Air4EU/AIR QUALITY ASSESSMENT SESSIONS

EVALUATION OF THE AIR QUALITY ASSESSMENT REPORTS BY MEMBER STATES UNDER THE EUROPEAN AIR QUALITY DIRECTIVES

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TREND ANALYSIS OF NO_2 URBAN BACKGROUND CONCENTRATIONS: IMPORTANCE OF DIRECT NO_2 EMISSIONS VERSUS OZONE/NO_x EQUILIBRIUM - AN AIR4EU CASE STUDY IN ROTTERDAM

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BASIC DATA ASSIMILATION METHODS FOR USE IN URBAN AIR QUALITY ASSESSMENT

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ESTIMATION OF MODELLING UNCERTAINTY FOR AIR QUALITY ASSESSMENT: THE AIR4EU BERLIN CASE

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UNCERTAINTY AND REGIONAL AIR QUALITY MODEL DIVERSITY: WHAT DO WE LEARN FROM MODEL ENSEMBLES?

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SOURCE APPORTIONMENT OF PARTICULATE MATTER ($PM_{2.5}$) USING DISPERSION AND RECEPTOR MODELLING - A CASE STUDY FOR OSLO

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ASSESSMENT OF NON-EXHAUST PM EMISSIONS BY ROAD TRAFFIC IN URBAN AREAS – AN AIR4EU CASE STUDY IN ROME, LONDON, OSLO AND ROTTERDAM

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STUDY OF THE PAST, PRESENT AND FUTURE STATE OF AIR QUALITY IN THE RUHR AREA

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A WEB BASED MAPPING TOOL FOR THE HARMONISED INTERCOMPARISON OF AIR QUALITY MAPS

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SPATIAL REPRESENTATIVENESS OF AIR QUALITY MONITORING DATA AND THE RELEVANCE FOR MODEL VALIDATION

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THE MODELLING SYSTEM SUPPORTING PIEMONTE REGION YEARLY AIR QUALITY ASSESSMENT.

Bande S.[1], Clemente M.[1], De Maria R..[1], Muraro M.[1] Picollo M.E.[1], Arduino G.[2], Calori G. [3], Finardi S.[3], Radice P.[3], Silibello C.[3], Brusasca G.[3] - [1] ARPA Piemonte, corso Unione Sovietica 216, Torino, Italy, [2] Regione Piemonte, via Principe Amedeo 17, Torino, Italy, [1] ARIANET, via Gilino 9, 20128 Milano, Italy

ANALYSIS OF INDUSTRIAL-URBAN NO_X SAMPLING DATA

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SIMPLE CURVES FOR ESTIMATING NITROGEN DIOXIDE IN INDUSTRIAL PLUMES.

D R Middleton[1], R S Sokhi[2], B E A Fisher[3] - [1]Met Office, Exeter EX1 3PB U.K., [2]University of Hertfordshire, Hatfield, [3]Environment Agency, Reading

ON THE IMPACT OF PCDD/Fs EMITTED FROM A 2006 LANDFILL FIRE NEAR THESSALONIKI, GREECE

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DETECTING CHANGES IN NO_2 AND NO_X AIR QUALITY IN VALENCIA (SPAIN)

C.Capilla, P.Aragon, J.Atienza - Universidad Politécnica de Valencia (Spain)

POM CHARACTERIZATION AT A PERI-URBAN SITE IN CENTRAL ITALY: THE RB-MIUR-2001 PROJECT "CHEMICAL INDEXES OF AMBIENT TOXICITY"

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SITUATION AND TENDENCIES IN AIR QUALITY IN A NORTH EUROPEAN MEDIUM-SIZED TOWN

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ASSESSMENT OF THE SPATIAL VARIABILITY OF AMBIENT NO_2 CONCENTRATIONS WITHIN THE URBAN AREA OF ATHENS

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AMBIENT AIR QUALITY OVER THE INDUSTRIAL CITY OF YANBU (SAUDI ARABIA)

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METHODOLOGIES AND RECOMMENDATIONS FOR EMISSION CALCULATION IN THE FRAME OF AIR4EU

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COMPARISONS OF DATA ASSIMILATION METHODS IN THE FRAMEWORK OF THE AIR4EU PROJECT

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FACILITATING THE COMPARISON OF URBAN AIR QUALITY – A NEW SET OF COMMON AIR QUALITY INDICES

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APPLICATION OF DATA ASSIMILATION IN OPEN ROAD LINE SOURCE MODELLING

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EVALUATION OF THE AIR QUALITY ASSESSMENT REPORTS BY MEMBER STATES UNDER THE EUROPEAN AIR QUALITY DIRECTIVES

Dick van den Hout [1] and Andrej Kobe [2]

ABSTRACT

EU Member States are assessing their air quality under the Air Quality Framework Directive and daughter directives and report this annually to the European Commission. These reports include data on the air quality zones, information on exceedances of air quality thresholds and on the assessment methods used.

In 2004, about 3700 monitoring stations were used for the assessment under the air quality directives. Several Member States sent also spatial statistics and/or maps of air quality calculated by modelling.

The limit value for daily mean PM_{10} concentrations was exceeded most extensively; it was followed by the limit value for the annual mean NO_2 concentration. The pattern of exceedance zones varied over the years, not only because of changing air quality, but probably also due to changes in the assessment methodology. Most exceedances for NO_2 and about half of the exceedances for PM_{10} related to traffic.

INTRODUCTION

In 2001 the First Daughter Directive[3] on sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead under the Air Quality Framework Directive[4] entered into force within the Member States. In 2003 it was followed by the Second Daughter Directive[5] on benzene and carbon monoxide and in 2004 by the Third Daughter Directive[6] on ozone. Under these directives Member States have, among other things, to assess the air quality throughout their territories and report the results to the European Commission. For the results several information streams exist, of which the following two, each specified in a legal Decision, are the most extensive:

- Reporting processed results that are most relevant to the directives, such as exceedances of limit values and target values and reasons for exceedance[7];
- Reporting the full time series of concentrations of the monitoring stations, including the metadata which describe details of the stations, under the "EoI" Decision[8].

This paper deals with the first information stream. Overviews of the information reported under the EoI Decision are annually published by the European Topic Centre on Air and Climate Change.

As a consequence of the stepwise introduction of the various daughter directives and furthermore of the accession to the EU of ten new Member States in 2003, the number of reports has increased over the years, both in terms of the number of Member States and the number of pollutants. This paper focuses on the year 2004, which was the first year in which 25 Member States had to report under the first three daughter directives (Luxembourg is not included because it did not send a report); it will also show changes over the period 2001-2004.

A more detailed overview is given in (Hout, 2006) and at the European Commission's air pollution website http://europa.eu.int/comm/environment/air/index.htm.

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[3] Council Directive 1999/30/EC relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air (amended by Commission Decision 2001/744/EC).

[4] Council Directive 96/62/EC on ambient air quality assessment and management.

[5] Directive 2000/69/EC relating to limit values of benzene and carbon monoxide in ambient air.

[6] Directive 2002/3/EC relating to ozone in ambient air.

[7] Commission Decision 2004/461/EC laying down a AQ questionnaire to be used for annual reporting on ambient air quality assessment under Council Directives 96/62/EC and 1999/30/EC and under Directives 2000/69/EC and 2002/3/EC of the European Parliament and of the Council.

[8] Council Decision 97/101/EC establishing a reciprocal exchange of information and data from network and individual stations measuring ambient air pollution within the Member States (amended by Commission Decision 2001/752/EC).

DESIGNATION OF AIR QUALITY ZONES IN MEMBER STATES

Member States are obliged to divide their territory into zones and agglomerations, which are the main territorial units for reporting the air quality assessment and management to the European Commission. Although Member States were entirely free in choosing the method of designing zones and indeed initially considered widely different methods, they have taken rather similar approaches, linking zones largely to administrative territories. In 2004, the total number of zones in the EU ranged between 759 and 971 depending on the pollutant and protection target. Numbers of zones per Member State and zone sizes vary widely, often reflecting country size, pollution levels, population densities and geographical conditions.

EXCEEDANCES OF AIR QUALITY THRESHOLDS

The directives set out several types of air quality thresholds for the various pollutants:

- Limit value (LV); this is a threshold that must be met from a certain year on.
- Limit value plus margin of tolerance (LV+MOT); the MOT is a decreasing increment set for most limit values, applying before the year in which the limit value must be met. If exceeded, a plan or programme has to be prepared or implemented and sent to the Commission ensuring that the limit value will be met in time.
- Target value (TV), a threshold for ozone to be met where possible from a certain year on.
- Long-term objective (LTO), a threshold for ozone to be attained in the long-term, save where not achievable through proportionate measures.

For ozone, the target value for health was exceeded in 30% of the zones reported and the target value for vegetation in 22%. The long-term objectives for health and vegetation were reported to be met in only 3% and 13% of the zones respectively. Figure 2 shows the exceedance percentages for the target values per Member State.

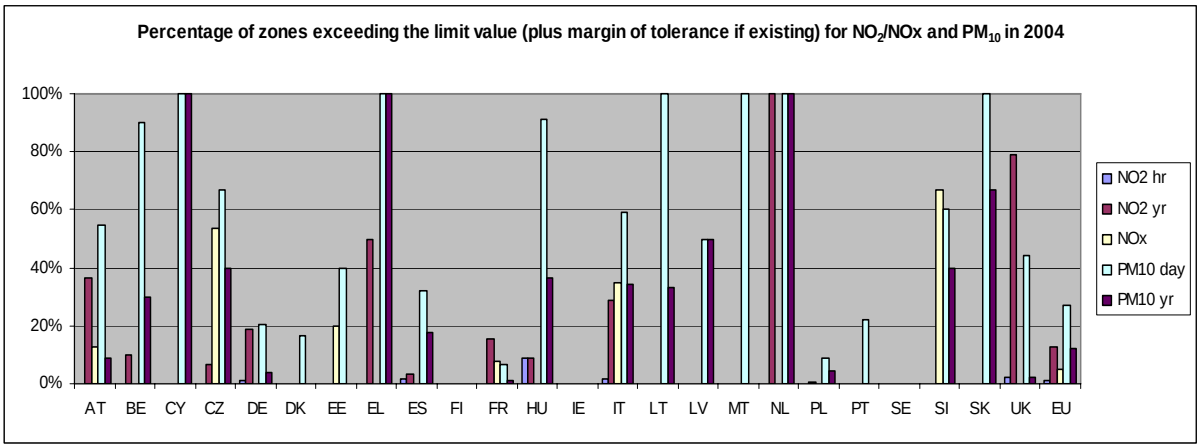

Figure 1 Percentage of zones exceeding the limit value (plus margin of tolerance if existing) for NO_2, NO_x and PM_{10} in 2004.

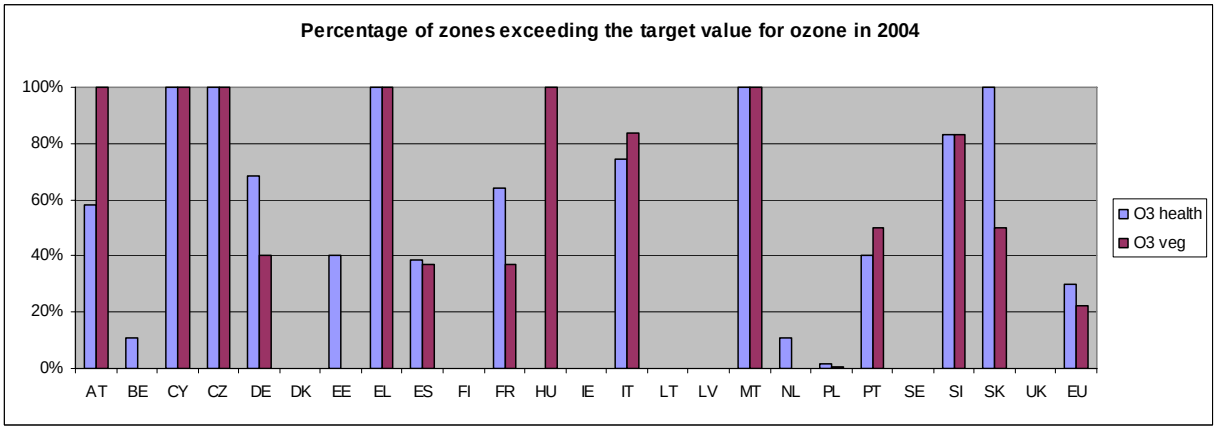

Figure 2 Percentage of zones exceeding the target values for ozone in 2004.

For NO_2, most exceedances related to traffic (see Figure 3). For PM_{10} about half the exceedances related directly to traffic and the others were due to a variety of causes. This was also reflected in the station types where exceedances occurred.

Of the limit values for ecosystems or vegetation, the NO_x limit value was reported to be exceeded more widely (5% of the EU15 zones designated for this limit value) than the limit value for SO_2 (1%).

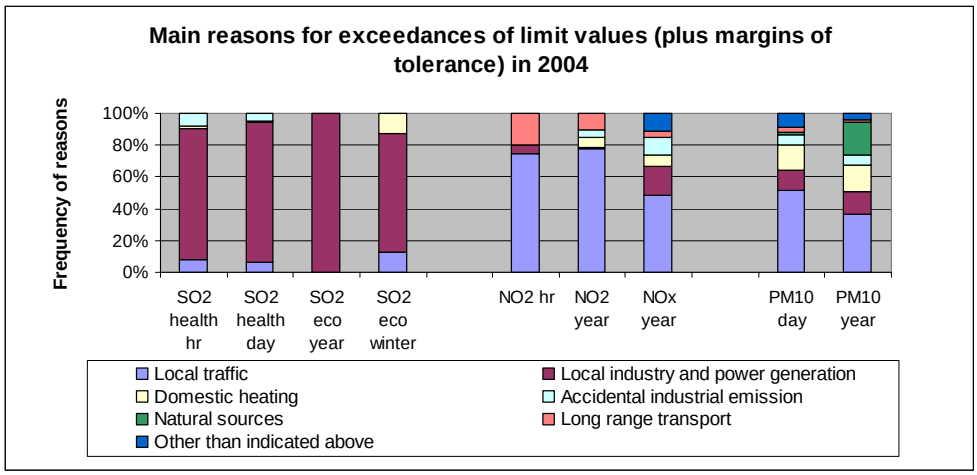

Figure 3 Frequency with which a reason for exceedance of a limit value (plus margin of tolerance) was indicated (2004). The detailed set of reasons reported by the Member States have been grouped here into seven reasons.

Figure 4 presents the change in exceedances of the limit value for the 13 Member States that reported for all four years (please note the logarithmic scale, which visually levels the trends off strongly). The number of exceedances in the 13 Member States is in 2004 slightly lower than in 2003 for the two limit values that are the most difficult to attain. For the other limit values the number of zones in exceedance was in 2004 either lower or about equal to the number in 2003.

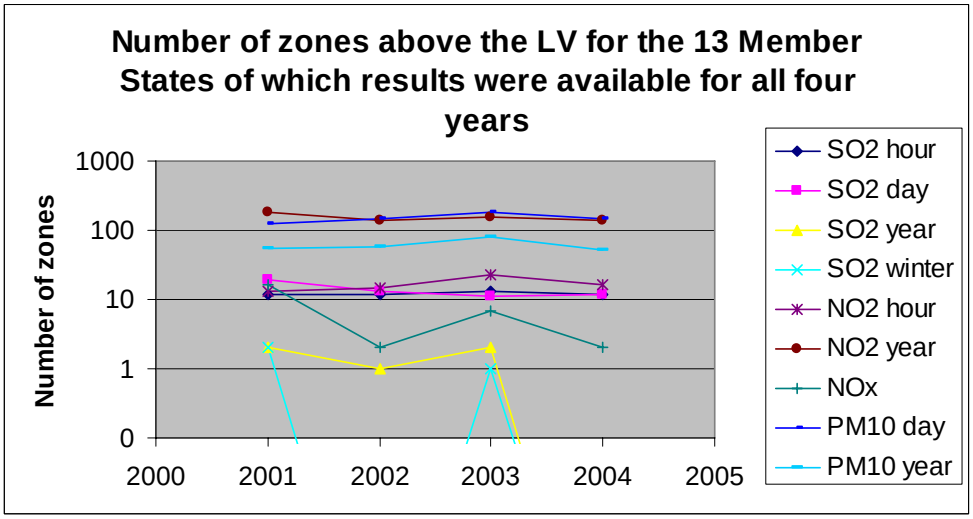

Figure 4 Change from 2001-2004 in the number of zones exceeding the limit values for the 13 Member States for which results were available for all four years.

Most Member States reported only exceedance for zones as a whole, without voluntary information on the surface area in exceedance; it should be noted that the percentages of zones with exceedance are not a good measure for the surface area or population exposed to levels exceeding thresholds.

In view of the differences between Member States in the methodology for air quality assessment (see below), significant differences in the extent to which exceedances are identified may be expected.

AIR QUALITY ASSESSMENT

In 2003, 4702 monitoring stations were reported to be used for measuring air quality under the air quality directives. 3418 stations were used for NO_2, 2851 stations for SO_2, 2296 for PM_{10}, 1765 for O_3 and less than 1500 stations for each of the other pollutants (see Figure 5). There were still considerable differences between Member States in the structure of their networks. It was also found that the information on the classification of stations needs to be improved. There were important differences between Member States in the correction of PM_{10} concentration measured with non-reference methods.

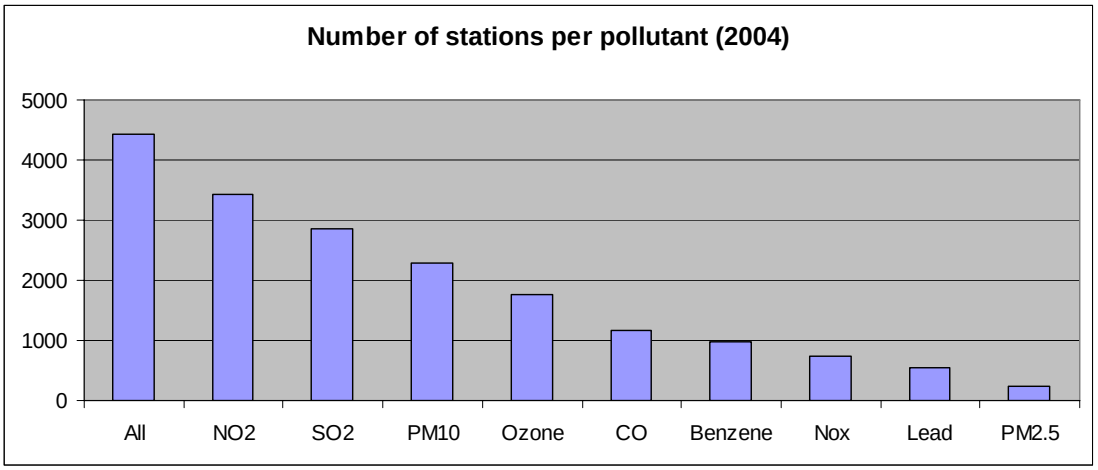

Figure 5 Total number of stations per pollutant in the EU in 2004.

