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An Empirical Assessment of European Urban Ozone: Concentrations and Exposure

Summary report

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Preface

Norwegian Institute for Air Research (NILU) is a co-ordinator of a project "Inclusion of health impacts in assessment modeling of Air Pollution to support the 1997 NO_x protocol". The project has received funding from the Nordic Council of Ministers, the Ministry of Public Housing, Spatial Planning and Environment of the Netherlands, and the European Environmental Agency, and has been further supported by the World Health Organization European Centre for Environment and Health. The project is done in co-operation between NILU, International Institute of Applied Systems Analysis (IIASA), UNECE/RIVM Co-ordinating centre for effects, and the University of Kassel, during the years 1996 and 1997.

The development in the preparatory work for the nitrogen protocol negotiations led to a change in scope of the project, originally aimed at nitrogen dioxide (NO_2) exposure. Early in the project period, ozone was identified as more important compound than NO_2, with known and quantified effects of the ambient levels on human health.

NILUs contribution has been to construct an empirical model to evaluate European urban ozone concentrations and exposure to ozone in relation to emissions of ozone precursors NO_x and VOCs on the basis of available international measurement data. This report provides a summary of the results of NILUs work.

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Summary

The aim of the presented work is to investigate if the urban ozone concentrations throughout Europe can be described with simple empirical relationships using internationally available measurement data. The goal is to provide a European-wide description of urban ozone concentrations based on precursor emissions, that may be used for assessment of damage to human health under different precursor emission scenarios. One of the requirements is that the urban assessment of health damage is consistent with the assessment done for natural vegetation and crops.

Measured urban ozone concentrations were related to precursor emissions using a statistical relationship. Such empirical approach has been tried successfully before for sulphur dioxide, for nitrogen dioxide and for regional ozone.

A database was assembled of urban and regional ozone measurements, urban and regional NO_x and VOC emissions, regional EMEP ozone and nitrogen compound modelling results, and other information. The following types of empirical relationships were studied:

- Relationships between annual averages and annual 98th percentiles of hourly values of urban ozone on one side and urban and regional NO_2 concentrations, NO_x and VOC emissions, and regional ozone concentrations;
- relationships between short-term urban and regional ozone concentrations
- relationship between measured annual AOT60 and measured 98th percentile of hourly concentrations in urban areas.

Based on the collected data, we did not find any simple statistical predictions of peak ozone concentrations in terms of precursor emissions. Urban ozone could be related to regional ozone. We therefore suggest to employ the regional photochemical model also for cities, as the regional concentrations quite consistently predict the upper envelope of urban ozone levels. The important exception is, however, the formation of urban plumes. We did not find an adequate statistical model that would describe the urban plume formation based on our types of data. Therefore, our results do not provide any estimate of ozone levels in urban plumes.

With the current emissions, over 90% of the European urban population are exposed to peak values of ozone over the 8-hour air quality guideline (120 µg/m^3 or 60 ppb as 8-hour average). This result is derived using an exposure assessment based on the regional ozone estimates from the EMEP regional photochemical model. With a reduction of NO_x emissions in one individual city, the average exposure to ozone is likely to rise there.

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Summary report

1. Introduction

Elevated tropospheric ozone has known harmful effects on natural vegetation, agricultural crops and human health. The concentrations of this secondary pollutant in the air are dependent on atmospheric concentrations of its precursors, nitrogen oxides (NO_x) and volatile organic compounds (VOCs), as well as on relevant atmospheric conditions. Therefore, the damage caused by elevated ozone concentrations has to be also considered in decision making about emission reduction strategies for NO_x and VOCs.

The negative impacts of exposure to elevated air pollution on the vegetation have by now been studied and quantified on large geographical scales, e.g., in North America, Europe and Asia (RAINS, RAINS ASIA, NAPAP). So far, no attempt has been made to quantify the extent of the pollution exposure of human population and resulting negative health impacts in relation to emissions on a large geographical scale.

In order to help assess the negative impacts of elevated air pollution levels also on humans, a study was devised to quantify the European population exposure to major air pollutants in relation to pollutant emissions. The aim is to estimate the health impacts resulting from different emission reduction strategies. So that this work would be of particular use to policy makers, the information has been compiled in a database to be used in an integrated computer model called EXPO: *EX*posure of the European *PO*pulation to Air Pollution.

EXPO uses air quality data officially reported in international databases to describe human exposure to air pollutants. It is not the aim of EXPO to duplicate international data collection, but rather to use the information already collected, complemented with other data when necessary.

Regional assessments of ambient pollution levels and their changes are done using the EMEP models for acidifying compounds and the EMEP oxidant model. These assessments take into account total amount of emissions, but do not specifically describe the urban pollution. Such approach is well suited when considering vegetation that is mainly found outside the cities, but further investigations are needed in order to describe the human exposure in cities.

The aim of the present phase of the project is to investigate if the urban ozone concentrations throughout Europe can be described with simple empirical models based on currently available data. The goal is to provide a European-wide description of urban ozone concentrations based on precursor emissions, that may

be used for assessment of damage to human health under different precursor emission scenarios. One of the requirements is that the urban assessment of health damage is consistent with the assessment done for natural vegetation and crops.

The assessment of short-term ozone concentrations in cities has many complicating factors. In the cities, emission intensities of ozone precursors are high, and follow a diurnal and seasonal pattern. Ozone concentrations resulting from similar emissions can be variable due to the variation in factors such as the concentration levels of NO_x and VOCs, the UV radiation, temperature gradient, wind conditions and deposition. The emission conditions are such that there almost always will be enough pollutants to produce ozone, provided there are favourable meteorological conditions.

A database based on the EXPO data was assembled for this project, comprising of monitoring, modelling and emissions data. Several kinds of empirical models were investigated that relate the ozone concentrations with local and regional precursor emissions and with regional ozone concentrations. If established, such relationships would provide a simple method to estimate the urban ozone concentrations under different emission conditions. This note describes the main results of the project, and suggests a simple model for urban ozone concentrations based on regional modelling.

2. Possible starting points for an empirical urban ozone exposure model

More than half of inhabitants of Europe reside in urban areas, so that urban concentrations of pollutants need to be described. The range of ozone concentrations, and their temporal and local variability are higher in cities than outside. The short-term high concentrations are known to have a direct relationship with damage to human health.

