 200 \text{ ng PAH}/\text{m}^3$ .



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POSTBOKS 130, 2001 LILLESTRØM (ELVEGT. 52), NORGE

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|--|--------------------------------|-----------------------------------|-------------------|
| RAPPORRTYPE<br>Oppdragsrapport   | RAPPORTRNR.<br>OR 41/85        | ISBN-82-7247-603-7                |                   |
| DATO<br>August 1985  | ANSV. SIGN.<br><i>Storland</i> | ANT. SIDER<br>103                 | PRIS<br>Nkr 90,00 |
| TITTEL<br>Final report - MIL 4.<br>Relative contribution of air pollutants from various sources to man and the environment.  |                                | PROSJEKTLEDER<br>Bjarne Sivertsen |                   |
|  |                                | NILU PROSJEKT NR.<br>0-8052       |                   |
| FORFATTER(E)<br>Bjarne Sivertsen, (ed.)  |                                | TILGJENGELIGHET*<br>A             |                   |
|  |                                | OPPDRAGSGIVERS REF.               |                   |
| OPPDRAGSGIVER (NAVN OG ADRESSE)<br>Nordisk Ministerråd<br>Kontaktgruppen for nordisk miljøvernforskning<br>Box 6753, St. Olavs pl., Oslo 1   |                                |                                   |                   |
| 3 STIKKORD (à maks. 20 anslag)<br>Population exposure   Source apportionment   Receptor models   |                                |                                   |                   |
| REFERAT (maks. 300 anslag, 7 linjer)<br>Gjennom 15 delprosjekter i MIL 4 er kildenes relative bidrag til forurensningsbelastningen via luft belyst. SO <sub>2</sub> -eksponeringen til befolkningen i 3 nordiske hovedsteder skyldes fra 50-80% energi og varmeproduksjon. NOx-eksponeringen skyldes vesentlig trafikkutslipp (~60%), mens partikkel- og PAH-belastningen varierer fra et område til et annet. |                                |                                   |                   |

TITLE

ABSTRACT (max. 300 characters, 7 lines)  
The results from 15 sub-projects, conducted by various institutions in the Nordic countries, have elucidated the impact of energy production and conversion to the environment relative to other sources.

\*Kategorier: Åpen - kan bestilles fra NILU                   A  
                  Må bestilles gjennom oppdragsgiver        B  
                  Kan ikke utleveres                            C