The reporting of certain data needed for judging the fulfilment of assessment requirements is voluntary and several Member States did not send such data. Compliance with the minimum number of stations required by the directives could only be checked for the zones on which sufficient information was received. It was found that the number of stations for benzene and PM_{10} – pollutants not regulated before – was in a substantial number of zones (about one-seventh of the zones where measurement was mandatory) below the minimum.

There were large differences in the use of models between Member States for compliance checking. The percentage of zones for which the compliance status was based on modelling varied between 2% and 21%, depending on the air quality threshold. In most cases modelling was used for demonstrating that limit values were not exceeded.

ACKNOWLEDGEMENT

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TREND ANALYSIS OF NO_2 URBAN BACKGROUND CONCENTRATIONS: IMPORTANCE OF DIRECT NO_2 EMISSIONS VERSUS OZONE/NO_x EQUILIBRIUM - AN AIR4EU CASE STUDY IN ROTTERDAM

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ABSTRACT

The annual air quality standard of NO_2 is often exceeded in urban areas near heavy traffic locations. Despite significant decrease of NO_x emissions in 1986-2005 in the harbour and industrial Rijnmond area near Rotterdam, NO_2 concentrations at the urban background remain at the same level since the end of the nineties. Trend analysis of monitoring data revealed that the ozone/NO_x equilibrium is a more important factor than increasing direct NO_2 emissions by traffic. The latter has recently been identified as an additional NO_2 source due to the introduction of oxy-catalytic converters in diesel vehicles. It is concluded that local NO_x emissions in Rotterdam need to be reduced by 50% before lower NO_2 *urban background* levels can be achieved. This is a relatively costly abatement strategy and therefore a hotspot approach aimed at reducing NO_x emissions by local traffic measures (e.g. traffic-low zones) is more effective to meet European Clean Air standards.

1. INTRODUCTION

In urban areas with a relatively high NO_2 background, extensive road traffic and limited dispersion of air pollution, the annual air quality standard of NO_2 is often exceeded. The effectiveness of local measures to control air quality for NO_2 is questioned in view of the importance of the contribution of regional and urban background to NO_2 levels, even at "hotspot" locations. This contribution of NO_x emissions to NO_2 levels is largely controlled by ozone. Ozone concentrations mainly depend on large scale emissions of precursors and meteorological conditions. Thus, the effect of local measures to control NO_2 concentrations seems limited. Recent, it has been reported [Carslaw, 2005] that direct NO_2 emissions by road traffic has increased significantly the last decade. The impact of this development on urban NO2 concentrations is still uncertain. In order to implement effective local measures, local authorities require information both on the contribution of local sources as well as large scale effects on NO_2 concentrations. In the framework of the Air4EU project (www.air4eu.nl), a case study in Rotterdam was implemented to demonstrate trend analysis as a tool to extract the required information from ambient air monitoring data [Air4EU, 2006].

2. METHODOLOGY

In cooperation with the City of Rotterdam - DCMR, the case study in Air4EU demonstrates trend analysis [Roemer, 2001] as an instrument to study urban background NO_2 concentrations. Monitoring data of NO_x, NO_2 and ozone were used from an urban background station in Schiedam (Rotterdam) and a regional station in Zierikzee at the coast in the south of the Netherlands. The monitoring data covered the period 1986-2005.

3. RESULTS AND DISCUSSION

In Figure 1, the trends for the annual average NO, NO_2 and O_3 concentrations at the urban background station in Schiedam are presented.

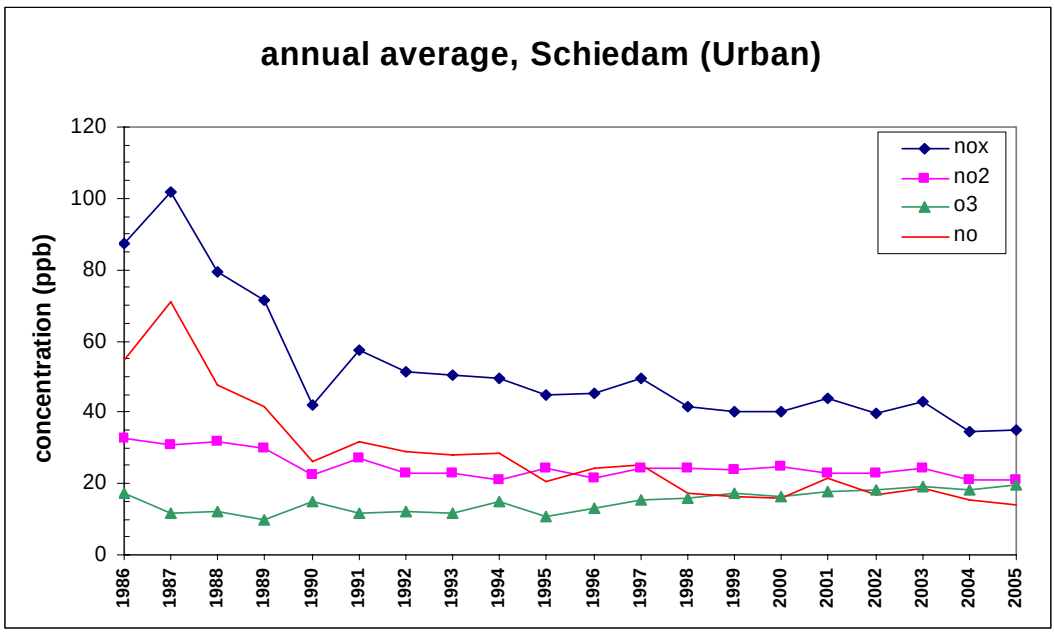

Figure 1: Annual trend of NO, NO_2, NO_x and O_3 over the period 1986 – 2005 at the urban background station in Schiedam.

The trends in Figure 1 show that NO_x concentrations in Schiedam decreased considerably, especially in the period from 1987 to 1991. This is attributed to the large scale introduction of 3-way catalysts, which significantly reduced NO_x emissions of road traffic. From 1991 onwards to 2005, the NO_x concentrations further decreased gradually with 30% in 15 years. The trends in Figure 1 also shows that in the 4 year period 1987 to 1991 NO_2 decreased by about 10%. This is in contrast with the remaining period of 15 years from 1991 to 2005, when NO_2 concentrations hardly decreased in Schiedam. *Hence, at the urban background station in Schiedam, the annual NO_2 concentration remains at around 35 µg/m^3 and consequently the annual NO_2 air quality limit value will be often exceeded near heavy traffic locations.* Thus, DCMR is interested in factors controlling the urban NO_2 concentrations in order to develop a cost-effective abatement strategy.

One hypothesis for the constant NO_2 concentrations, despite the reduction in NO_x emissions, is an increase in ozone concentration at the urban background. Another hypothesis is an increase in direct NO_2 emissions due to the introduction of oxidising particle trap catalysts for diesel vehicles. The trend analysis in Figure 1 shows indeed an increase in ozone concentrations at the urban background station in Schiedam from 1991 to 2005 due to the reduction of NO_x emissions. So, increasing ozone at urban background seems to be a likely explanation for the trend of NO_2 concentrations.

A method to research the possible contribution of increasing direct NO_2 emissions by local sources is based upon the use of the "oxidant" concentration. The oxidant concentration (Ox) is the sum of the ozone and NO_2 concentrations. Under the NO_x/ozone equilibrium Ox should be constant. Due to direct NO_2 emissions by local sources in an urban area, Ox increases when comparing urban Ox to regional Ox. Thus, the "delta Ox" from an urban and a regional background station pair over the years is a method to identify direct NO_2 emissions: Carslaw, 2005. The method is demonstrated using Rotterdam hourly monitoring data and consists of the following steps:

1. the delta Ox and NO_x of Schiedam (urban background) minus Zierikzee (regional background) are calculated from the monitoring data for each year 1986 to 2002;
2. the delta Ox is plotted against the delta NO_x per year;
3. the slope of the regression line is the amount of direct NO_2 emission per year;
4. the slope for each year is plotted against each year from 1986 to 2002: this trend quantifies the increasing direct NO_2 emissions in this period.

The results of this exercise are shown in Figure 2.

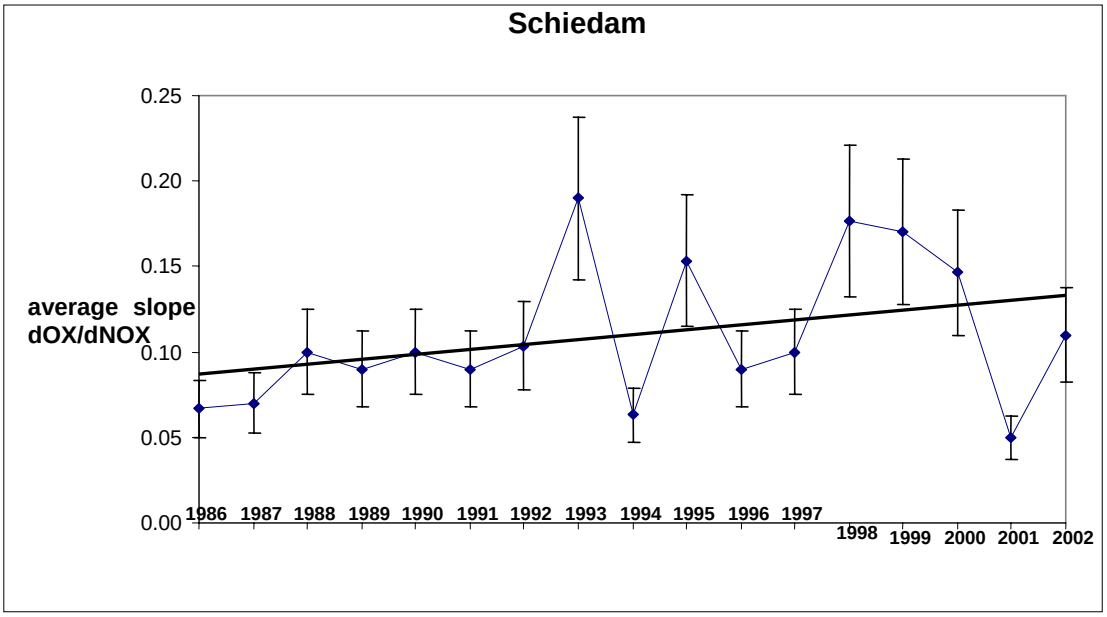

Figure 2: The trend in direct NO_2 emissions at the urban background at Schiedam (Rotterdam) from 1986 to 2002 including the average standard deviation with in 95% confidence level.

Despite some variation form year to year, the method results in a positive trend of direct NO_2 emissions in 1986 of 9 % to 13% in 2002. In London a similar trend was identified from 5-6% direct NO_2 road traffic emissions in 1997 to 17% in 2003: Carslaw, 2005.

The importance of both developments "$NO/NO_x/O_3$-equilibrium" and "direct NO_2 emissions" relative to each other was explored for the data in 1986 and 2002 in Schiedam and Zierikzee:

1. *regional background at Schiedam;* From the measured NO_2 at Zierikzee, the regional background at Schiedam is estimated;
2. *direct NO_2 emission at Schiedam;* The fraction of direct NO_2 estimated from the trend analysis is respectively 9% (1986) and 13% (2002). The contribution of direct NO_2 emission (to NO_2 concentrations at Schiedam) can be estimated from the delta NO_x (Schiedam minus Zierikzee data) in ppb: 68.1 (1986) and 24.1 (2002) multiplied with the fraction NO_2;
3. *NO to NO_2 conversion at Schiedam;* The fraction of NO from NO_x emissions is 91% (1986) and 87% (2002). Using the delta NO_x at Schiedam, this results in a concentration of 62.0 (1986) and 21.0 (2002) ppb NO. The measured *regional* background at Zierikzee is 6.3 (1986) and 4.9 (2002) ppb NO. This results in an expected value of NO at Schiedam of 68.3 (62.0 + 6.3; 1986) and 25.9 (21 + 4.9; 2002) ppb NO. The *measured* values for NO at Schiedam were 54.6 (1986) and 16.9 (2002). The difference between expected and measured values is an estimate of the conversion of NO into NO_2: 13.7 (1986) and 9 (2002) ppb NO_2;
4. *total NO_2 concentration at Schiedam;* The total NO_2 concentration at Schiedam in 1986 and 2002 can be estimated from the sum of the regional background, direct NO_2 emissions and conversion of NO into NO_2. Comparison with measured data provides an indication of the validity of the approach.

In Table 1, a summary is provided of the aforementioned approach.

Table 1: The NO_2 budget for Schiedam in 1986 and 2002 based upon the regional background, direct NO_2 emissions and NO to NO_2 conversion, and compared with measured data.

	1986 **ppb NO_2**	**2002** **ppb NO_2**
1. Contribution *regional* NO_2 background	13.8	10.7
2. Contribution direct NO_2 emission	6.1	3.2
3. Contribution NO/O_3 conversion into NO_2	13.7	9
Concentration NO_2 *estimated* by the sum of 1-3	**33.6**	**22.9**
Concentration NO_2 *measured*	**33.5**	**23.1**

The estimated and measured concentrations at Schiedam in Table 1 show good agreement which underlines the validity of the approach. Table 1 illustrates that chemical conversion is the dominant contributor of local sources to NO_2 relative to direct NO_2 emissions.

4. CONCLUSION

Two hypotheses have been explored to explain the constant trend in measured NO_2: 1.) reduction of NO_x emissions results in lower NO concentrations but due to the high ratio NO/O_3 less to lowering NO_2 concentrations and 2.) increasing direct NO_2 emissions by traffic results in higher NO_2 concentrations. The data confirms that the NO/O_3 ratio is relatively high at urban background sites, and this causes NO_2 concentrations to decrease less than NO_x concentrations. The data furthermore indicate, that direct emission of NO_2 by traffic has increased from 9 to 13% (as fraction of the NO_x emission) but the total amount of emitted NO_x has decreased substantially. Thus, chemical conversion of emitted NO_x remains the dominant contributor of local sources to NO_2.

5. ACKNOWLEDGEMENT

The Air4EU project was financed under the FP6 programme of the European Commission. The Environmental Agency – DCMR for the Rijnmond area is acknowledged for providing the necessary monitoring data to perform the trend analysis. The data at the regional stations in Zierikzee was retrieved from the National Air Quality Monitoring Network managed by RIVM in the Netherlands.

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BASIC DATA ASSIMILATION METHODS FOR USE IN URBAN AIR QUALITY ASSESSMENT

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ABSTRACT

In this paper a number of basic data assimilation methods are applied to improve maps of air quality for the city of Prague. Annual mean concentration fields for PM_{10}, SO_2 and NO_2 provided by the ATEM model are combined with data from 12 monitoring sites available for the year 2003. The methodologies employed include linear regression, kriging, kriging of residuals and a form of Bayesian assimilation. Uncertainty maps are also produced. These methods can be implemented using standard software packages, making them accessible to city authorities as well as research institutes.

1. INTRODUCTION

There are a number of methodologies available for combining monitoring and modelling data. Many of these are complex applications involving data assimilation techniques that require direct interaction between model calculations and observed data. Examples of these are variational methods, e.g. 4Dvar, Kalman filters and other particle filter methods. There are also other methods that can be applied 'offline' where pre-calculated model fields can be adjusted using monitoring data. These methods do not require interaction with the model and make use solely of the resulting model fields. Examples of such methods are 3Dvar, optimal interpolation, regression modelling and kriging. Such methods are easier to implement without having to interact with complex models and some can be implemented using standard software.

As part of the 6'th framework programme Air4EU, a number of these simpler methods have been explored and this paper addresses the use of these for the city of Prague (Denby, 2007). Other recent studies using similar statistical methods, such as kriging and regression, on varying spatial scales include Kassteele et al. (2006), Horálek et al. (2005) and Blond et al. (2003).

The aim of this paper is to apply and test basic data assimilation methodologies that can be used to combine monitoring and models to improve pre-calculated model fields and produce more reliable maps of concentrations for urban regions. Pre-existing model fields of annual mean concentrations for PM_{10}, SO_2 and NO_2 are used in combination with 12 monitoring sites for the city of Prague. A number of assimilation methods are tested and the results are objectively assessed using the cross-validation root mean square error (RMSE). In addition to the maps of concentrations fields the uncertainty of these maps is discussed and maps showing uncertainty are calculated and displayed.

2. METHODOLOGY

A number of assimilation methodologies are applied to the available model fields and monitoring data for the pollutants PM_{10}, SO_2 and NO_2. These methods include:

1. Linear regression of the model fields
2. Ordinary kriging of the observed fields
3. Kriging of the residual fields (observed – model)
4. Bayesian combination of kriged observations and regression model fields

Model and observations

The ATEM model is a statistical Gaussian model that produces annual mean pollutant fields at a resolution of 250 x 250 m for the city of Prague. The model is originally based on the US EPA model IST2. Emissions for the model include point, line and area sources within the model regions as well as a number of point sources external to that region. A set of statistical wind roses and stability classes are used for the calculation based upon results from a local mesoscale model. More information concerning the model can be found in Brechler (2000). Model fields used in the calculations for NO_2 can be seen in figure 2a.

12 monitoring stations are available for the calculations. Their positions and concentrations are shown in the maps in figure 2. The stations used are assumed to be representative of the modelling scale, that being of a 250 x 250 m region surrounding the monitoring station.

Objective assessment

In order to assess the results a number statistical parameters are used. However, central to the assessment is the use of cross validation (CV) as a comparative test. CV involves carrying out the assimilation using all the observations except one. The excluded observation is compared to the calculated one and this process is rotated around all the observations. To give a single value assessment of the quality of the assimilation

method the root mean square error (RMSE) of the CV errors is used. CV is also applied to all the methods, including the regression analysis.