Summaries from urban monitoring networks provide usually annual peak and mean statistics. Such reporting may be too restricted to adequately describe the short-term variation in concentrations, but indicates presence or absence of high concentrations that may have health impact. In Europe, ozone concentrations have been measured and reported internationally for a number of cities, most representatively for the EU.

Deterministic models of urban ozone concentrations are currently available only for a small number of cities, while monitoring data are more widely available. If empirical relationships were established between urban precursor emissions and urban ozone concentrations based on the measurement data, the European emission inventory for ozone precursors could be used to estimate urban ozone concentrations in the whole of Europe. The empirical relationships could be also used to estimate ozone concentrations under different emission situations. Such empirical approach has been tried successfully before for sulphur dioxide, nitrogen dioxide (IVM/IIASA/NILU, 1997) and for regional ozone (Heyes et al. 1996).

2.1 Atmospheric processes involving ozone

When an air mass is advected over a city, the nitrogen oxides (NO_x, the sum of NO_2 and NO; emissions are dominated by NO) will almost immediately deplete ozone by the reaction

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (1)$$

During sun-lit hours, O_3 will be quickly (time scale of minutes) regenerated by the photodissociation of NO_2:

$$NO_2 + h\nu\ (+O_2) \rightarrow NO + O_3 . \qquad (2)$$

Close to emission sources, and as long as the NO_x concentration is sufficiently high, NO, NO_2 and ozone will be in equilibrium by (1) and (2), the so-called photostationary state. This implies that Ox, defined as

$$Ox = NO_2 + O_3 ,$$

is constant.

Photolysis of ozone is to some extent controlling the formation of RO_2 radicals which are products from the oxidation of VOC. This proceeds via the formation of OH radicals in the simplified route:

$$O_3 + h\nu\ (+H_2O) \rightarrow 2OH + O_2 \quad (3)$$
$$VOC + OH \rightarrow RO_2 + VOC' \qquad (4)$$

Net formation of ozone and Ox occurs only through the subsequent reaction

$$NO + RO_2 \rightarrow NO_2 + RO. \qquad (5)$$

Thus, only at some distance away from the emission sources, when the combined effect of dilution and chemistry has to some extent replenished ozone (and depleted the NO_x), the air mass will experience an influence from the photochemical Ox and ozone generation. Furthermore, the relative rate of ozone production (dO_3 / dNO_x) is non-linear, and would normally increase with decreasing NO_x concentration. Box model calculations have indicated that maximum OH and, hence, maximum rate of VOC oxidation, occurs at a NO_x concentration about one ppb.

In a city one would therefore expect a negative, linear relationship between ozone and NO_2 at high NO_x concentrations (ozone depletion), whereas an additional influence from photochemical production would be expected at some distance downwind of the most important NO_x sources.

3. The project database

The following data were compiled from the EEA TC AQ databases, the EMEP database, the EMEP modelling results, and from information provided by individual countries:

- Annual measured ozone statistics per site and year (annual average and 98th percentile of hourly values). The database contains over 1100 site-years, with most data reported from Germany, covering one or more years from the period 1988-1994.
- Annual measured ozone statistics at the EMEP site nearest to each urban area (annual average and 98th percentile of hourly values).
- Results of the EMEP photooxidant model calculations, given as annual statistics.
- Annual measured NO_2 statistics per site and year (annual average, 98th percentile of hourly values). This database contains ca. 800 site-years within the period 1988-1994, mostly in the EU.
- Annual calculated average of regional NO_2 concentrations (EMEP model average for 10 years).
- Emissions of NO_x and VOCs in sub-national regions (NUTSIII areas) and in EMEP 150 x 150 km^2 grid, covering Europe.
- Number of inhabitants separately for all cities above 50.000 inhabitants in Europe, and with sub-national resolution (NUTSIII areas).

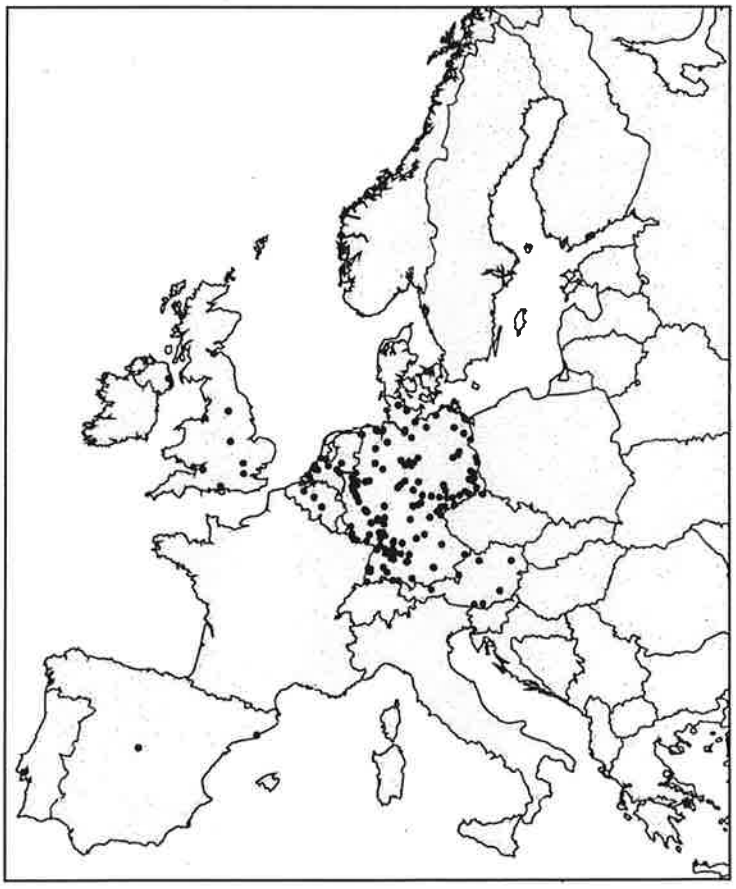

Figure 1 Map of cities ozone measurement data used in the analysis.

3.1 Representativity of the urban monitoring data

The database is limited (see Figure 1), mainly to Western Europe. In Germany, all cities over 20.000 inhabitants are covered, while outside of Germany, only selected large cities are represented. Figure 3 shows, for cities with more than 50.000 inhabitants, the distribution in size classes of cities covered in the measurement database, compared to the distribution of cities for the area covered by the EMEP oxidant model. In the area covered by the EMEP model, there are 1478 cities larger than 50.000 inhabitants, altogether 303,6 million inhabitants. Our database overrepresents large cities, and is not geographically representative. (136 cities with more than 50.000 inhabitants, with 52,7 mill. inhabitants).

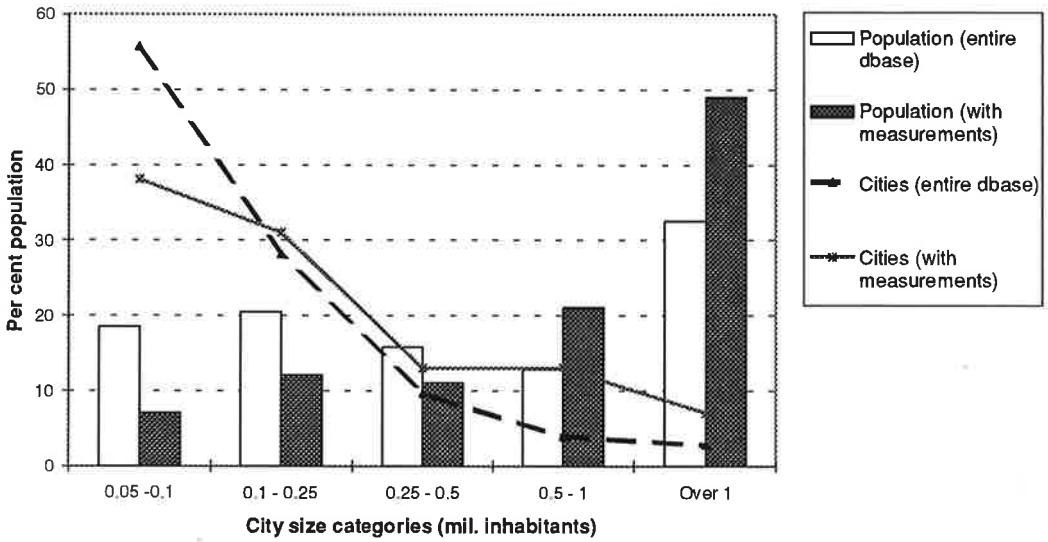

Figure 2 Percent of inhabitants in city size categories (columns) and percent of cities in individual city classes (lines), population of cities with valid measurement data (52,7 mil.) compared to all cities (303,6 mil.).

4. Establishing empirical relationships

Two topics were addressed:

- a relation between the reported measured annual average and 98th percentile of hourly values and AOT60 value (Accumulated dose Over Threshold 60 ppb ozone in a calendar year, given in ppb-hours), and
- a relationship describing ozone as a function of precursor emissions, accounting for regional O_3 and NO_2 concentrations.

4.1 Relation between different annual statistics

For vegetation, cumulative indices of dose acquired over the growing season, are indicative of adverse effects. These are given as AOT*xx* values (accumulated dose over threshold *xx* ppb ozone in a given period, e.g. growing season, given in ppb-hours). The AOT60 value indicates a threshold for damage at 60 ppb ozone

(about 120 µg/m³ ozone), corresponding approximately to the short-term air quality guideline. Although air quality standards to protect human health are defined as short or long-term concentrations, a cumulative index such as AOT60 may be used as indicative of compliance (UNECE/WHO 1996).

The AOT60 values are not reported by the monitoring networks. Based on sites with available hourly data, the AOT60 statistics (accumulated over a calendar year) was related to the 98th percentile, a value that is often reported. For sites where the AOT60 is non-zero, there is a close relationship between the 98th percentile and the AOT60 value (Figure 4), represented by an equation

$$\begin{aligned} &\text{AOT60} = (\text{P98}*1.45-112)^{**}2 && \text{for P98} > 80\ \mu g/m^3\ , \\ &\text{AOT60} = 0 && \text{for P98} \leq 80\ \mu g/m^3\ , \end{aligned} \qquad (6)$$

where the P98 is the annual 98th percentile of hourly concentrations measured in µg/m³ at a given site. The equation (6) gives a good approximation of the AOT value, but some sites report an AOT60 much higher than their 98th percentile value would suggest. Such sites are likely to be infrequently hit by an ozone plume, a phenomenon that is difficult to predict.

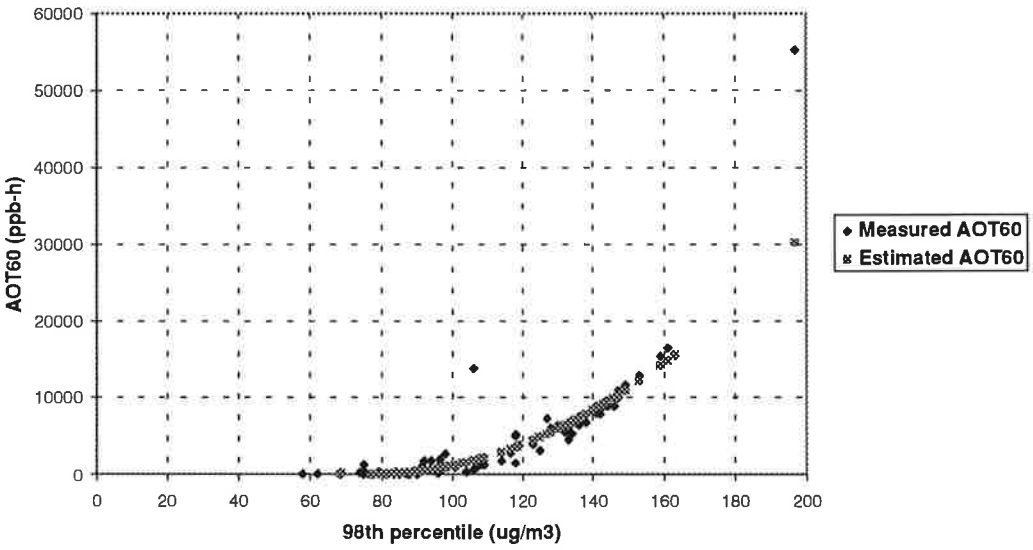

Figure 3 Measured urban AOT60 values and AOT60 values calculated using equation (6), related to the 98th percentile of measured hourly values.