Linear regression
Linear regression can be used to produce regression models that relate model calculated concentrations to observed quantities. Linear regression produces a regression model that minimises the RMSE between the model and the observations. Linear regression does not take into account spatial variations and is an appropriate assimilation technique when intrinsic model bias is due to spatially homogenous errors in the model calculations such as emission factors, dispersion and meteorological model formulations and regional background contributions. However, linear regression will not help improve results when the variations are local in nature.

Ordinary kriging and residual kriging
Kriging is an often used statistical interpolation method in the geosciences. It is based upon the assumption that there is a spatial correlation between points in space that is related to the distance between the points. This spatial correlation is described by a spatial variance function, called the semi-variogram. The interpolation is carried out by weighting the nearby measurement points so that the variance at the interpolation point is minimised. In other words, the interpolated point is statistically the most likely one. Defining the sem-variogram is thus critical to the method.

Kriging works best when the spatial variability occurs on scales larger than the distance between measurement points. In an urban setting where spatial variation is much finer in detail, kriging by itself is less likely to give useful results. In the current application the kriging parameters are based on the use of the model variance, rather than observed variance, since the spatial resolution is higher for the model fields. An alternative method, also addressed in this study, is to set the semi-variogram parameters by selecting those that minimise the cross validation RMSE. For a general description of kriging methods see Cressie (1993).

Ordinary kriging assumes some form of spatial stationarity to the field, i.e. there are no spatial trends present. This is often not the case and so a number of methods for accounting for spatial trends, or 'external drift', can be applied. The method used here is to assume the regression model field represents the general character of the field, i.e. the drift, and to apply kriging to the residuals (also termed innovation). This has been shown in other studies (Horálek, 2005; Blond et al., 2003) to be an effective method for improving mapped concentration fields on regional scales.

Bayesian assimilation
Another assimilation method, which takes into account the uncertainties in both modelled and observational fields, is to use Bayesian statistics to combine these fields. Optimal interpolation and variational methods are based on this type of statistics. In this case the observational field, described by the kriging interpolation, is combined with the regression model field. Both these fields have associated uncertainty, for the observational field this is the kriging variance and for the regression model this is based on the normalised RMSE scaled by the model concentrations. If the uncertainty is assumed to be Gaussian in nature, ie. the probability density function (PDF) is normally distributed and unbiased, then an assimilated field can be produced by combining the observational and model field in a straightforward manner.

$$M_{BA}(x,y) = \frac{\sigma_O^2 M(x,y) + \sigma_M^2 O(x,y)}{\sigma_O^2 + \sigma_M^2} \qquad (1)$$

$$\sigma_{BA}^2(x,y) = \frac{\sigma_O^2 \sigma_M^2}{\sigma_O^2 + \sigma_M^2} \qquad (2)$$

where σ is the standard deviation at that point in space of the model (M) and observational (O) field. The resulting uncertainty, in terms of standard deviation of the assimilated field, is given by equation 2. The assimilated concentration field will then be a weighted combination of the observed and modelled field, the weight given to the assimilated values being dependent on the relative uncertainties of the observed and modelled fields.

Uncertainty mapping
Uncertainty maps are created in different ways, dependent on the methodology employed. For model and linear regression model fields the uncertainty maps are constructed by determining the CV RMSE. This is then normalised with the mean model concentration (taken at the observational sites). This normalised uncertainty is then scaled by the model concentrations. For kriging and residual kriging methods the calculated variance is used to map the uncertainty. For the Bayesian assimilation equation 2 is used.

3. RESULTS AND DISCUSSION

Details of the analysis can be found in Denby (2007) and the results are summarized here.

Cross validation RMSE

The normalised CV RMSE calculated for the different methods is shown in figure 1 and compared to the standard deviation of the observations. This last value indicates the quality of the 'simplest model', that being the mean of the observations. Using the CV RMSE as a measure of the uncertainty in the assimilation, the results indicate that improvement is obtained using all the assimilation methods, in regard to the initial model calculations. The application of regression significantly reduces the uncertainty and this can be, but is not always, reduced by the extra application of residual kriging or Bayesian assimilation. The use of pure kriging gives similar CV RMSE values for SO_2 and NO_2 but gives higher values for PM_{10}.

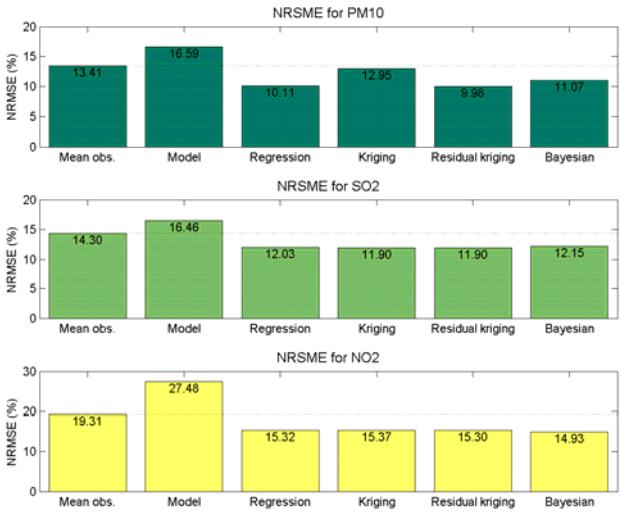
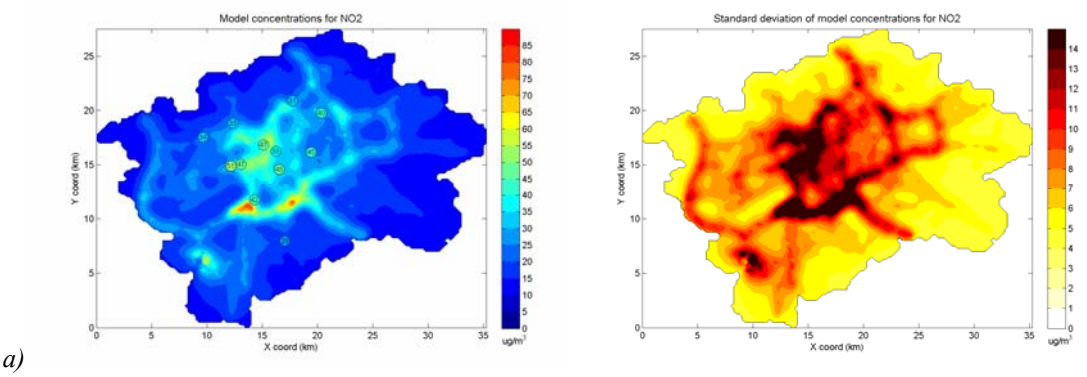


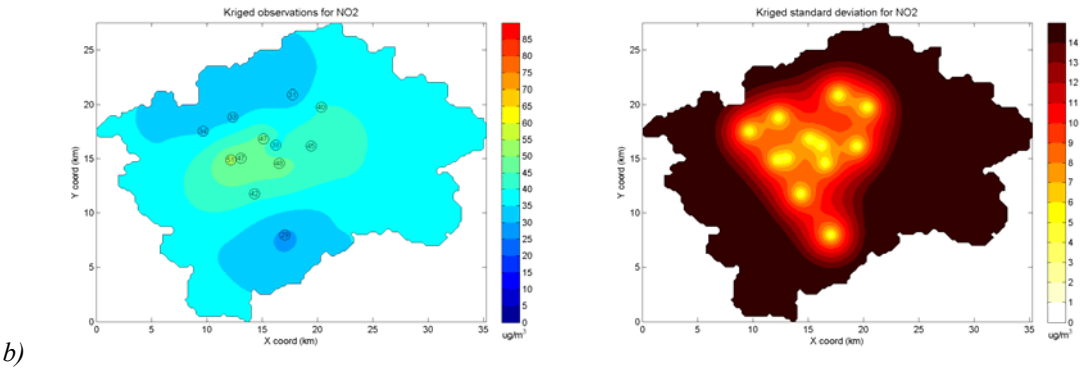
Figure 1. Summary of the cross validation RMSE of the assimilation methods tested showing the normalised RMSE as %. The thin horizontal line corresponds to the observational standard deviation. Top PM_{10}, middle SO_2 and bottom NO_2.

Concentration and uncertainty maps

Figure 2 below shows maps of the concentration and uncertainty fields of NO_2 for the initial model field, kriged field, the regression model field and the Bayesian assimilated field. These maps provide valuable information concerning the quality of the mapping method.



a)



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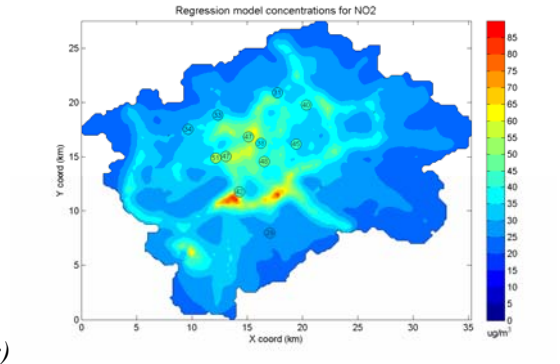

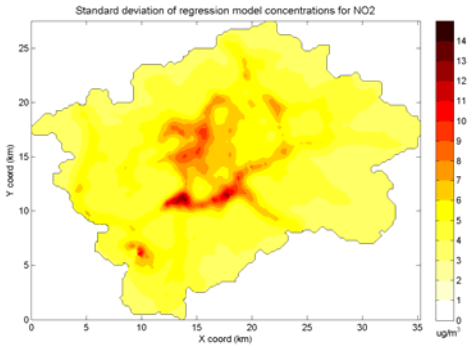

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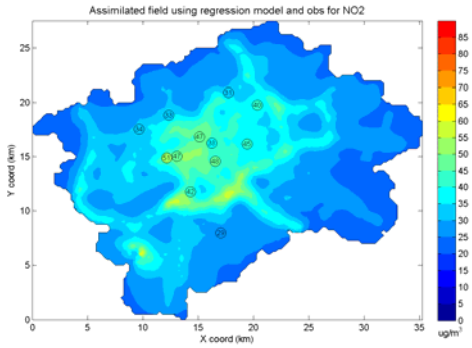

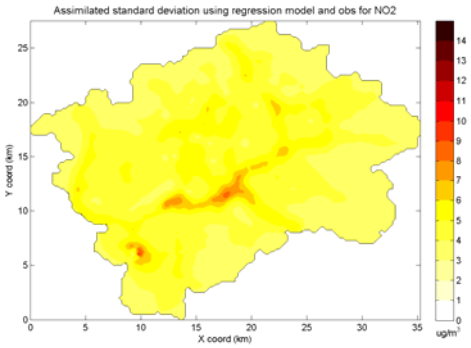

Figure 2: Annual mean concentration maps (left) and uncertainty maps showing standard deviation (right) for NO_2. a) Model field, b) Ordinary kriged field, c) Model regression field and d) Bayesian assimilation field. Units are μgm^{-3}.

Clearly the kriged field has high uncertainty far from the observations where no data is available. The regression model field has lower uncertainty than the initial model field, due to the removal of general model bias. The Bayesian assimilation field shows reduced uncertainty in the areas surrounding the monitoring stations but with the same uncertainty as the regression model field far from the observations.

4. CONCLUSIONS

This study has applied basic data assimilation methods to the urban scale. The results indicate that the simple application of linear regression, when sufficient stations are available, can significantly reduce the uncertainty in the model concentration fields. In addition the use of residual kriging or Bayesian assimilation of kriged fields can help spatially reduce the uncertainty. This case study has tested these methods on only one urban area, that being the city of Prague. To assess further the usefulness of these methods on the urban scale then applications in other cities using other data and models is required.

5. ACKNOWLEDGEMENTS

Air4EU is has been funded by the EU 6'th framework programme and is part of the CLEAR cluster.

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Estimation of modelling uncertainty for air quality assessment: the AIR4EU Berlin case

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ABSTRACT
Air quality models need to be properly evaluated before their predictions can be used with confidence, because model results often influence decisions with consequences on health and economy. A systematic description of the methodologies for modelling uncertainty estimation was performed and discussed in the scope of the AIR4EU European project based on a bibliography review and addressed according to the end users needs. This methodology consists first on the estimation of model uncertainty and comparison with the data quality objectives defined by the EU legislation and, consecutively, a complementary analysis of the total model uncertainty based on a statistical analysis. The developed procedure was applied to the city case of Berlin, considering both mesoscale and local scale modelling applications. The uncertainty exercise shows an incomplete definition of the Quality Assurance/Quality Control procedures for air quality modelling by the present EU legislation. The performed statistical analysis suggests the use of a specific group of indicators in order to complete the model uncertainty estimation.

1. INTRODUCTION

The uncertainty concept is one of the crucial points of Quality Assurance/Quality Control procedures that should provide quantitative information about the modelling precision, identifying the uncertainty sources and their potential reduction. The present European legislation defines the Modelling Quality Objectives as an acceptability measure, to guarantee good model performance and reliable modelling results for decision makers. However, a practical application of these requirements and interpretation of the uncertainty analysis results based on the recommended methodology is ambiguous, and in some cases incomprehensible for non-expert users. The development of a consistent procedure for uncertainty evaluation is still a challenge for the scientific community. Model quality measures described in the EU Directives can be interpreted as the relative maximum error without timing, which is the largest concentration difference of all percentile differences normalized by the respective measured value. An alternative relative error at the percentile correspondent to the allowed number of exceedances of the limit value was suggested and tested by Stern and Flemming (2004), showing that it is more robust and also evaluates the model performance as required. Besides the legislation procedures, a statistical analysis is suggested to evaluate the model performance and to estimate uncertainties, comprising a set of parameters, that give information about the ability of the model to predict the tendency of observed values, errors on the simulation of average and peak observed concentrations, and type of errors (systematic or unsystematic): the correlation coefficient (r), the root and normalized mean square errors (RMSE and NMSE) and the systematic error (Bias) (Borrego *et al.*, 2005).

2. METHODOLOGY

The aim of this case study is to provide an example of the methodology described in the Air4EU-M2 cross cutting report (Borrego *et al.*, 2006) regarding the estimation of air quality modelling uncertainty. An uncertainty analysis was performed to the modelling results of the Berlin case, concerning the urban and hot spot scales for the main critical pollutants. According to the basic recommendations, defined in the Air4EU-M2 report, the estimation of total model uncertainty will be determined through comparison between model predictions and air quality observations, based on the Air Quality Framework Directive (FWD) settlements and on statistical parameters. The referred recommendations consider three levels of application adaptable to end-users needs, goals and ability. The first level is a simple qualitative analysis. The estimation of total model uncertainty based on statistical parameters and on FWD settlements is covered by the second level. In order to improve model performance, it is necessary to investigate the contribution of the different components of total model uncertainty. Therefore, the third level details the total model uncertainty, through the estimation of its different components. The variability is addressed here as a first step. For a more complete evaluation, sensitivity analysis to model and input parameters should also be performed. This sequence of recommendations should be regarded in these three levels of complexity and could be applied by end-users according to their desire and need of detail (Borrego *et al.*, 2005). For the Berlin case study the second level of recommendations will be applied. The alternative model error RPE (Relative Percentile Error) will be considered to estimate model uncertainty, corresponding to the allowed number of exceedances of the limit value normalized by the observation. The Berlin case study is composed by two different scale model approaches. Simulations were performed with the RCG model over the urban area of Berlin, with 4x4 km^2 grid resolution. The Gaussian multi-source dispersion model IMMIS^{net} was used for the local scale modelling simulations over Berlin at specific main roads, and with a resolution of several meters. The modelling and PM_{10} and NO_2 monitoring data were provided by the Senat Berlin for 2002 year.

3. RESULTS AND DISCUSSION

3.1 Local scale

A total of 121 air quality measuring devices were located at different heights over the study domain according to Figure 1, providing hourly measured concentrations of NO_2 and PM_{10} pollutants during 2002 over an XZ plane.

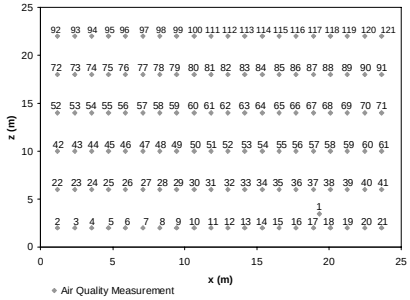

Figure 1. Location of the air quality measuring devices over the study domain for NO_2 and PM_{10} pollutants (XZ plane).

Figure 2 presents the RPE values for each pair of observed/modelled data of the 121 measuring points, for NO_2 and PM_{10}, respectively. In the case of NO_2, RPE is estimated for percentile 99.79, corresponding to the 18th maximum hourly mean within one calendar year (2002 year), and according to the target value defined for the protection of human health. For PM_{10}, the RPE is based on the annual average concentration, according to the annual limit value for human health protection. In both figures are indicated the data-quality objectives defined by the FWD for modelling uncertainty: 50-60% for NO_2 hourly mean and 50% for PM_{10} annual averages. For NO_2, the RPE values increase with height, concluding that the Modelling Quality objectives of the FWD are overpassed for heights above 15 m (RPE>50-60%). This will mean that the applied model has the capability to predict well the transport and formation of NO_2 near the ground. The high discrepancy between observed and modelled values over the 15 m can justify RPE values higher than 1.0. For PM_{10} most of the RPE values determined are above the modelling quality objective of 50%. These results allow to conclude that the model has a deficient performance concerning the modeling of PM_{10} pollutant. Moreover, these modeling results should not be considered with care before their use for air quality modeling assessment. Again, it is possible to observe lower RPE values near the ground that will increase with height.

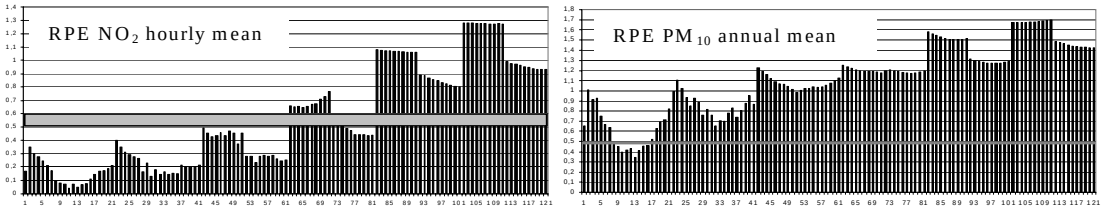

Figure 2. RPE hourly mean for NO_2 and RPE annual mean for PM_{10} estimated for each air quality measuring point.