4.2 Comparison between urban and background ozone measurements

Measurement data for each city represented in the database were complemented with measurements taken at "nearest" EMEP ozone monitoring site. Usually, the nearest site would be the nearest-lying background EMEP site within 200 km of a city, not separated from the city by mountains.

A comparison of the annual statistics for the urban and the "nearest" site shows that the annual average concentration in a city is usually lower than on the EMEP site, however, the 98th percentiles are of the same size (Figure 4 and Figure 5). This indicates that in the cities, ozone is most of the time depleted due to nitrogen oxides emissions, however, in periods with ozone formation, the concentrations can attain values higher than in the rural areas. This is also illustrated in Figure 6, that shows simultaneous hourly measurements at the site Central London (urban background site) and Bottesford (suburban site located in open farmland). Most of the time, the concentrations within city are lower than those outside, but in several cases, high short-term peaks occur in the city and not outside. The site Bottesford is about 70 km away from the centre of London.

Figure 7 illustrates the relationship between measured AOT60 in the cities and measured AOT60 at the nearest EMEP site. Our database of hourly data shows that the regional AOT60 values in most cases provide an upper estimate of the urban AOT60 values, and this serves as a basis for an exposure model.

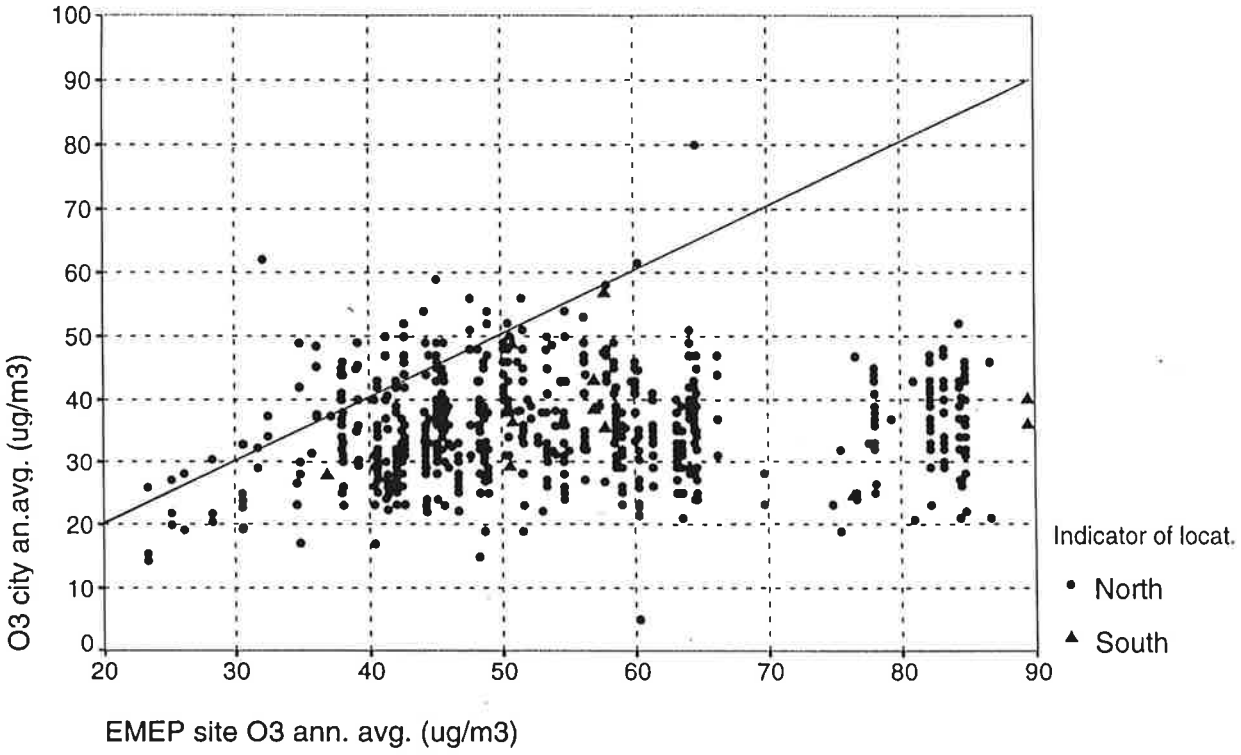

Figure 4 Relationship between urban ozone and nearest EMEP site ozone (annual averages) with indicated 1:1 line.

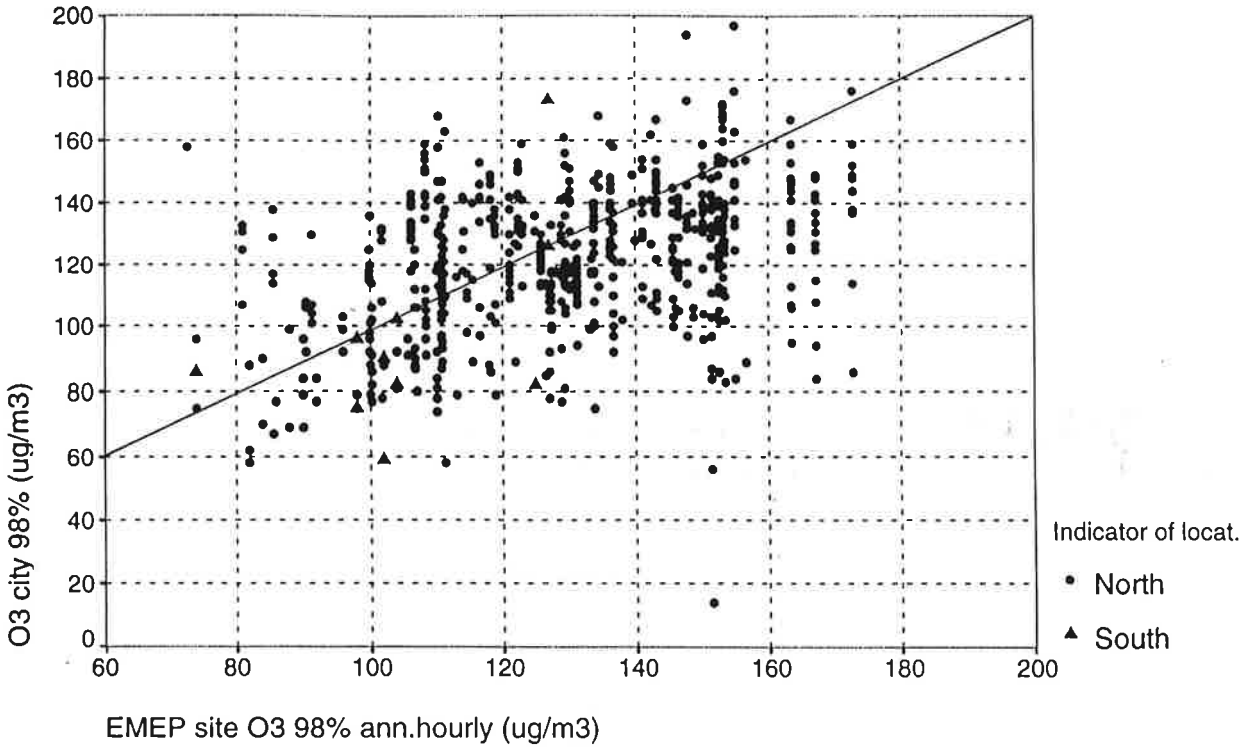

Figure 5 Relationship between urban ozone and nearest EMEP site ozone (98th percentiles of hourly values over one year) with indicated 1:1 line.

4.3 Relationship between ozone precursor emissions and measurement statistics at urban sites

Emissions of NO_x and VOCs provided in the CORINAIR inventory were proportionally assigned to cities based on population. No natural emissions were assumed to be taking place in the cities. The NO_x and VOC emissions are highly correlated, and the resulting city emissions of the two compounds were almost perfectly correlated. Several combinations of NO_x and VOC urban and regional emissions were investigated.

Other data used in the statistical models included:

- annual measured concentrations of nitrogen dioxide for each site,
- geographical parameters (latitude, longitude), and site location relative to Alps,
- regional ozone concentrations (AOT60) calculated by the EMEP oxidant model,
- measured ozone concentrations at the nearest EMEP site
- annual average regional NO_2 concentrations calculated by the EMEP model for acidifying compounds.

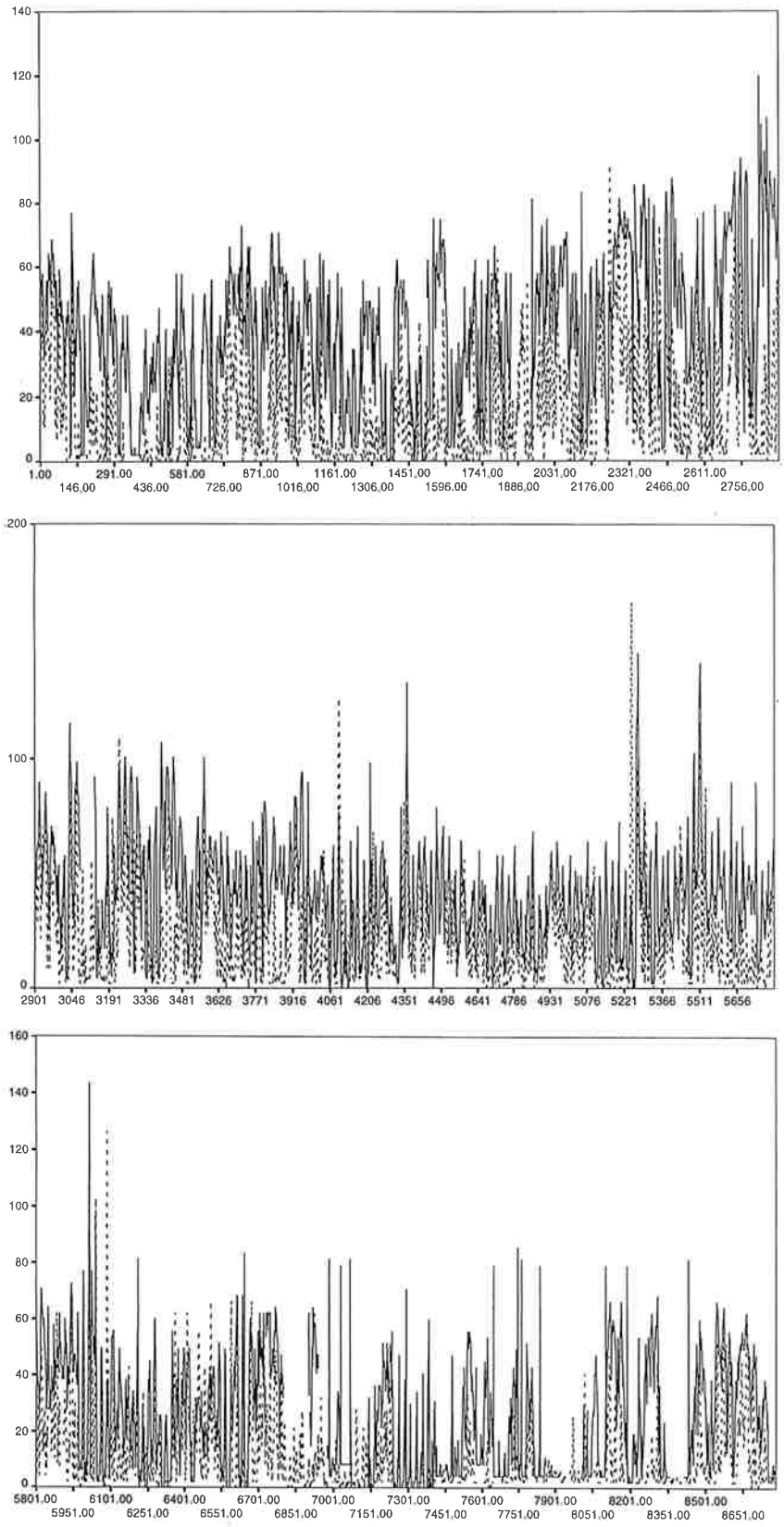

Figure 6 Time series of simultaneous ozone measurements in urban background site Central London (dashed line) and suburban site Bottesford (full line) for the year 1988. Time in hours, concentrations in µg/m³.