A statistical analysis was performed using the referred set of parameters, in order to give some additional information to the model uncertainty analysis. As an example, in Figure 3 are plotted some of the statistical parameters for NO_2. The correlation coefficient (r) is significantly higher for PM10 (>0.51) and less for NO_2 (>0.48), suggesting that the model is not able to simulate correctly all the physic processes involved in the dispersion of the above mentioned pollutants. The RMSE presents an average of 50 $\mu g.m^{-3}$ for NO_2 and 10 $\mu g.m^{-3}$ for PM_{10}, showing that the magnitude of the errors between observed and modelled values are lower for PM_{10} than for NO_2. The relative error, NMSE presents an average of 0.24 for NO_2 and 0.79 for PM_{10}, indicating that besides the absolute error is higher for NO_2, the normalized error is lower for this pollutant. Finally, the Bias presents averages of -40 and -9.6 $\mu g.m^{-3}$ for NO_2 and PM_{10}, respectively, with values always negative for both pollutants, suggesting an overestimation of modelled concentrations.

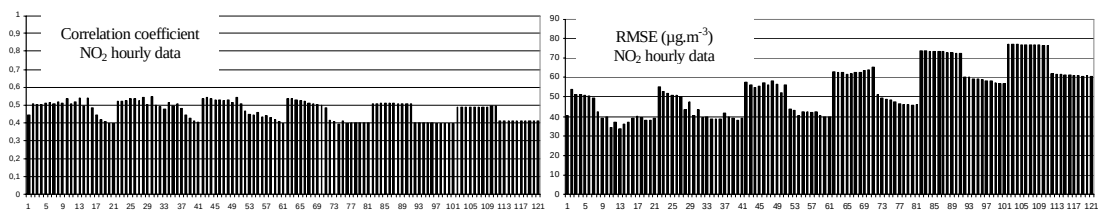

Figure 3. Statistical parameters for the local scale simulation for NO_2 pollutant.

3.2 Urban scale

The German HOVERT campaign dedicated daily measurements in a high density network (61 monitoring sites) for one year period (September 2001 to September 2002) providing valuable information to perform a thorough model evaluation and being the database for the presented uncertainty analysis of the RCG model. This model, developed at Free University of Berlin with the support of the German Environmental Protection Agency, is a Chemical-Transport-model specially designed for the regional and urban scale, and was applied for Berlin area over a domain that covers 11º-15ºW of longitude and 51º-53,5ºN of latitude, with 4x4 km^2 resolution. According to the model data quality objectives defined by the EU legislation, the second level of the model uncertainty estimation consisted in the calculation of the RPE parameter. In Figure 4 are plotted the RPE values concerning O_3 and PM_{10}. In the case of ozone, RPE is estimated for the percentile 99.7, corresponding to the 26th maximum daily 8-hour mean within one calendar year (2001 year), and according to the target value defined for human health protection. For PM_{10}, RPE is based on the annual average concentration, according to the annual limit value for human health protection. In both figures is indicated the data-quality objectives defined by the FWD for modelling uncertainty (50%). Results show that, for both pollutants, the RPE values are below 50%, fulfilling the data quality objectives for allowed uncertainty of model assessment. There is only an exception regarding a specific monitoring site for ozone that presents a RPE close to 0.5, which can be justified by the traffic type and urban influence of the station (not representative of the model grid resolution). In summary, according to the FWD, the RCG model is able to simulate appropriately and with an acceptable level of uncertainty the Berlin case study at urban scale.

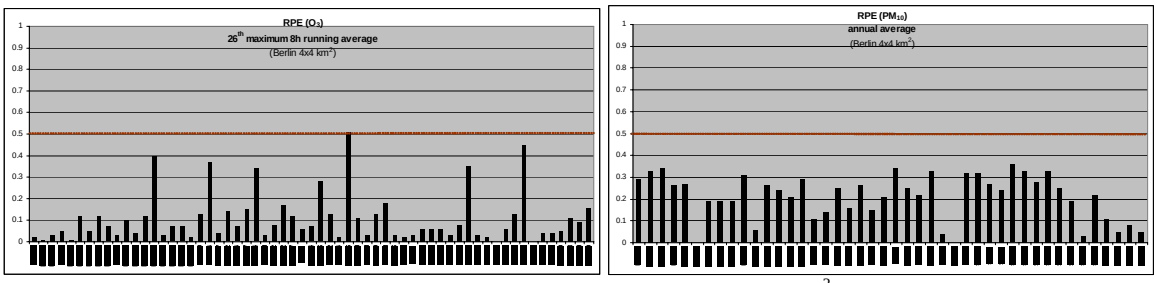

Figure 4. RPE estimated for each O_3 and PM_{10} monitoring station, for the 4x4km^2 Berlin case study simulation.

In Figure 5 are plotted some of the statistical parameters for ozone O_3. The correlation coefficient (r) is significantly higher for ozone (>0.75) and less for PM_{10} (>0.6), suggesting that the model is able to simulate the physics and chemistry processes involved in the O_3 formation and also (but more deficiently) the aerosols chemistry. The RMSE presents an average of 15-25 µg.m^{-3} for O_3 and 10-20 µg.m^{-3} for PM_{10}, showing that besides the lower correlation, the magnitude of the errors between observed and modelled values is lower for PM_{10} than for O_3. Concerning the relative error, NMSE presents an average of 0.2-0.25 for O_3 and 0.4-0.85 for PM_{10}, indicating that besides the absolute error is higher for O_3, the normalized error is lower for this pollutant. This could be due to the PM_{10} values that are usually lower than O_3 concentrations. Finally, the Bias presents an average range between -10 and 5 µg.m^{-3} for O_3 and is always positive for PM_{10}, suggesting that there is an overestimation of the PM emissions, but not existent regarding ozone precursors (that could inclusive be underestimated). It should be noted that there are some exceptions to these average ranges, all explained by the monitoring station traffic type or strong urban influence.

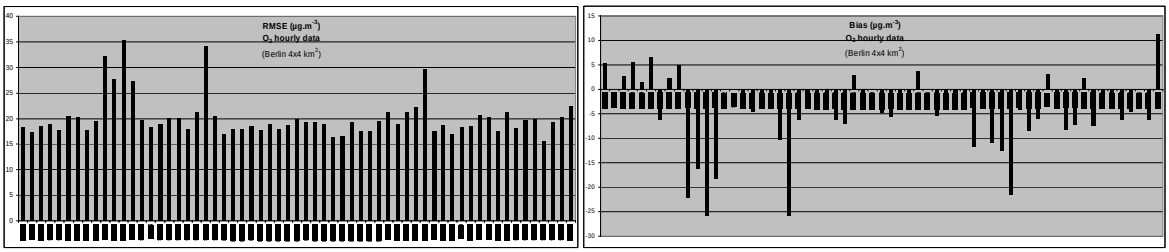

Figure 5. Statistical parameters obtained for the urban scale Berlin simulation, for each O_3 monitoring station.

According to the basic recommendations, since there is a considerable high density monitoring network over the study area, it is suitable to spatially interpolate the error data. In Figure 6 is presented, as an example, the spatial mapping (using Kriging interpolation method) of the statistical parameters at urban scale, for O_3 and PM_{10}. For both cases, it is possible to verify that each error parameter can be used as a (relative or absolute) indicator of the model uncertainty. In fact, the spatial distribution is similar for all the variables presented,

reflecting the same pattern when identifying the areas with higher uncertainty levels. The ideal should be to combine the different maps to produce a final one for model uncertainty spatial representation.

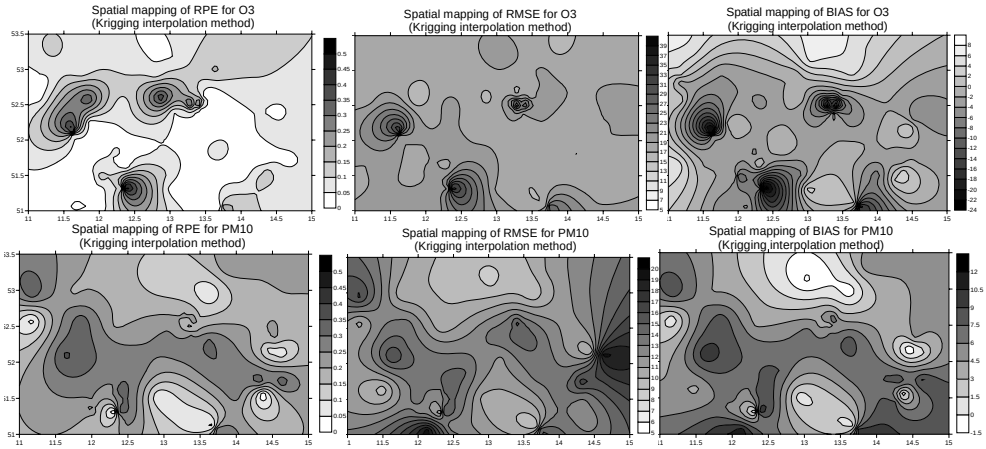

Figure 6. Spatial mapping of model uncertainty: kriging interpolation of RPE, RMSE, r and Bias for O_3.and PM_{10}.

4. CONCLUSIONS

According to the basic level of recommendations, defined in the cross cutting report (Air4EU - M2), the total model uncertainties for Berlin case were determined through comparison between model predictions and a wide air quality monitoring network for the urban and hot spot scale. The first level comprises the application of an alternative model error measure to the FWD and the use of a set of statistical parameters. Results of the first step point to the fulfilment of the modelling acceptability criteria defined in the FWD, for all the analysed pollutants for the urban scale and partially for NO_2, at the local scale. For the same scale and for PM_{10} most of the RPE values determined are above the modelling quality objective of 50%. In the second step, the statistical analysis, comprising a set of parameters, gave information about the ability of the model to predict the tendency of observed values (r), relative and absolute errors on the simulation of average and peak observed concentrations (RMSE and NMSE), and type of errors (Bias). This analysis shows that the urban scale model is able to predict concentrations for the Berlin case with correlation factors higher than 0.6 for O_3 and for PM_{10}. For the local scale, the averaged correlation factor is 0.48 and 0.51 for NO_2 and PM_{10} pollutants, respectively, indicating that the model is not able to simulate correctly all the physic processes involved in the dispersion of the referred pollutants. Besides that, the models average error shows no significant discrepancies between model values and observed data. At the urban scale the Bias shows no model tendency for some pollutants, suggesting that local phenomena could be responsible for model errors at each specific monitoring site, but a clear model overestimation for PM_{10}. The uncertainty spatial mapping showed that all the analysed indicators are adequated to perform the spatial representation of the uncertainty. Considering that the basic level of recommendations is completed for this case study, it is recommended for future work to investigate the stochastic contribution to the total model uncertainty and to perform a sensitivity analysis or model intercomparison to evaluate the different model modules.

5. ACKNOWLEDGEMENTS

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UNCERTAINTY AND REGIONAL AIR QUALITY MODEL DIVERSITY: WHAT DO WE LEARN FROM MODEL ENSEMBLES?

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ABSTRACT

Recently several Clean Air For Europe (CAFE) regional air quality projects were carried out in order to predict the impact of emission control policies, using an ensemble of models. This was the methodology used within the two projects CITYDELTA (phase 2), where simulation of urban and suburban air quality was studied in a major intercomparison over 4 european cities, and EURODELTA, where the regional scale was studied. Pollutants considered were ozone and PM10. After describing and intercomparing the skill of these models over long simulation periods, we examine whether this ensemble provides a spread of concentrations of ozone and aerosols that is representative of the simulated uncertainty. Using tools borrowed from the evaluation of ensemble weather forecasting, we analyze statistics of simulated concentrations over an entire summer season. Although the ensemble may have biases, the distribution of simulated concentrations is representative of the uncertainty.

For ozone the ensemble spread is partly due to fluctuations resulting from different model formulations and input data, but also to the spread between individual model systematic biases. The variability of the uncertainty is fairly well reproduced by the ensemble. The skill of the ensemble in predicting uncertainty is also demonstrated by evaluating the reliability of probabilistic prediction of threshold exceedances. These results indicate that the ensemble can be used for evaluation of the impact of emission reduction policies and its uncertainty.

1. MODEL INTERCOMPARISONS IN CAFE PROGRAM

Predicting future regional or urban air quality in the future is a major challenge for health-related questions in our societies. The evolution of air quality depends on many factors, among which the evolution of the development worldwide, the associated pollutant emission changes and the subsequent evolution of the global atmospheric composition. In Europe, future air quality for about a decade appears affected by a combination of the evolution of continental emissions and changes in remote emissions (Parrish et al., 1993) and baseline concentrations (Szopa et al., 2006). While the latter factor is impossible to control at the European scale, the former mainly results from concerted efforts put into environmental policies. The evaluation of such policies efficiencies can only be carried out with the use of numerical models.

In support of the European Union Clean Air For Europe (CAFE) Programme and in the framework of the Convention on Long Range Transport of Atmopheric Pollution, (CLRTAP United nations – Economic Commission for Europe), several projects have been set up in order to evaluate the regional responses to emission reduction scenarios. These projects strongly rely on long-term air quality simulations using several chemistry-transport models. CITYDELTA, the first project (Cuvelier et al., 2007), and was devoted to the evaluation of emission scenarios for 2010 at the scale of European cities. EURODELTA is the regional

counterpart of the former project (Van Loon et al., 2007), at a European scale. In both projects several models are used, giving one access to the spread of possible modelling responses to emission scenarios.

2. THE USE OF MODEL ENSEMBLES

Using an ensemble of models, instead of a unique model, in order to predict air quality in the future, actually gives two new informations:

(i) The average over this ensemble of responses is a new response by itself, which can smooth the errors of individual models, and may therefore be more reliable.
(ii) The spread of the ensemble may represent the uncertainty in our calculations.

Unfortunately it is not possible to verify future responses of several scenarios and their associated uncertainties directly. However, a first evaluation of whether these new possibilities, offered by model ensembles, are realistic can be by simulating past periods. For point (i) a direct intercomparison between individual models skills and ensemble mean model skills can be carried out using routine air quality observations. For point (ii), the evaluation of the relation between models simulation spread and the uncertainties can be made using methods developed in the framework of ensemble weather forecasting (Molteni et al., 1996). Here we summarize the results obtained by Van Loon et al. (2007) and Vautard et al. (2006), which address both questions.

3. THE SKILL OF THE MODEL ENSEMBLE AVERAGE

Within the EURODELTA project, 7 regional air quality models have been run over an extended period of time (6 months), the year 2001 (see Van Loon et al., 2007). Figure 1 shows, as an example, the mean diurnal cycle of Ox=NO2+O3, over about a hundred of monitoring sites in Europe, for each model, the ensemble mean and the observations. The model ensemble diurnal cycle is clearly closer to the observations than any individual model, because model biases cancel each other when averaging. The dispersion of biases, with negative and positive values, was also found in the intercomparison of the models used at city scale in the CITYDELTA experiment (Vautard et al., 2007).

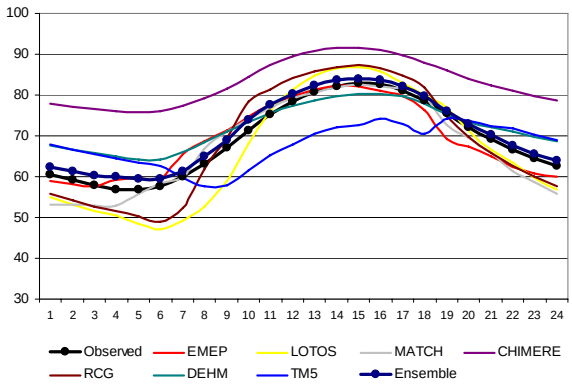

Figure 1: Yearly average diurnal cycle of $Ox=O3+NO_2$, in g/m^3, for all models, and their ensemble mean, averaged over all monitoring stations. After Van Loon et al. (2007).

The better skill of the ensemble average as compared to that of individual models is not always found in diurnal cycles, but also in root mean square error and correlation for almost all seasons, as shown for instance (for correlation) in Table1. Other statistics can be found in Van Loon et al. (2007).

	daily average					daily maximum				
	year	DJF	MAM	JJA	SON	Year	DJF	MAM	JJA	SON
EMEP	0.72	0.67	0.55	0.50	0.55	0.75	0.60	0.59	0.61	0.53
LOTOS	0.70	0.49	0.54	0.49	0.43	0.76	0.47	0.70	0.66	0.48
MATCH	0.80	0.68	0.66	0.60	0.67	0.81	0.58	0.68	0.7	0.61
CHIMERE	0.76	0.62	0.58	0.64	0.60	0.84	0.62	0.71	0.77	0.62
RCG	0.71	0.58	0.59	0.52	0.36	0.76	0.56	0.70	0.61	0.44
DEHM	0.64	0.45	0.41	0.56	0.31	0.75	0.45	0.60	0.68	0.45
TM5	0.67	0.69	0.44	0.35	0.62	0.72	0.63	0.47	0.51	0.58
Ensemble	0.79	0.74	0.66	0.68	0.58	0.84	0.69	0.76	0.78	0.59

Table 1: Correlation coefficients for daily average and daily maximum O_3. After Van Loon et al. (2007).

4. SPREAD AND UNCERTAINTY

Does the spread of the model ensemble give any information on uncertainty of simulations? In the best case with respect to estimation of uncertainty, modellers have, independently from one another, selected model options or parameter values with a range of choices that is representative of the uncertainty on these parameters. In the worst case, all modellers have selected the same options or parameter values, or missed the same key processes. In the former case one expects observations to lie within the range of simulated concentrations, while in the latter observations should be "outliers" of the simulations distribution. Therefore the consistency between observations and the distribution of ensemble simulated concentrations measures our ability to represent uncertainty of simulations.

In order to measure the representativeness of the ensemble, we borrowed tools from the evaluation of uncertainty estimates using ensemble weather forecasting (see e.g., Jollife and Stephenson, 2003). The rank histogram (Figure 2) measures the distribution of the rank of the observation within the ensemble of simulated values. In this figure the example bears on the ozone daily maxima for the summer season. In the perfect cased, the distribution of the observation within the ensemble is equiprobable, and the rank histogram must be "flat". The figure shows that the first two bins (0 and 1) have a number of counts much larger than the other bins, reflecting a difficulty of models to simulate low daily maxima, which is a bias of the ensemble. This bias could result from the larger modelling effort put for skilful prediction of high concentrations rather than of lower ones. This "ensemble bias" can be removed, at each station, by subtracting the average difference between simulated (all models together) and observed ozone daily maxima. After this operation, daily maxima ensembles are only shifted but their distribution, spread and model rank are unchanged. In this case, the rank histogram becomes flatter.