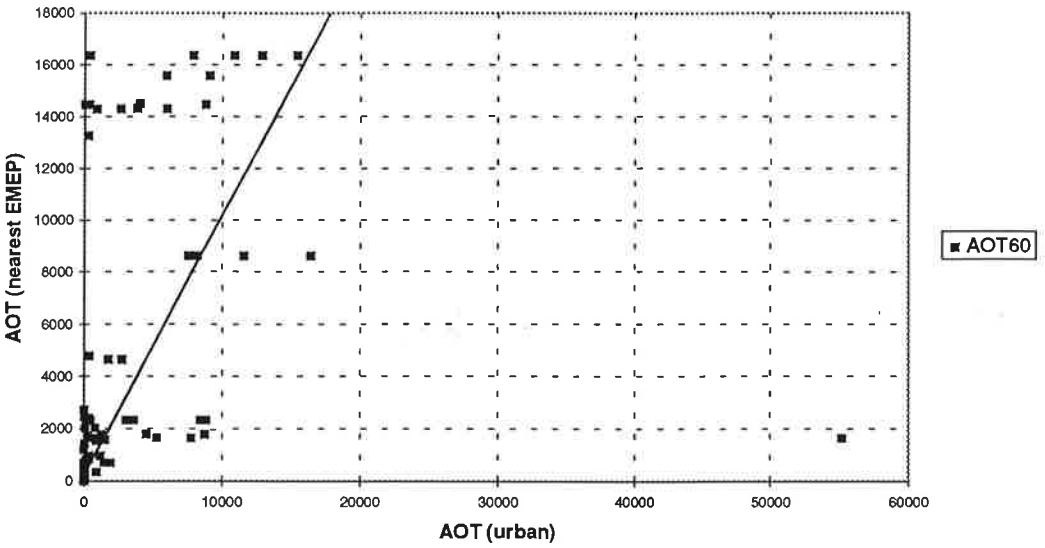

Figure 7 AOT60 values measured at urban sites and at their "nearest" EMEP site.

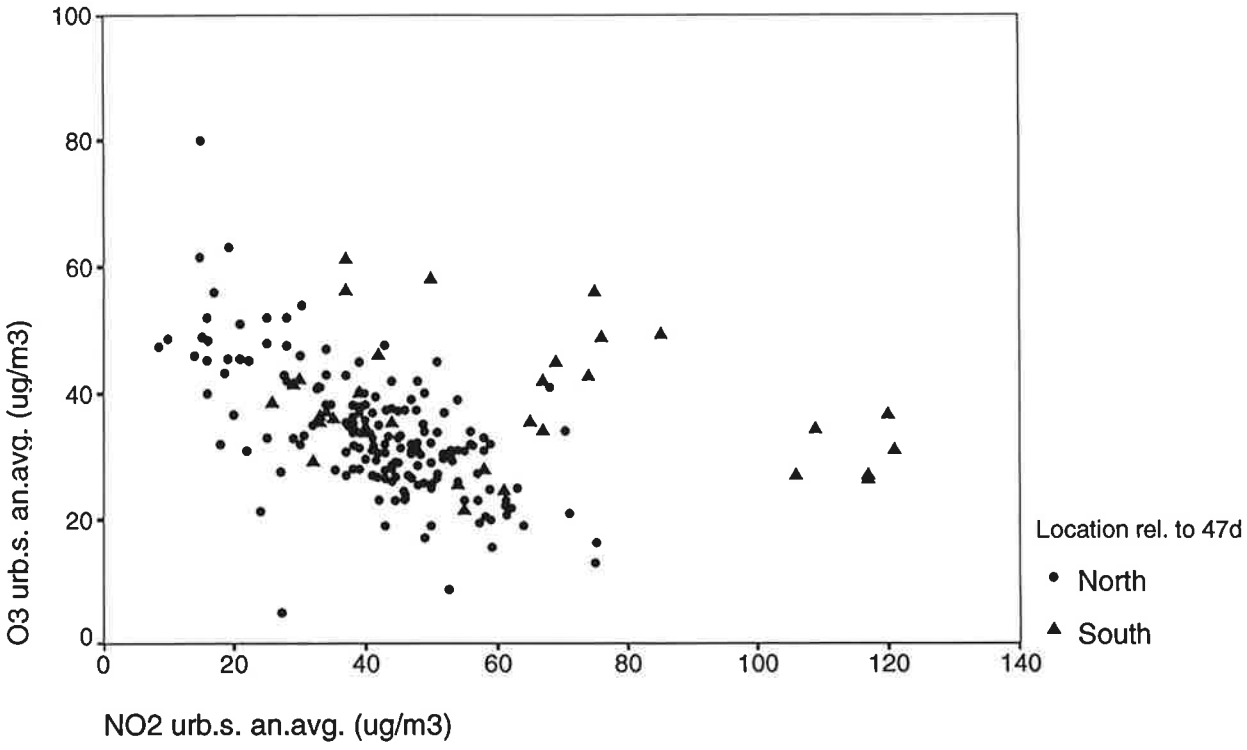

Figure 8 Scatter diagram of ozone urban annual average concentration against NO_2 annual average measured same year at the same site.

The expression relating annual averages of ozone and NO_2 measured at the same site confirmed the negative relationship between NO_2 and O_3 , a decrease of approximately 1 µg/m³ O_3 with 2 µg/m³ increase of NO_2 (see Figure 8). No direct relationship with ozone precursor emissions was found.

For the 98th percentile, a weak but statistically significant proportional relation was found between the urban and nearest EMEP site measured annual percentiles of hourly ozone values. No relation was found between ozone and NO_2 concentrations or NO_x or VOC emissions.

This indicates that the statistical data available do not allow conclusions about the influence of local urban emissions and photochemical activity on short-term ozone concentrations. This confirms that the complex processes influencing ozone are not adequately described by the monitoring data used.

5. A simple urban concentration and exposure model

Regional ozone concentrations seem to provide an upper estimate of the urban concentrations (Figure 4 and 5). As the regional ozone levels are adequately modelled with the EMEP photochemical oxidant model, the results of the EMEP model may be used as an approximation of the urban concentrations when assessing the impact of different emission scenarios.

The human urban ozone exposure is calculated from data on cities with over 50.000 inhabitants, complemented with the results of the EMEP oxidant model.

5.1 Exposure models

The ambient concentrations, expressed as AOT60, are an indicator of exposure to values over the health-related guideline value of about 60 ppb (120 µg/m³ O_3) as an 8-hour average. The following three exposure models were considered.

M1. The total city population is exposed to an average AOT60 calculated from a measured annual 98th percentile of hourly values using relationship (6).

M2. The total city population is exposed to regional AOT60 value (estimated by the EMEP regional oxidant model).

M3. If the regional AOT60 value (EMEP model) is below or equal to 5000 ppb-hours:

the total city population is exposed to the regional AOT60 value.

If the regional AOT60 (EMEP model) is above 5000 ppb-hours:

75% of the total city population is exposed to regional AOT60 reduced by 50% (0.5*AOT60), and 25% of the total city population exposed to regional AOT60 with no reduction.

The reduction of the regional values in the city used in model **M3** is a crude approximation to the relationship between urban and rural AOT60 values seen in

Figure 7. It is further based on an assumption that a significant part of the urban area, say 25%, is affected mainly by ozone concentrations in the air masses coming from outside of the city. These proportions are arbitrary, as more detailed empirical evidence is lacking.

5.2 Comparison of the exposure models based on the measurement database

Figure 9 shows the estimated number of inhabitants exposed to AOT60 values lumped into bins < 100 ppbh, 101-1000 ppbh, 1001-2000 ppbh, ..., > 7000 ppbh. The models M2 and M3, based on calculated regional AOT60 values, underrepresent low and very high exposures when compared to M1 based on measured annual 98th percentile.

These exposure models are very simple. Urban ozone concentrations have short-term peaks, so that the population distribution and mobility within the city are important factors in determining population exposure but such information is not readily available.

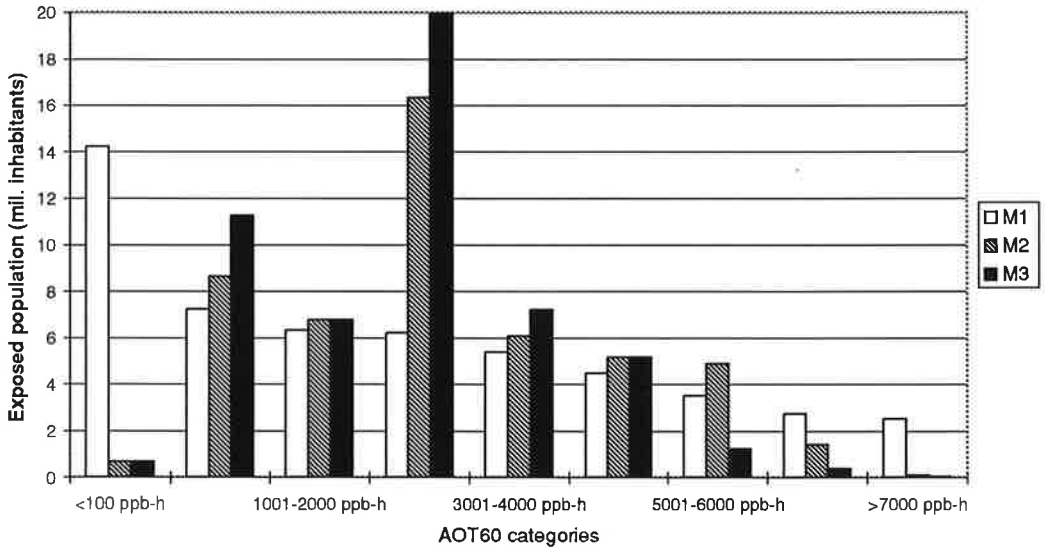

Figure 9 Comparison of exposure calculated by three exposure models M1, M2 and M3.

5.3 Urban exposure based on regional AOT60 values for the whole region covered by the EMEP oxidant model

A measurement-based model that with reasonable accuracy predicts changes in urban ozone concentrations in relation to changes in NO_x and VOC emissions could not be derived. We therefore model the European urban exposure by linking regional model results with the urban database. For this, we have used exposure models **M2** and **M3**.

Figure 10 shows urban population distributed into classes of AOT60 values. The lowest class includes very low non-zero AOT60 values, indistinguishable from

zero. More than 30 million urban inhabitants are not exposed to short-term (hourly) values over 120 µg/m³ , indicating that the 8-hr guideline (120 µg/m³) is not exceeded. This result is based on 3-year (1993-1995, April-August) average results of the EMEP oxidant model (D. Simpson, 1997).

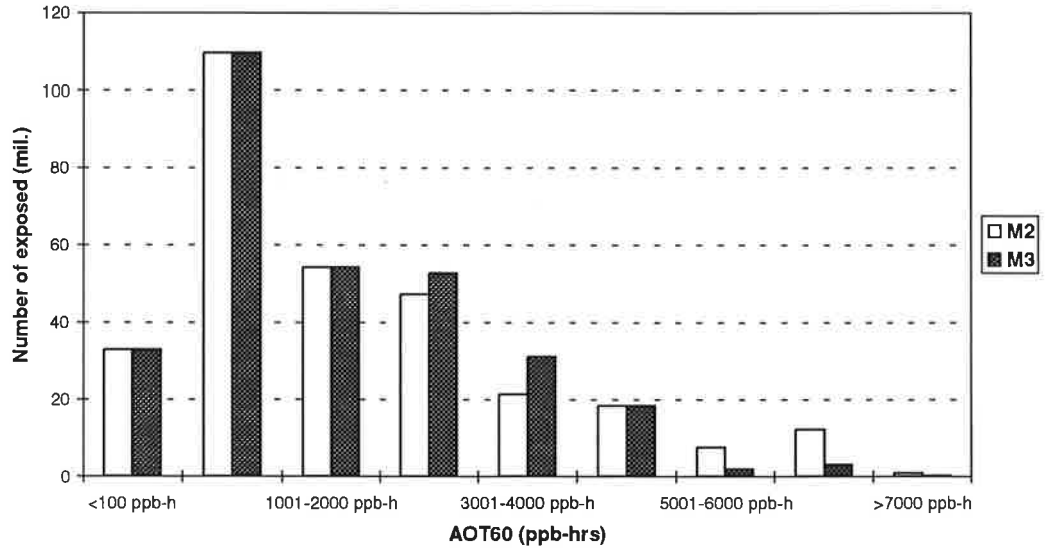

Figure 10 European urban population exposure to ozone estimated from the EMEP regional oxidant mode using exposure models M2 and M3.