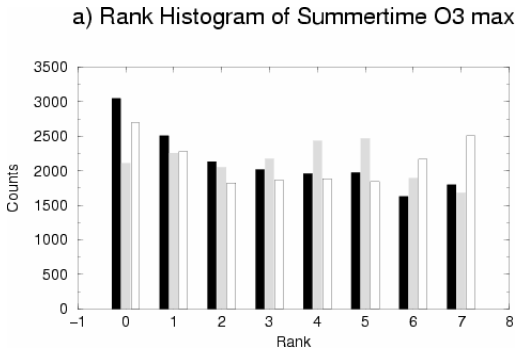

Figure 2: Rank histograms of observed summertime ozone daily maxima among the 7 simulated values, all stations and days, from April to September 2001 being put together. The black bars show the rank histogram from the raw ensemble. Gray bars stand for the histogram of unbiased estimates, the ensemble bias being removed. Empty bars show histograms for simulated values where bias has been removed separately for each model and each station. After Vautard et al. (2006).

5. CONCLUSION

The cooperative modelling work that was done within the CAFE projects has brought evidence that the use of model ensembles provides additional crucial information than the use of single models. However this demonstration is only done using past air quality data and cannot be directly extrapolated to future emission scenario simulations.

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SOURCE APPORTIONMENT OF PARTICULATE MATTER ($PM_{2.5}$) USING DISPERSION AND RECEPTOR MODELLING - A CASE STUDY FOR OSLO

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ABSTRACT

As a part of the EU-funded project Air4EU, a source apportionment study has been performed on PM data from a traffic hot-spot monitoring station in Oslo. In this study, the quality of the emission inventory for particulates ($PM_{2.5}$) was assessed. This assessment was performed by comparing source contribution estimates from a dispersion model, using the official emission inventory, with source apportionment estimates from a receptor model. The chemical composition of $PM_{2.5}$ filter samples from a measurement campaign performed from January 2004 to April 2005 was analysed. With these data as input, Positive Matrix Factorization (PMF) was applied to detect and quantify the various source contributions. For the same observational period and site, we performed emission and dispersion model calculations using the Air Quality Management system AirQUIS. Based on comparison of the two methodologies, the emission inventory was adjusted for individual source categories and the $PM_{2.5}$ concentrations were recalculated. These updated $PM_{2.5}$ concentrations were compared with measurements at three other independent stations to evaluate the improvements of the updated inventory.

1. INTRODUCTION

Air pollution emission inventories are in many cases the basis for air quality assessment and management. The quality of the inventories is of great importance since these data are part of the fundamental basis for air pollution impact assessments using dispersion models.

According to the EU Air Quality Directives (EC, 1999) national and local authorities are required to assess the air quality in zones, throughout the territory of the Member State. To obtain a territorial coverage, it is necessary to use dispersion models, especially in areas where the limit values are approached or exceeded. Also, source contributions must be assessed if limit values and guidelines are exceeded, since abatement measures are required in that case. For this reason it is not only important to calculate the concentration levels properly, but also to quantify the most important source contributions in order to implement effective abatement measures.

As part of the EU-funded project Air4EU, a source apportionment study has been performed on $PM_{2.5}$ data from a traffic hot-spot monitoring station in Oslo (Laupsa et al., 2007). In this study, the quality of the applied emission inventory for $PM_{2.5}$ was assessed. This assessment was performed by comparing source apportionment estimates from a dispersion model, using the official emission inventory, with source apportionment estimates from a receptor model (PMF). Based on this analysis, individual source categories within the emission inventory were adjusted and $PM_{2.5}$ concentrations were re-calculated to reveal improvements in the results.

2. METHODOLOGY

Measurements of $PM_{2.5}$

In 2004, the air quality monitoring network in Oslo consisted of 10 monitoring stations, five street stations and five urban background stations. At five of these stations $PM_{2.5}$ was measured using TEOM instruments (Lützenkirchen and Lutnæs, 2004).

In parallel and for two different periods, from January 2004 to April 2004 and from October 2004 to April 2005, a PM measurement campaign was carried out at the same site as one of the street stations, identified as Rv4. 12 hour filter samples of $PM_{2.5}$ were collected using a Kleinfiltergerat (KFG) instrument (Hagen et al., 2005).

Measurements of hourly averages of $PM_{2.5}$ at four of the stations, Kirkeveien, Løren, Aker hospital and Rv4, were used to evaluate the results from the dispersion model and the impact of adjusting the emission inventory. Kirkeveien, Løren and Rv4 are traffic stations and Aker hospital is an urban background station.

Daily values of $PM_{2.5}$ from a regional background site, Birkenes, were used as boundary conditions in the dispersion model calculations. Birkenes is located approximately 300 km south-west of Oslo (58º 23'N, 8º 15'E).

Receptor modelling using Positive Matrix Factorization (PMF)
78 $PM_{2.5}$ filter samples were selected for chemical analysis (Hagen et al., 2005). The components and elements analysed were selected to provide as much information as possible for the source identification. The filter samples were analysed with respect to major anions and cations using ion chromatography, and 30 elements using ICP-MS (NILU, 2006). Thermal Optical analysis (TOT) was applied to quantify the samples' content of elemental (EC) and organic carbon (OC). (Birch and Cary, 1996), whereas levoglucosan (Dye et al., 2005; Yttri et al., 2005) and NCBA where quantified using HPLC/HRMS.

Based on the chemical analysis of the $PM_{2.5}$ samples, receptor modelling was performed to detect and quantify the various source contributions. Positive Matrix Factorization (PMF) (Paatero, 1993 and Paatero and Tapper, 1993, 1994) was used in the present study. The PMF model uses both measured concentrations and uncertainty estimates to generate chemical profiles and time series associated with each profile. The method applied in this study is two dimensional PMF (PMF-2).

Dispersion modelling
The AirQUIS modelling system (AirQUIS, 2006) was applied in this study to calculate ambient concentrations of $PM_{2.5}$. AirQUIS is a GIS based integrated management system that includes a user interface, comprehensive measurement and emission inventory databases, and a suite of models for simulating ambient air concentrations and exposure.

The models used in the calculations are the diagnostic wind field model MATHEW (Sherman, 1978; Foster et al., 1995) and the dispersion model EPISODE (Slørdal et al., 2003). The EPISODE model contains a Eulerian model with embedded sub-grid line source and point source models for calculating ambient concentrations. The line source model HIWAY-2 (Petersen, 1980) is used to calculate traffic related contributions at receptor points close to roads. The grid applied for the Oslo region is a 22x18 km grid, with grid size 1 km, and 10 vertical layers ranging up to 2400 m. The meteorological wind field is calculated using input from a meteorological mast located within the modelling domain.

Concentrations calculated at receptor points corresponding to the positions of monitoring stations in Oslo are stored for comparison with the monitoring data.

The emission inventory for industrial and area distributed sources in Oslo has been compiled by Statistics Norway. The traffic emission inventory is based on official data from The Norwegian Public Roads Administration.

To carry out the source apportionment using the dispersion model, calculations of hourly concentrations from single source categories, or a suite of categories, were performed from 1 November 2003 to 1 May 2004. The classification of the sources was made according to the sources indicated by the receptor modelling. 12 hour averages were generated for the periods corresponding to the data collection period for the filter samples.

3. RESULTS AND DISCUSSION

Source apportionment for 40 filters collected from January 2004 to April 2004 provide the basis for the comparison between the receptor model and the dispersion model results. The PMF model calculations resulted in separate factors which were interpreted as the following $PM_{2.5}$ sources: 3 different traffic related sources (suspension of road dust, exhaust particles from gasoline and diesel vehicles separately), a long range transport (LRT) source, and a wood burning source for domestic heating. In addition to the sources identified by the PMF model, the emission inventory also included emissions from industrial activities, space heating using oil, and harbour and non-road traffic sources. In the results from the PMF model, these sources are probably incorporated with the sources interpreted as traffic exhaust.

The results identified differences between the dispersion and the receptor model results (Figure 1). Particularly for contributions from wood burning and traffic, there were significant differences. For domestic wood burning, the dispersion model estimated an average $PM_{2.5}$ concentration twice the concentration estimated by the receptor model. On the other hand, the receptor model estimated a contribution from traffic-induced suspension that was 7 times higher than the dispersion model estimate. For the other sources, i.e., long range transport and sum of vehicle exhaust and other combustion sources (sources in bars 4, 5, and 6 in Figure 1), the models agreed fairly well. In spite of the deviation for individual sources, the average $PM_{2.5}$ deviation between the dispersion model (20.9 $\mu g/m^3$) and filter samples (23.9 $\mu g/m^3$) was only 13%.

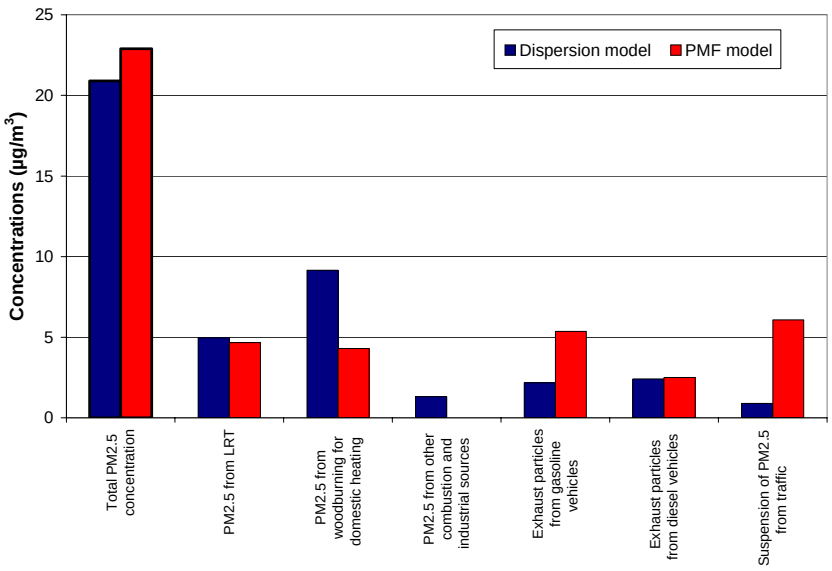

Figure 1: Estimated average concentrations of $PM_{2.5}$ (µg/m^3) from different source categories using the dispersion (blue) and receptor models (red) at Rv4 (traffic hot-spot station). The comparison was performed for the period where the 40 filter samples were collected, winter/spring 2004.

To assess if the differences between dispersion and receptor model calculations, found at the one site, were generally applicable throughout Oslo, we adjusted the emission inventory for individual source categories by a simple rescaling of the emission rates and recalculated $PM_{2.5}$ concentrations using the dispersion model. Based on the above results, the emission inventory was updated for the wood burning and traffic induced suspension. The emissions from wood burning were reduced by a factor of 2 and the emissions for traffic induced suspension were increased by a factor of 7. New $PM_{2.5}$ estimates from November 2003 to May 2004 were calculated using the dispersion model. The newly estimated $PM_{2.5}$ concentrations were compared with measurements at three other independent stations, Kirkeveien, Løren, Aker hospital, to evaluate the improvements of the updated inventory. New statistics were calculated for all the four stations. The statistical analysis showed a general improvement in the estimated concentrations for $PM_{2.5}$ at all sites (Table 1). The correlation coefficient (based on daily means) increased, RMSE (Root Mean Squared Error) decreased and the regression parameters (slope and intercept) generally improved after updating the emissions. The average value increased or decreased dependent on which sources dominates at the specific locations.

Table 1: Measured (hourly) and estimated (dispersion model) $PM_{2.5}$ concentrations from November 2003-April 2004 (based upon daily (24 hour) values) at 4 stations in Oslo before and after rescaling of the emission inventory.

µg/m^3	$PM_{2.5}$											
	Løren			Kirkeveien			Aker sykehus			RV4		
	Meas.	Estim.	Re-est.	Meas.	Estim.	Re-est.	Meas.	Estim.	Re-est.	Meas.	Estim.	Re-est.
Max	31.8	61.0	45.4	36.4	71.7	47.5	27.1	58.0	37.6	29.8	60.3	40.6
Ave.	15.3	14.2	14.5	13.6	14.1	11.8	10.9	11.8	10.2	13.0	14.3	14.2
St. Dev.	6.5	10.6	9.2	5.7	10.7	7.3	5.0	11.0	7.7	5.3	11.0	8.0
Corr. Coeff.		0.59	0.67		0.66	0.72		0.61	0.67		0.55	0.65
Slope		0.96	0.94		1.24	0.91		1.34	1.04		1.14	0.98
Intercept		-0.56	0.06		-2.81	-0.59		-2.8	-1.08		-0.62	1.45
RMSE		8.62	6.84		8.15	5.39		8.82	5.74		9.25	6.16

The new estimated average concentration map showed a reduction in the spatial concentration values of $PM_{2.5}$ (Figure 2) This is due to the reduction of the source strength of wood burning for domestic heating, which is an area distributed source. The source strength for traffic induced suspension is however a local hot spot source and will therefore have less influence on the spatially distributed concentrations.

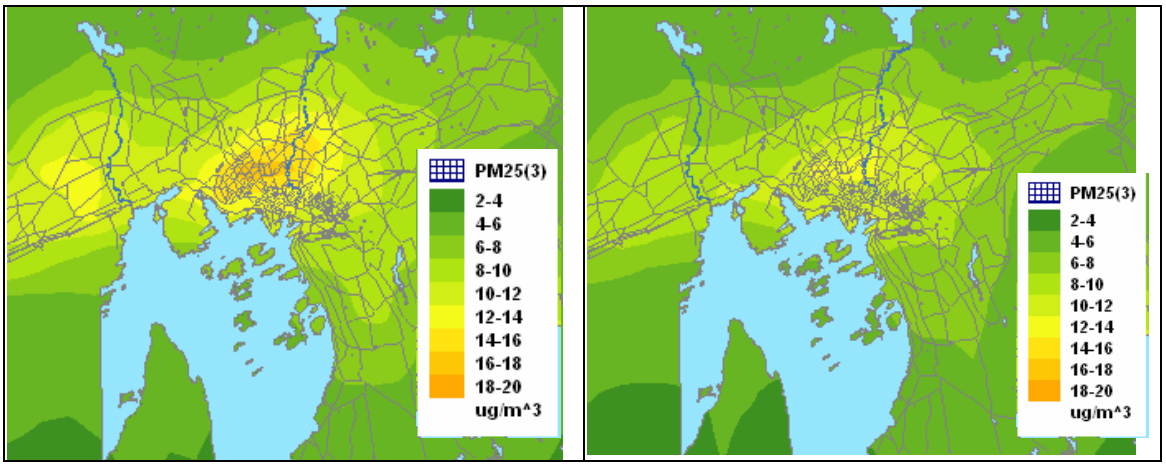

Figure 2: a) Average $PM_{2.5}$ concentration field from 1 November 2003 to 1 May 2004. b) New estimate of average $PM_{2.5}$ concentration field after rescaling of the emission inventory.

4. CONCLUSIONS

Comparison of source apportionment estimates of $PM_{2.5}$ using receptor modelling and dispersion modelling, provides a good basis for independent assessments of emission inventories. Even though both dispersion and receptor modelling have weaknesses, comparison of source apportionment estimates can reveal both differences and conformities and is a good basis for a detailed analysis of the results. Although the measured and modelled $PM_{2.5}$ concentrations on the average are in good agreement at all sites in Oslo, the analysis has shown large deviations for individual sources. Largest deviations were revealed for wood burning and traffic induced suspension. This study has shown how updated emission estimates for single sources may have significant impact on the spatial concentration distribution estimates and the associated exposure estimates and abatement measures.

5. ACKNOWLEDGEMENTS

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ASSESSMENT OF NON-EXHAUST PM EMISSIONS BY ROAD TRAFFIC IN URBAN AREAS – AN AIR4EU CASE STUDY IN ROME, LONDON, OSLO AND ROTTERDAM

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ABSTRACT

Traffic emits both exhaust and non-exhaust particulate matter (PM). The latter involves non-controlled friction processes (e.g. brake lining and tire wear) and re-suspension of road dust. In cooperation with the cities of Rome, London, Oslo and Rotterdam in Air4EU, a method has been demonstrated to assess the contribution of non-exhaust PM emissions by road traffic. The method applies hourly monitoring data of NO_x and PM collected both at an urban background location and an inner-urban road. The study shows that the contribution of non-exhaust emissions of traffic is of the same order as exhaust emissions and mainly consists of coarse $PM_{2.5-10}$ particles. In Rome the contribution of non-exhaust is significantly higher than exhaust emissions. This may be related to drier meteorological conditions in the south of Europe. In Nordic countries, due to the use of studded tyres and salting in early spring, non-exhaust contribution of PM exceeds an order of magnitude average PM exhaust emissions.

1. INTRODUCTION

PM_{10} air quality limit values are frequently exceeded in streets in urban areas. This underlines, that traffic is an important local source for PM_{10} (and $PM_{2.5}$) emissions. Monitoring is hardly feasible to assess air quality in all relevant streets and therefore, monitoring is often combined with (street canyon) modelling. These models require emission factors for PM, as input parameter. Traffic emits both exhaust and non-exhaust PM. The former is relatively well defined in emission inventories, e.g. [COPERTIII, 2000] and [UBA, 2004] but the latter involves non-controlled friction processes (e.g. brake lining and tire wear) and re-suspension of road dust. Non-exhaust emissions depend on traffic intensity, road type and meteorology, which vary in different cities in Europe. In the framework of the Air4EU project (www.air4eu.nl), a case study was implemented to assess non-exhaust PM emissions by road traffic [Air4EU, 2006].

2. METHODOLOGY

In co-operation with the cities of Rome, London, Oslo and Rotterdam, a method has been demonstrated to estimate the non-exhaust contribution of PM by road traffic in urban areas. The method was developed in European Topic Centre Air Quality and Climate Change project Street Emission Ceilings (SEC) [EEA, 2004]. Monitoring data for NO_x and PM_{10} (and $PM_{2.5}$ in London and Oslo) collected at an urban background station and a street station are used for each city. Also, traffic data (e.g. number of vehicles, average speed and fleet composition) in the street with air quality data was available. For London, Rotterdam and Rome this concerned annual data, while for Oslo only winter data were applied. Increments (i.e. differences between street and urban background concentrations) of PM and NO_x, and their ratio are calculated using hourly monitoring data. Subsequently, the increment ratio of *measured* PM/NO_x is compared with the ratio of *emitted* PM/NO_x. From the difference between measured and emitted PM/NO_x ratios the non-exhaust contribution of PM can be estimated.