6. Discussion.

Our aim has been to express urban ozone concentrations in terms of precursor emissions using a statistical model and available internationally reported measurement data. Such empirical approach has been tried successfully before for sulphur dioxide, for nitrogen dioxide and for regional ozone.

In the empirical model for regional ozone concentrations, the output of several scenario calculations with the EMEP oxidant model is used to construct regression models (Heyes et al., 1996). The regression models relate ozone concentrations to "effective emissions" of NO_x and VOC, i.e. emissions accumulated along trajectories and corrected for dilution. An approach using an urban photochemical oxidant model was not possible within this project as a model appropriate for individual cities was not available.

When observational data are input to a statistical model, rather than the results of a deterministic photochemical model, there is a substantial increase in the random scatter in the data, reducing any statistical significance. Secondly, the relationships between emissions (by city or region) of NO_x and VOC and ozone concentrations are very different in urban areas compared to rural areas due to differences between urban and non-urban areas in emissions, transport time from the sources, dry deposition, UV radiation, temperature and other factors. Also, the measured data in cities available here are not continuous (parallel) time series, but

rather annual averages and percentiles, and as such describe only average conditions.

As Ox (NO_2 + O_3) is a better conserved quantity than O_3 and NO_2 individually, we have also considered this quantity. However, it was not possible to derive a relationship between urban and rural Ox from the available data. Therefore, we have investigated a relationship between city ozone concentration on one hand and on the other hand city NO_2 concentration, city NO_x and VOC emissions, and an estimate of dilution for each monitoring site, but this did not bring out a positive result either.

The urban networks are not set up in all cities in Europe, and the data are not always reported to international bodies. Where the monitoring data are available, the networks are usually not designed to adequately describe ozone concentrations for the entire city. The empirical measurement data will therefore not provide detailed information relevant to ozone formation or depletion, as many factors, such as the position of the monitoring sites up- or downwind of NO_x and VOC sources, are generally not known.

A possible approach would be to collect several years of measured time series data for ozone and NO_2 from sites representing adequately individual selected cities, together with enough site classification. Complemented with meteorological information, this may provide a sufficient basis for city-specific statistical ozone model. The approach would need extensive data collection, and the result would have to be extrapolated to whole Europe. This was not possible within this project.

A feature of EXPO is to use as much of officially available information as possible. For practical reasons, such data have to be quite condensed, and are most often reported as annual summaries, not time series. For the empirical urbanozone relationships, the EXPO database was extended with additional short- and long-term ozone measurement data, and with information updates.

Further uncertainty of the results is brought about by the oversimplified model for exposure used. Several such broad models were investigated earlier, (WHO ECEH 1995, Sluyter et al. 1995). These models allocated concentrations to population proportionally to the available monitoring data assuming simple concentration distributions, while we assign one concentration value to the whole city. Ours may be a conservative approach that underestimates the high short-term concentrations.

7. Conclusions

We have assembled database of urban and regional ozone measurements, urban and regional NO_x and VOC emissions, and of other information. The following empirical relationships were studied:

- Relationships between annual averages and annual 98th percentiles of hourly values of urban ozone on one side and urban and regional NO_2 concentrations, NO_x and VOC emissions, and regional ozone concentrations on the other side;
- relationships between short-term urban and regional ozone concentrations;
- relationship between measured annual AOT60 and measured 98th percentile of hourly concentrations in urban areas.

Based on our data, we did not find any simple statistical predictions of peak ozone concentrations in terms of precursor emissions. Urban ozone could be related to regional ozone. We therefore suggest to employ the regional photochemical model also for cities, as the regional concentrations quite consistently predict the upper envelope of urban ozone levels. The important exception is, however, the formation of urban plumes. We did not find an adequate statistical model that would describe the urban plume formation based on our types of data. Therefore, our results do not provide any estimate of ozone levels in urban plumes.

With the current emissions, over 90% of the European urban population are exposed to peak values of ozone over the 8-hour air quality guideline (120 $\mu g/m^3$ or 60 ppb as 8-hour average). This result is derived using an exposure assessment based on the regional ozone estimates from the EMEP regional photochemical model. With a reduction of NO_x emissions in one individual city, the average exposure to ozone is likely to rise there.

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AN EMPIRICAL ASSESSMENT OF EUROPEAN URBAN OZONE: CONCENTRATIONS AND EXPOSURE. Summary report	Alena Bartonova NILU PROJECT NO. O-96077
AUTHOR(S) Alena Bartonova	CLASSIFICATION * A CONTRACT REF.

REPORT PREPARED FOR: United Nations European Comission for Europe, Task Force for Integrated Assesment Modelling and Nordic Council of Ministers

ABSTRACT The aim of the project is to establish a European-wide model for urban exposure to ozone in relation to NO_x and VOC emissions. As a basis, internationally reported data are to be used as much as possible. Database was created for the project, assembling urban O_3 and NO_2 measurement data, NO_x and VOC emission data, urban and regional population data, and other information such as geographic information and results of regional modeling of ozone and nitrogen species. No relationships were established between urban ozone concentrations and urban NO_x and VOC emissions, however, based on relations between urban and regional ozone measurements, exposure model was suggested using the results of EMEP regional oxidant modeling. It is estimated that currently, 90% of urban population are exposed to short-term concentrations above the 8-hour air quality guideline of 120 µg/m^3 ozone.

NORWEGIAN TITLE

Empirisk vurdering av ozon i byer i Europa: konsentrasjoner og eksponering.

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* *Classification*

- *A Unclassified (can be ordered from NILU)*
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