3. RESULTS AND DISCUSSION

As an example, increments for NO_x and PM_{10} concentrations at the Marylebone Road in London are presented in Figure 1. These graphs illustrate the impact of morning and evening traffic peaks on NO_x and PM_{10} concentrations, especially for working days. The example for London indicates that lower traffic volume and lower percentage of heavy duty traffic in the weekend results in lower contributions to NO_x and PM_{10} in Marylebone Road.

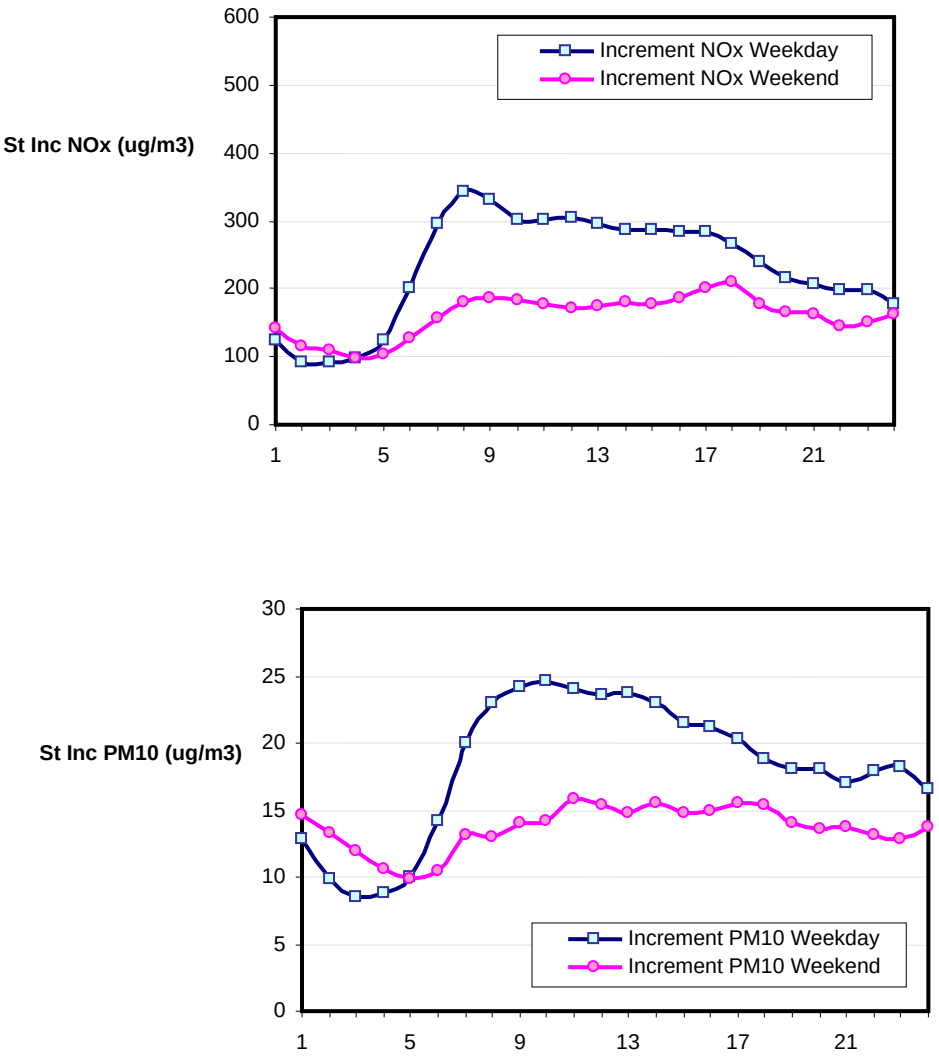

Figure 1: Annual diurnal average of hourly street increments in Marylebone Road (London) in 2004 for NOx and PM_{10} during working and weekend days.

Differences in increments in the four cities are related to traffic volume (highest in London), fleet composition (highest fraction of heavy duty in Rome), dispersion of air pollution (highest around the highway in Oslo), meteorology (wind speed and rainfall) and monitoring period (only winter period in Oslo, while in the other cities an annual period).

The next step is to compute the ratio of the increments for NO_x and PM_{10}, as well as for NO_x and $PM_{2.5}$ (only in London and Oslo these data are available). In Figure 2 the results are presented for the ratio of the increments NO_x/PM_{10} in London.

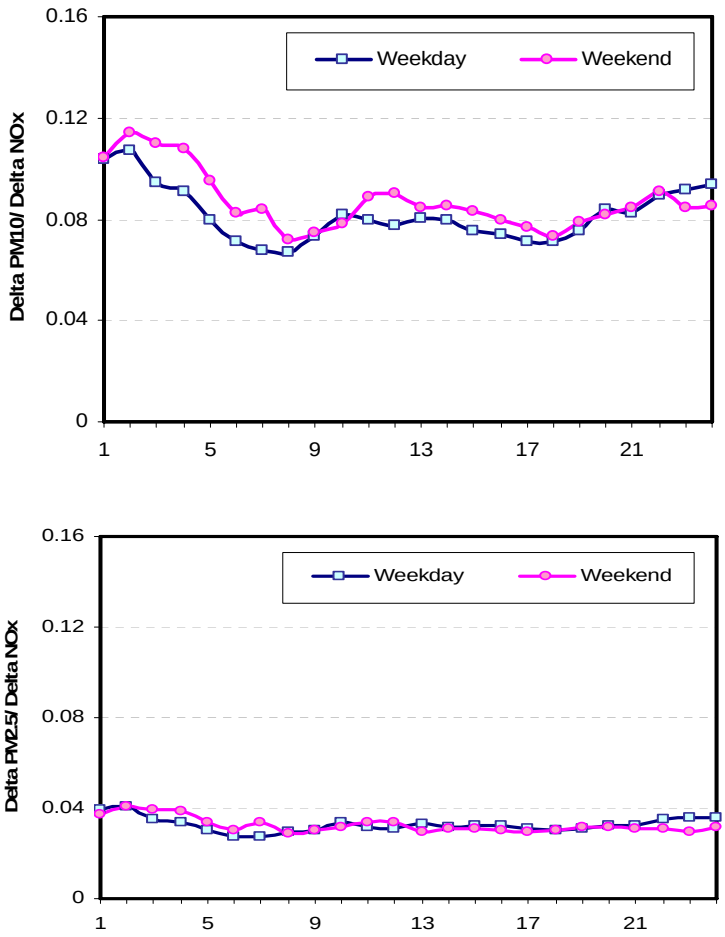

Figure 2: Annual diurnal average of hourly increments in Marylebone Road (London) in 2004 for PM_{10}/NO_x and $PM_{2.5}/NO_x$ during working and weekend days.

The ratio of the increments of PM_{10}/NO_x in Figure 2 indicate that the ratio PM_{10}/NO_x emitted by local traffic varies between 0.06 – 0.12 in London for both weekend and working days. The ratio of the increments for $PM_{2.5}/NO_x$ are in the range of 0.03 – 0.04 and in the order of 50% of the PM_{10}/NO_x ratio. As exhaust emissions of PM are mainly $PM_{2.5}$, the larger ratios of PM_{10}/NO_x as compared to $PM_{2.5}/NO_x$ indicate a *non-exhaust source of PM_{10} emissions*. This is further discussed in the next section on increments of emitted PM/NO_x. In Table 1 the results are presented for the study in Rome, London, Oslo (winter period) and Rotterdam.

Table 1: The ratio of increments PM/NO_x of monitoring data and exhaust pipe emission factors in Magna Grecia (Rome; 2003), Marylebone Road (London; 2004); highway (Oslo; winter 2005) and Bentinckplein (Rotterdam; 2005).

	Measured ratio PM_{10}/NO_x		Measured ratio $PM_{2.5}/NO_x$		Emitted ratio PM*/NO_x
	Weekend	Working	Weekend	Working	
Rome	0.15	0.17	-	-	0.05
London	0.08	0.08	0.04	0.04	0.05
Oslo (winter)	0.40	0.30	0.04	0.04	0.05
Rotterdam	0.05	0.12	-	-	0.05

*: Exhaust emissions are measured as PM but actually only concern $PM_{2.5}$.

Table 1 illustrates that the *measured* ratio PM_{10}/NO_x is in the order of a factor 2 (Rotterdam and London) and a factor 3 (Rome) larger than the *emitted* ratio PM/NO_x. This indicates an additional source of PM_{10} than exhaust emissions such as re-suspension of road dust and friction processes. In Oslo even a factor 10 is encountered but Oslo is a special case due to the use of studded tyres in Nordic countries in the winter period. The lower measured ratio PM_{10}/NO_x during the weekend in Rotterdam shows the importance of heavy duty traffic (trucks and buses) for non-exhaust emissions, as in the weekend in Rotterdam the number of heavy duty trucks is practically zero and the number of buses is reduced.

Table 1 also shows that the measured ratio $PM_{2.5}/NO_x$ is in the same order as the ratio of emitted PM/NO_x of urban traffic. This illustrates that exhaust emissions are mainly $PM_{2.5}$ and hardly any $PM_{2.5-10}$. It also shows that non-exhaust emissions is mainly in the size range of $PM_{2.5-10}$.

4. CONCLUSION

The case study in Rome, London, Oslo and Rotterdam on the assessment of non-exhaust emissions of $PM_{10/2.5}$ demonstrate the applicability of the methodology earlier developed in the Street Emission Ceiling project. Earlier findings were confirmed, that non-exhaust emissions mainly consist of $PM_{2.5-10}$, while exhaust emissions are $PM_{2.5}$. Presently, non-exhaust emissions are in the same order as exhaust emissions in urban areas with the exception of Nordic countries where studded tyres in early spring result in additional high PM_{10} emissions. The importance to monitor both PM_{10} and $PM_{2.5}$ have been shown, as exhaust and non-exhaust emissions can be best differentiated by $PM_{2.5}$ (exhaust) and PM_{10} (non-exhaust). It is recommended to perform more research on factors controlling re-suspension of road dust such as traffic volume, traffic speed, fleet composition, road type, dispersion and meteorological conditions.

5. ACKNOWLEDGEMENT

The Air4EU project was financed under the FP6 programme of the European Commission. The cities of Rome, London, Oslo and Rotterdam are acknowledged for providing the necessary monitoring data to perform the case study.

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STUDY OF THE PAST, PRESENT AND FUTURE STATE OF AIR QUALITY IN THE RUHR AREA

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ABSTRACT

Emission reduction scenarios have been applied to study the possible future development of air quality in Europe in general and in the Ruhr Area in particular. The EURAD model system allowing nested calculations with high resolution has been employed. Annual simulations were carried out for 2002, 2005 and 2010 to demonstrate the sensitivity of air quality in the boundary layer to expected emission changes. Considerable improvements are predicted for the Ruhr region though certain problem areas will remain regarding the requirements of the EC air quality directives for particulate matter and NO_2 in particular.

1. INTRODUCTION

The Ruhr Area of the State North-Rhine Westphalia (NRW) is a densely populated region with 11 cities and 4 counties. It encompasses an area of 4.435 km² with a population of 5.3 million. It belongs to the most congested areas in Europe together with Ile de France, Moscow, Greater London, the Po Valley and Istanbul, and shows typical features of a mega city. It contains the main centre of European integrated steel production. Due to severe economical changes it has gradually been converted from a “black country” into a cleaner region. The proverbial air and water pollution of the area is largely a thing to the past. Yet it is still characterized by elevated emissions of anthropogenic pollutants. Intense industrialisation started in the 19th century leading to strong increase of the number of coal mines (from more than 300 in 1850 to 3200 altogether). Coking plants and steel mills started to develop with an impressive growth rate. At the same time the population increased exponentially. After the coal crisis started in 1958 the number of coal mines and coking plants was drastically reduced. Only 6 coal mines and 3 coking plants are still operating and the future of the mines is presently in strong political dispute.

This shows that the Ruhr Area is in a transformation process from a region completely depending on mining and heavy industry to more complex economical conditions. The major old industries left enormous buildings and open spaces behind which are now converted into areas to be used by modern industry, business, leisure industry and tourism. The change from totally grey to more and more green of the former black country is clearly evident from the improvement of air quality (example of SO_2, also including the Rhine area in NRW, Fig. 1). It is emphasized that a considerable part of the improvement is due to regulation and control measures in Europe in general and particularly to those imposed by the German federal and NRW state government as indicated in Fig. 1 (TAL). The conversion of the Ruhr Area from a heavily polluted to a cleaner environment forms a grand experiment of transformation of a mega city from adverse to acceptable conditions and may provide useful material for the treatment of environmental problems of other mega cities. Measurements show that the region is on a promising way to acceptable atmospheric environmental conditions, but that the standards set by the European Commission (EC 1999) still needs further mitigation measures leading to emission reductions. The aim of this study is to demonstrate what changes can be expected in the Ruhr area if available reduction estimates will be realized in the near future.

2. THE MODEL

The trend of air quality to be expected from estimated future emission reductions has been calculated with the EURAD model system. A description of the model is given in Memmesheimer et al. (2004). It allows nesting of selected areas (Jakobs et al. 1995) with increasing horizontal resolution up to 1 km. In this study the procedure was applied to the State of NRW with 5 km grid size proceeding from the European domain (125 km grid size, coarse grid calculations) to Central Europe (25 km, nest 1) to NRW (nest 2). The RACM-MIM chemical mechanism was employed (Geiger et al. 2003, Karl et al. 2006). Secondary anthropogenic and natural inorganic and organic aerosols are simulated with the modules MADE (Ackermann et al. 1998) and SORGAM (Schell et al. 2001) including interactions with clouds. Hourly grid box values of emission intensities needed for the chemistry transport model of the EURAD system are derived from annual emission inventories employing the EURAD Emission Model (EEM). The model has been evaluated in a large number of applications to demonstrate its applicability to air quality studies like the one discussed in this paper.

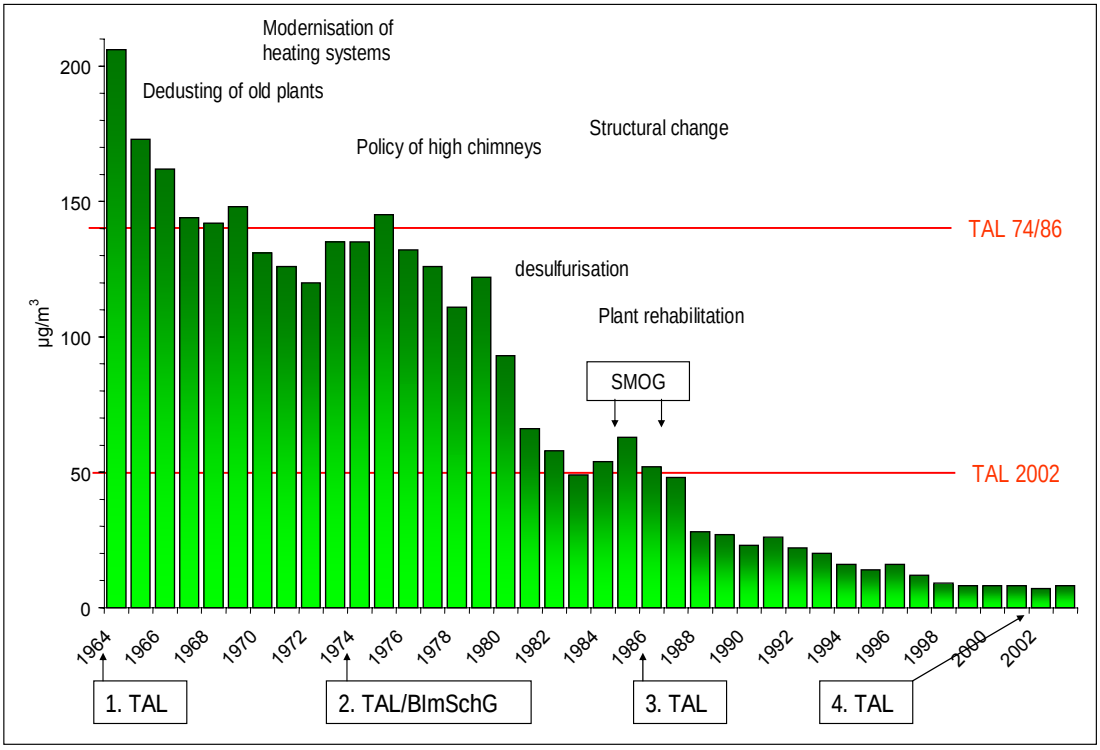

Figure 1. Time evolution of the annual average SO_2 load in the Rhine-Ruhr-Area 1964 – 2003. With indication of applied mitigation measures. TAL: Federal air quality directives.

Table 1. Annual emissions, nest 2 (mostly NRW), kto/year

Source category	**NO_x**				**VOC**			
	2000	**2002**	**2005**	**2010**	2000	**2002**	**2005**	**2010**
road transportation	4,6	4,0	3,1	1,8	90,3	74,6	51,0	24,6
point sources	151,2	151,2	151,2	127,8	7,1	7,1	7,1	6,6
small scale sources	19,9	19,7	19,4	18,9	146,2	135,5	119,4	98,0
Total		**174,9**	**173,7**	**148,5**		**217,2**	**177,6**	**129,1**

Source category	**Particles**				**Soot**			
	2000	**2002**	**2005**	**2010**	2000	**2002**	**2005**	**2010**
road transportation	6,5	5,5	3,9	1,9	6,1	4,9	3,0	1,0
point sources	33,2	33,2	33,2	30,2	20,0	20,0	20,0	18,0
small scale sources	2,2	2,1	1,8	1,4	1,8	1,7	1,6	1,3
Total		**40,7**	**39,0**	**33,5**		**26,6**	**24,6**	**20,3**

3. EMISSION REDUCTION ESTIMATES

The EMEP inventory of annual emissions has been applied to estimate hourly emission intensities for the reference year of the simulations, namely 2002, on the greater and Central European scale, whereas specific inventories provided by the former Environmental Agency of the State NRW (LUA) have been employed for nest-2 calculations. The reduction scenario is based on estimates of IIASA, EMEP, the German Federal Environmental Agency (UBA) and LUA. Examples are presented in Table 1 for NOx, VOC, particulate matter and soot as derived for nest 2 (mostly NRW).

4. RESULTS AND DISCUSSION

Using estimated emission intensities of the years 2002, 2005 and 2010 (Table 1 for NRW) together with the meteorology of 2002 the sensitivity of air quality to changing emissions has been analyzed (Memmesheimer et al. 2004a,b). Since gradual reduction of emitted pollutants is expected for West Europe

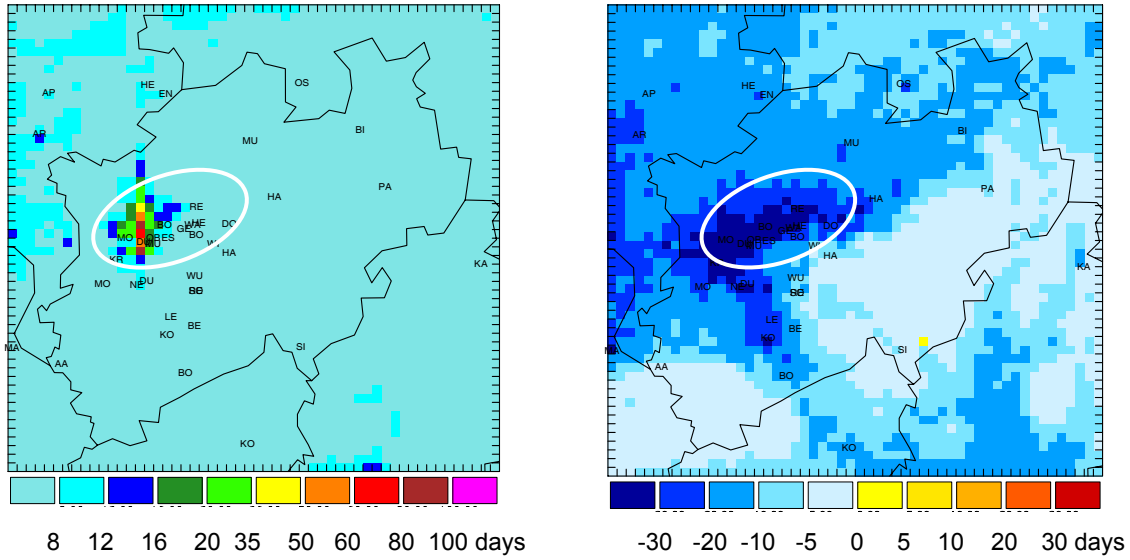

Figure 2. PM_{10} in 2010. Left panel: Number of days/year with PM_{10} exceeding 50 µg/m³. Right panel: Difference (2010 - 2002) of the number of days/year with PM_{10} exceeding 50 µg/m³. EC limit values for PM_{10} are 40 µg/m³ for the annual average concentration and 35 exceedance days/year from 2005 on. NRW/nest 2. The Ruhr Area is approximately indicated by ellipses.

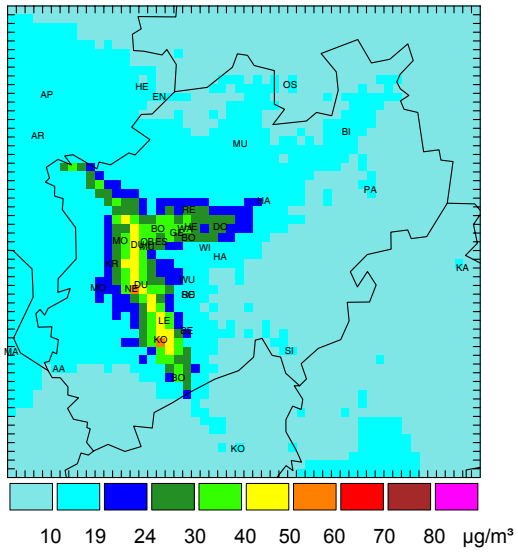

Figure 3. Annual average of NO_x mixing ratios (µg/m³), NRW/nest 2, 2010.

the simulations reveal a decrease of primary and most secondary pollutant concentrations. The analysis was carried out for SO_2, CO, NO, NO_2, NO_x, PM_{10}, NH_3, HNO_3, NH_4^+, SO_4^{2-}, NO_3^- and ozone. $PM_{2.5}$ was also considered in some instances. Ozone shows a peculiar behaviour with respect to reduced emissions of its precursors. In NRW/nest 2 one finds that the annual mixing ratio average increases in most cases since higher NO emissions keep it at lower levels through titration. This effect is stronger in the Ruhr Area and also along the Rhine than in neighbouring regions. In contrast, maximum hourly values decrease at most places pointing to a reduction of strong photo-oxidant episodes as a consequence of decreasing mixing ratios of the precursors NO_x and VOC

As regards air quality in general the Ruhr Area will considerably gain from expected emission reduction. This is clearly evident from the simulated PM_{10} response. The right panel of Fig. 2 exhibits the difference of the years 2010 and 2002 obtained for the estimated number of days/year with hourly values of PM_{10} exceeding the EC limit of 50 µg/m³. The most efficient reduction of days with high aerosol load is obtained along the Ruhr. Regarding the number of critical aerosol days (left panel) there seems to be a good chance to meet the EC standard (EC 1999) in most of the nest 2 domain in 2010 except for a region in the Ruhr area where strong industrial aerosol sources exist. Street canyons with high traffic amounts are potential further neuralgic spots regarding atmospheric PM_{10} and NO_2 loads. It is noted that secondary aerosol formation is a large-scale phenomenon and therefore not only local, but also Europe-wide reduction of the main aerosol precursors, namely ammonia, sulphur dioxide and nitrogen oxides, is needed to reduce background aerosol formation so efficiently as seen in the Ruhr Area.

The anticipated emission reductions are sufficient to reduce annual averages of NO_x mixing ratios below the limit values enforced by the EC in most parts of NRW (NO_2: 40 µg/m³ for the protection of human

health in 2010; NO_x: 30 μg/m³ for the protection of vegetation, valid since 2001). Though this means considerable improvement of air quality the values of NO_x will continue to be exceeded in the Ruhr Area and along the Rhine in NRW, but not in rural and remote areas as indicated in Fig. 3. It should be noted that in street canyons which are not resolved by the model the limit value of NO_2 for the protection of human health may considerably more be exceeded than indicated in the figure.

5. CONCLUSIONS

The Ruhr Area exhibiting features of a mega city is presently still suffering from mainly anthropogenic air pollutants considerably exceeding EC air quality limit values, though the concentrations of some major pollutants have strongly been reduced during the last decades. Simulations of air quality with the EURAD model system were carried out to assess future changes in the Ruhr Area, in particular, resulting from the expected reduction of the emission of anthropogenic pollutants in Europe. It can be shown that the Ruhr Area gains a lot from the anticipated mitigation measures regarding overall air quality. Yet additional control is needed for some pollutants which have not met the limit values of the EC air quality directives in 2005 (e.g., PM_{10}) and probably also not quite meet those in 2010 (e.g., PM_{10}, NO_2) according to our reduction sensitivity studies. This may happen for ozone because the calculated AOT40 values could still be slightly to high, for NO_2 at traffic hotspots, and for particulates in the vicinity of industrial sources and in street canyons. Further work is needed to explore possible trends of the finest fraction of aerosols below 2.5 μm down to nanometers because of their special relevance for health effects.

6. ACKNOWLEGDEMENTS

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A WEB BASED MAPPING TOOL FOR THE HARMONISED INTERCOMPARISON OF AIR QUALITY MAPS

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ABSTRACT

The Air4EU mapping tool is a web based GIS mapping tool which was developed as part of the EU 6th framework project Air4EU. The mapping tool allows maps on all scales to be displayed, along side each other if required, using a homogenous colour scaling and map projection that allows easy intercomparison of maps from different regions, of maps on different scales and of maps produced by different methodologies. It contains a number of examples of different methodologies such as modelling, interpolation of monitoring data and various data assimilation methods. A large number of the maps are also accompanied by uncertainty maps that have been produced based on recommendations from the Air4EU project. These uncertainty maps allow the viewer to directly see the comparative quality of the maps displayed. The basic technology used for this system and its application in Europe wide air quality assessment are discussed in this paper.

1. INTRODUCTION

The Air4EU project has produced valuable results and useful recommendations for improving air quality assessment in Europe (www.air4eu.nl). In order to facilitate the widespread access of these results a web based GIS tool was developed by the Norwegian Institute for Air Research (NILU). This system allows maps of air quality to be shown, in a friendly and coordinated manner, for a wide variety of pollutants and all spatial scales using an interactive GIS system. The maps available include case studies carried out within Air4EU as well as other contributed assessment maps. Figure 1 shows the home page layout. The GIS based mapping tool web page can be visited at the following web address: www.air4eumaps.info

Figure 1 – Left: Home page of the mapping tool www.air4eumaps.info. Right: Showing the 'mapping room' selection tool where multiple maps can be selected based on criteria defined by the user.

2. OVERVIEW OF THE AIR4EU MAPPING TOOL

Web page set up and technical platform

The tool's technical platform is based on a web map server (WMS) and the associated GIS software MAPPER both developed by Demis (www.demis.nl). As shown in figure 2, the interactive GIS window within the web browser, is based on a flash application. In order to update a map, the flash application sends a request to the WMS using the OpenGIS WMS protocol developed by the OpenGIS consortium (www.opengeospatial.org). The WMS is a server where all GIS maps are registered. In order to display the appropriate map, the request to the WMS is filtered using the application server. The application server contains an application, written in ColdFusion, that defines the layer and information is to be shown. The application server is connected to a SQL Oracle database where information describing each map is stored. Once the request is filtered the map server sends a map image as a response to the flash application request.

The GIS data displayed are of 2 types. The first is the air quality data itself, which are stored as either ASCII grid or BIL file formats, and the second is the layer information pertaining to geo-referenced data such as roads, water, parks etc., in the form of standard ESRI shape files.

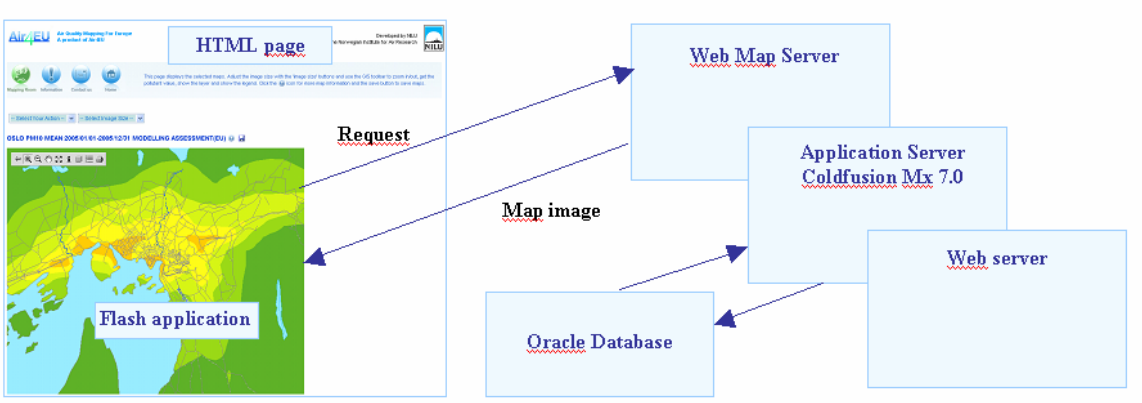

Figure 2 – Air4EU mapping tool dataflow. See the text for details.

Map properties and selection

Figure 1 (right) shows the map selection tool layout. The selection tool allows the database to be searched for all available maps using six map properties. These are region, pollutant name, indicator, period, mapping method and map type. Selection of any property will automatically update the list of available maps as well as repopulating the available property selection. The map properties are defined as follows:

- **Region:** The name of the geographical area covered by the map. It can be a city, a country or a specific region such as Oslo, England, Europe, etc. The tool is flexible and new regions can be added.
- **Pollutant name:** The name of the chemical pollutant addressed. Currently available pollutants are Benzene, CO, NO_2, O_3, PM_{10}, $PM_{2.5}$ and SO_2 but this is dependent on the actual maps available. The tool is flexible and additional pollutants can be added.
- **Indicator:** The statistical parameter, related to the pollutant, shown in the map. This can be any indicator, such as annual mean, number of exceedances, percentiles, AOT, etc. The indicator is usually related to limit values for particular pollutants.
- **Period:** This simply indicates the time period for which the map is valid, usually a year for assessment purposes.
- **Mapping method:** This indicates the methodology used for creating the maps. Currently available are: monitoring, modelling and assimilation as named methods. See figure 3 for an example. New methods can be added.
- **Map type:** There are currently two map types available, assessment and uncertainty. However, the mapping tool is flexible and there is no limitation to the number of map types.

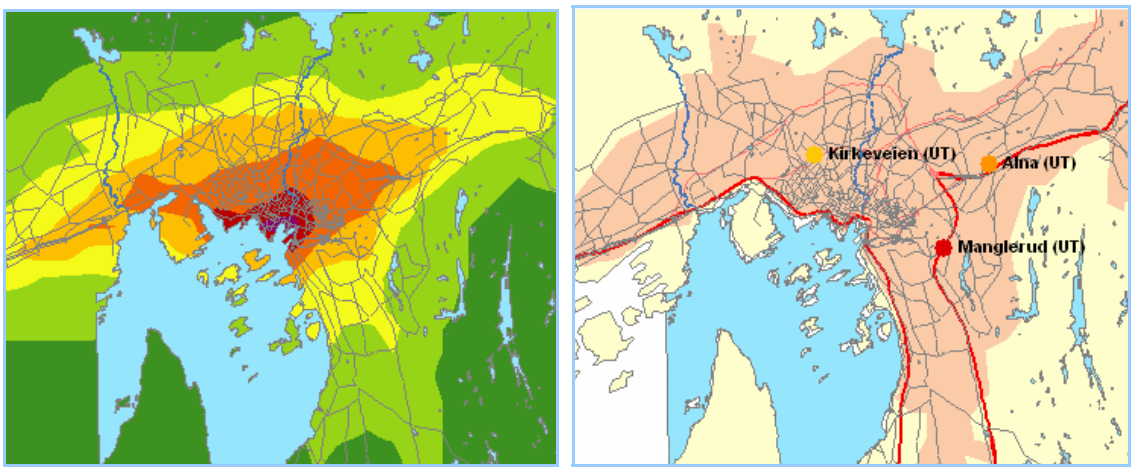

Figure 3 – Example of mapping method, modelling (left) and monitoring (right)

In addition to the maps, information for each map is available as a pop-up window. The information displayed includes text describing the map properties and any other relevant information concerning the model, the monitoring data, the methods used to construct the map or URL references to relevant links.

After filtering of the maps using the map properties the user can select any number of maps to be displayed. These will then be shown in a number of interactive GIS windows.

Map display

One of the main added values of this system is that it can display multiple interactive GIS windows at the same time, as shown in figure 4. This allows the user to view and directly compare assessment maps from different regions or cities, assessment maps of different compounds, assessment maps on different scales, assessment maps for different periods, assessment maps produced using different methods, and uncertainty maps for different mapping methods and regions. Each GIS window is interactive because it allows the user to edit the maps by zooming, selecting and viewing layers, viewing the legend, and printing and saving the edited maps.

Figure 4 – Screen shots showing examples of single (left) and multiple (right) interactive GIS windows. In this screen shot PM_{10} concentrations for Europe, Oslo, Prague and Rotterdam are shown.

Harmonisation of maps

There are two types of legends (contour colour scaling), currently in use in the mapping tool. One for assessment maps and one for uncertainty maps. The legend is harmonised so that maps are directly comparable. Each map may have a different contour interval, dependent on the range of concentrations, but the colour scaling has uniquely associated values so that any particular colour indicates the same value in all maps. Within the legend the pollutant level given by the European directives is indicated in red. Examples of these colour contours are shown in figure 5.

Within the system the monitoring stations are classified according to the DG Environment guidance on exchange of information: http://ec.europa.eu/environment/air/pdf/guidancetoannexes97101ec.pdf. The most common station categories are: UB (Urban background), UT (Urban traffic), UI (Urban industry), SB (Suburban background), ST (Suburban traffic), RNC (Real near city) and RR (Rural regional). See figure 3 (right) for an example.

During the mapping tool development EU guidelines for dissemination of maps in Europe were taken into account. These concern in particular the choice of the spatial reference. According to the guideline all maps (for Europe) are projected using the European spatial reference grid ETRS 1989 LAEA. EU guideline documents are available at the following websites: www.eea.eu.int, http://eionet.eu.int/gis and http://eionet.eu.int/gis/docs/EEA_GISguide_v2.doc.

3. Example

In figure 5 an example, extracted from the mapping tool, is shown. It displays a comparison of two different mapping methods used for calculating the annual mean PM_{10} concentrations for Europe in 2003. The two methods shown are calculations from the Unified EMEP model and results from the data assimilation of the EMEP model using Bayesian assimilation. In addition to the concentration maps their associated uncertainty maps are also shown. In this example it is possible to compare the results of the 2 methods and the uncertainty related to each of the methods. In this way maps from different models, and regions as well, can be compared.

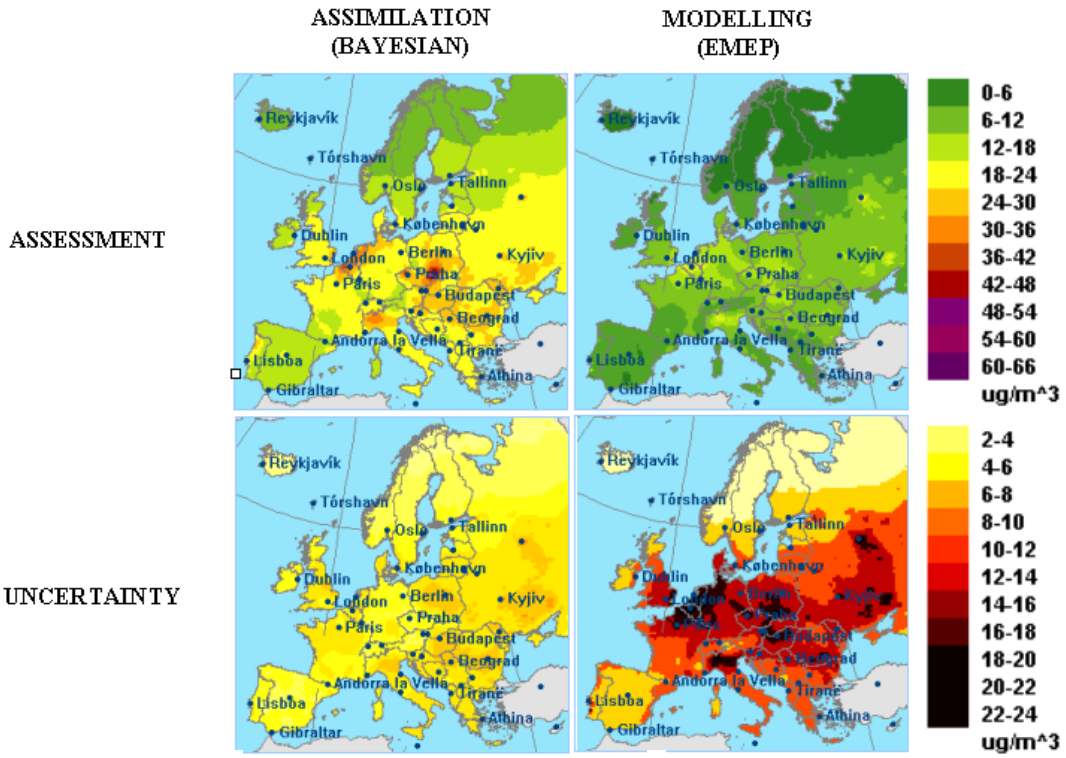

Figure 5 – Comparison of 2 different mapping methods showing annual mean PM_{10} concentrations for Europe in 2003. Bayesian assimilation assessment map (top left), EMEP model assessment map (top right), Bayesian assimilation uncertainty map (bottom left) and EMEP model uncertainty map (bottom right).

4. Conclusions

The tool improves and harmonises the dissemination of mapping information for both authorities and institutions involved in air quality assessment. It allows the simultaneous presentation, and comparison, of air quality assessment maps on all scales and for any number of compounds and indicators.

The features which make the mapping tool unique are as follows:

- The database of maps can be viewed and selected in a simple and interactive manner.
- Selection criteria for the maps include region, pollutant, assessment period and mapping method.
- Any number of maps can be simultaneously presented.
- Contour colour scaling (legend) is homogenised for direct intercomparison, with clearly defined limit values based on EU directives.
- Information and links for each map is provided.
- Maps and information can be saved or printed directly.
- Maps of uncertainty can also be viewed concurrently with the assessment maps.
- GIS integration allows for a more comprehensive and visual assessment.

The mapping tool is presented here to encourage the harmonised presentation of maps and encourage the production of uncertainty maps associated with them. The tool provides centralised and ordered access to a mapping database and can be used to aid dissemination of mapping information for both city users and institutes involved in air quality assessment.

If you wish to add maps or are interested in utilising the Air4EU mapping tool then please contact: Dr. Agnes Dudek at the following email address: agnes.dudek@nilu.no

www.air4eumaps.info

5. Acknowledgement

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SPATIAL REPRESENTATIVENESS OF AIR QUALITY MONITORING DATA AND THE RELEVANCE FOR MODEL VALIDATION

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ABSTRACT

A methodology is presented for the assessment of the spatial representativeness of air pollution monitoring data. The methodology relies on a statistical approach that connects air quality expectation values with land use characteristics. The relevance of this issue for model validation is addressed and the technique is illustrated for the validation of BelEUROS NO_2 model results.

1. INTRODUCTION

In a small country such as Belgium, air quality levels are sampled by a rather dense network of monitoring stations. More than 50 measuring stations are collecting pollutant concentrations (O_3, NO_2, PM_{10},…) on a half hourly basis. A great deal of these measuring stations is located in urban, suburban or industrial areas since it is there air pollution levels are higher and adverse health effects due to air pollution are more important. When these measurements are used for air quality assessment, an essential point that needs to be addressed is the spatial representativeness of the urban and industrial monitoring sites. This certainly holds for those urban or industrial sites that are used for model validation. For those locations it is questionable if point measurements can be directly compared to volume-averaged modelled concentrations.

In this paper a technique is presented which addresses this spatial representativeness of monitoring locations. Subsequently, the relevance of this issue for model validations is examined making use of NO_2 data obtained with the BelEUROS-model. The BelEUROS model is an Eulerian chemistry transport model (Deutsch *et al.*, 2007, Deutsch *et al.*, 2006) that is used as a policy supporting tool in Belgium.

2. METHODOLOGY

The methodology for the assessment of spatial representativeness presented here relies on a general relation between mean air pollution concentration levels and a parameter that determines the land use characteristics. Before the relation as such is discussed, the definition of the land use indicator will be briefly expounded.
The land use indicator, hereafter called the β-parameter, is calculated from the CORINE Land Cover data set. For a given area (~10 km²) the CORINE Land Cover pixels are determined and classified according to the 44 CORINE classes. The resulting classification histogram can be seen as a spectrum that represents a fingerprint of the land use characteristics in that particular area. As pointed out above, it is the aim to define a single value land use indicator that represents the characteristics of the local behaviour of the air pollution phenomenon. Therefore, the CORINE class distribution is transformed into a land use indicator according to the relation:

$$\beta = \log\left[1 + \frac{\sum_i a_i \cdot n_{CORINE\ Class\ i}}{\sum_i n_{CORINE\ Class\ i}}\right]$$

In this formula, the index i runs over all CORINE classes. $n_{CORINE\ Class\ i}$ is the number of pixels of class i inside the specified area and a_i is the pollution coefficient for the CORINE class i. As such, the β-parameter is the logarithm of a normalised sum of the CORINE class distribution in that particular region. The pollution coefficient a_i represents the impact of a particular CORINE class on the air pollution levels. Contributions from different urban, industrial, traffic and agricultural classes are expected to be important and are retained in this analysis. As a first approximation, the pollution coefficients of all natural CORINE classes are set to zero. For the 12 remaining classes, the set of pollution coefficients is optimized for each of the three pollutants in this study (O_3, NO_2, PM_{10}).

In order to pin down the relation between the mean air pollutant concentrations and the land use characteristics, the β-parameter is determined inside a 2 km buffer around each monitoring site. After all, it is only at those locations that such a relation can be established since measurement data has to be at hand. When the expectation values are plotted versus the β-parameter for all available monitoring sites, a scatter

plot as in Figure 1 is obtained. From this figure, a clear trend is revealed. Rural stations (low β) have low mean NO_2 concentrations, in urban or industrial sites (high β) increased NO_2 levels are observed. This relation between land use and air pollution can be approached by a second order polynomial fit. This functional form can then be used as an estimator for the pollutant concentration as a function of the land use indicator β. Similar relations are obtained for the pollutants O_3 and PM_{10} and they constitute the core of this methodology.

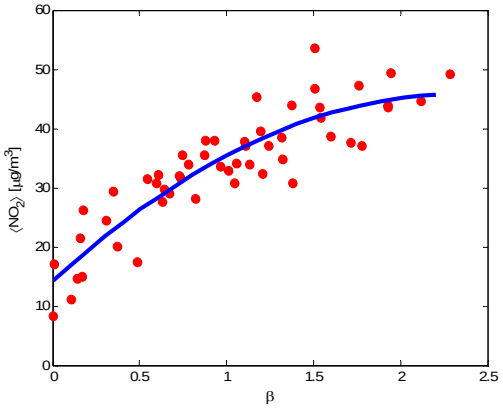

Figure 1: NO_2 expectation values at the Belgian monitoring locations as a function of this β-parameter. A second order polynomial fit is printed as the blue line.

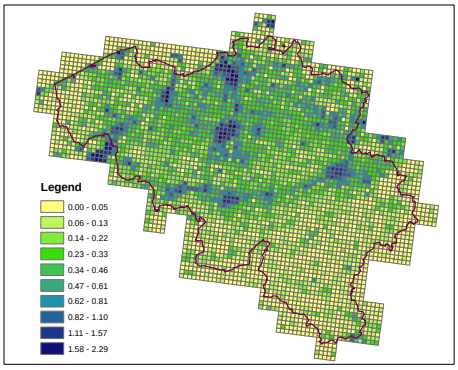

Figure 2: β-map for the NO_2 pollutant on a 3x3 km² regular grid. NO_2 monitoring stations are indicated by the red dots.

3. RESULTS AND DISCUSSION

3.1. Spatial representativeness of monitoring data

Up to now, the β-values were determined for the monitoring sites (2 km buffer) only. However, the β-values can also be calculated on a regular grid (here 3x3 km²) for the entire Belgian territory. An example of such a β-map is given in Figure 2. In this map, the urbanized and industrialized regions in Belgium are clearly pronounced due to their high β-values. Rural areas like the Ardenne region in the south of Belgium show lower β-values.

In order to assess the spatial representativeness of the monitoring locations, the variability of the β-values in the vicinity of the site is examined. Therefore, all grid cells of the β-map which fall inside a 7.5 km buffer of the station locations are collected and the variability of the β-values is determined. The results for the NO_2 stations are presented in Figure 3. For each station in this plot, the mean and standard deviation of the β-distribution is printed (blue dots with errors) in combination with the β-value of the corresponding monitoring site (green diamonds). As additional information, the minimum and maximum β-value in the buffer are also plotted (red triangles). This figure contains all relevant information for the assessment of the spatial representativeness of a monitoring site. First of all, it is interesting to explore the variability of the β-values in the buffer. This is expressed by the width of the error bars (one σ). If this value is large, a significant variability in the land use characteristics is noticed within a 7.5 km radius. As a consequence it can be expected that the air pollution concentrations in the same area are subject to a similar variability which clearly reduces the spatial representativeness of the monitoring site. Further, it can be examined if the mean β-value in the buffer (blue dot) approaches to the actual β-value of the monitoring site (green diamond). For a number of stations, both quantities differ to a large extent. In such cases, there is additional evidence for reduced spatial representativeness of the sampling site. After all, if the β-value of the site is far apart from the average land use characteristics in its surrounding area, its sampling values will by no means be representative for this same region. Similar plots can be obtained for the ozone and PM_{10} monitoring sites. Since the β-parameter represents a unique parameterisation for each pollutant, it has to be stressed that the spatial representativeness of a monitoring site also depends on the type of pollutant.

3.2. Model Validation

The methodology developed so far can also be applied to improve model validations. As a matter of fact, a great deal of the air quality sampling sites are located in (sub-)urbanised or industrialized regions. As

indicated before, it is questionable if the sampling values collected at those locations are representative for the volume averaged concentrations in the model grid cells.

In this paragraph, the methodology will be illustrated with NO_2 results of the BelEUROS model. The BelEUROS model is an Eulerian chemistry transport model that yields concentration fields on a 15x15 km² grid. In order to improve the comparison of model results with measurement values, the relation between air pollution and land use established in §2 is applied in a downscaling procedure. Based on this relation, a mass conserving distribution into 3x3 km² cells of the modelled pollutant concentrations is performed inside each 15x15 km² grid cell. The distribution is carried out according to the variability of the β-parameters of the parent BelEUROS grid cells. The result of this downscaling procedure of annual mean NO_2 concentrations is presented in Figure 4. By examining the two plots, it can be recognized that mass conservation of the BelEUROS results is fulfilled. When the high resolution map (3x3 km² cells) is aggregated again to the low resolution BelEUROS grid (15x15 km²), the original map is obtained again. On the other hand it becomes clear that especially in urbanized regions, much more detail is introduced in the 3x3 km² air pollution map which makes a comparison with measurement data more appropriate.

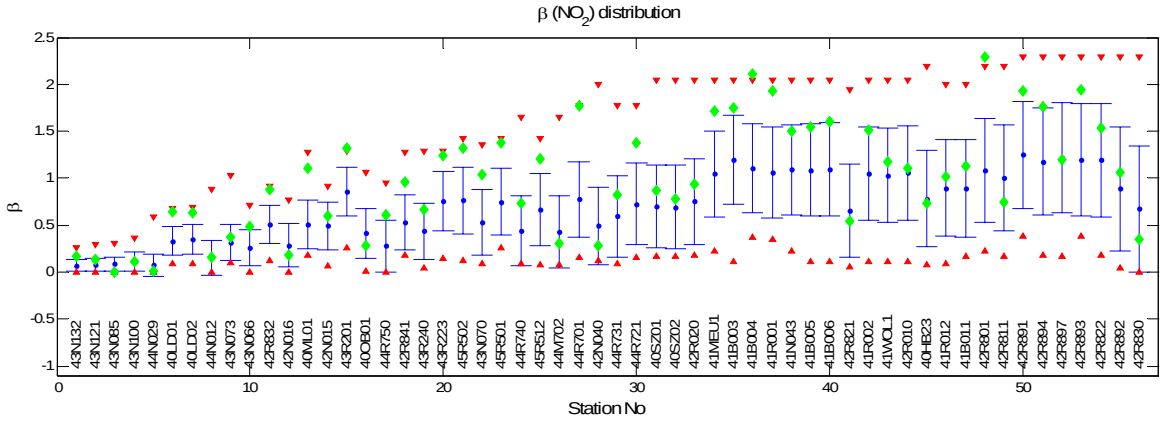

Figure 3: Variability of the NO_2 β-parameter in the vicinity (7.5 km radius) of the Belgian monitoring sites. The average values +/- one standard deviation are given as blue dots with error bars. The maximum and minimum β-values in the buffer are given as the triangles. The β-values of the monitoring sites are printed as green diamonds. Stations are sorted according to the standard deviation of the β-distribution.

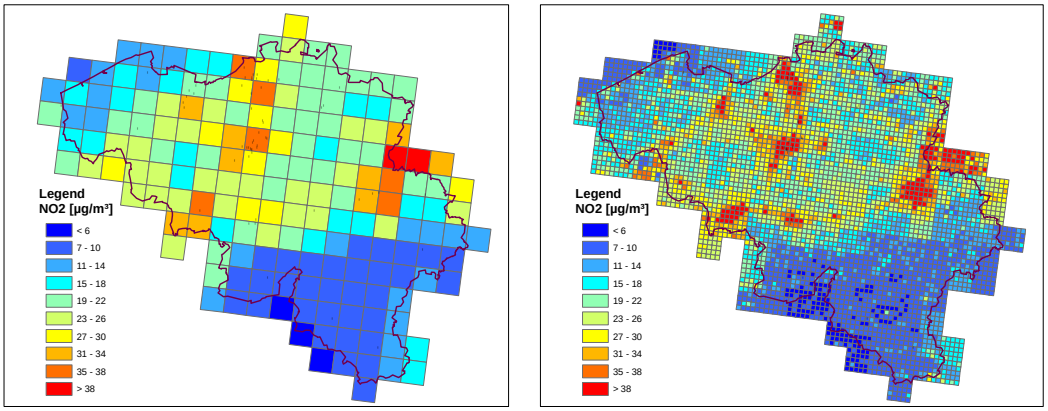

Figure 4: NO_2 map of Belgium for the annual mean concentrations for the year 2002. BelEUROS results on a 15x15 km² grid are presented on the left, the downscaled results on a 3x3 km² grid are presented on the right. Station locations are indicated by the black dots.

The improved model validation can now be demonstrated by comparing measurement data with the original BelEUROS 15x15 km² concentration fields and with the high resolution data downscaled into the 3x3 km² gridcells. The validation results are summarized in Figure 5. For the rural stations (low β-values at the left) no much difference is observed between the original and the downscaled model results. For stations with a higher β-value, the spatial representativeness becomes lower (see Figure 3) and more discrepancy is revealed between the original and the downscaled results. A positive bias is introduced in the majority of the measurement stations. This can be explained by the fact that most of those stations are located in an

urbanised area (higher β-value), compared to their direct environment. When both data sets are confronted to the measurement data, it is clear that the downscaling procedure does not have a positive effect in all stations. However, in the majority of the sampling sites, the shift introduced by the downscaling has a positive effect. This is also confirmed in the statistical analysis given in Table **1**. The three quality indicators presented in this table clearly show a general improvement in the model validation. The overall improvement of taking into account this spatial representativeness can be quantified as greater than 15%.

Table 1: Validation statistics for the annual mean NO_2 concentrations (µg/m³). Root Mean Square Error (RMSE), Mean Absolute Error (MAE) and bias are obtained by comparing model results with measurements in all monitoring sites. Statistics are given for the original low resolution BelEUROS model results and the high resolution downscaled results.

NO_2 (µg/m³)	High resolution (3x3 km² grid)	Low resolution (15x15 km² grid)
RMSE	8.18	9.69
MAE	6.16	7.26
BIAS	0.25	-5.07

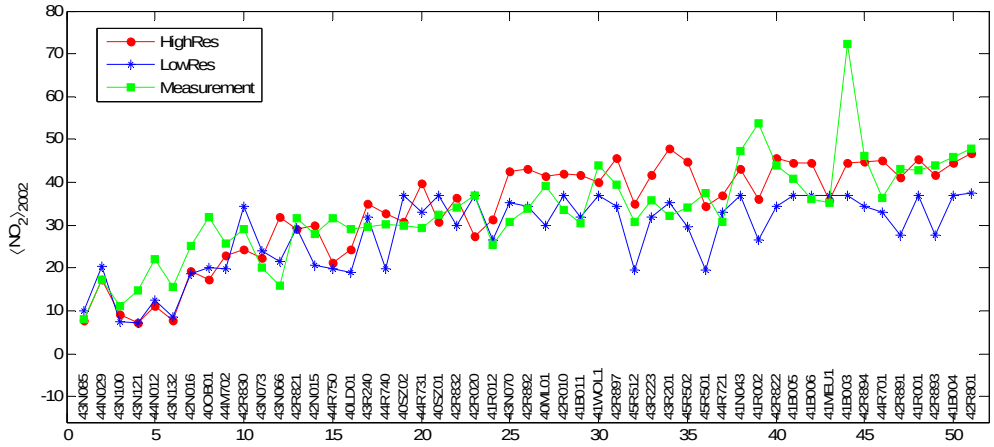

Figure 5: Validation results of the downscaling procedure for the 2002 annual mean NO_2 concentrations (µg/m³). Measurements are printed as green squares, low resolution (15x15 km²) BelEUROS results as blue stars and high resolution downscaled results (3x3 km²) as red dots. Stations are sorted according to their β-value: rural stations to the left, urban and industrial stations to the right.

4. CONCLUSIONS

In this paper a methodology is presented for the assessment of spatial representativeness of air quality monitoring data. The methodology relies on general relations between air pollution expectation values and land use characteristics. These characteristics are captured by a single parameter, calculated from the CORINE Land Cover data set and optimized per pollutant. The technique is applied here for the NO_2 measuring stations but is applicable for other pollutants as well.

The methodology developed for the assessment of spatial representativeness can also be applied to improve model validations. The technique was illustrated by making use of NO_2 concentration fields of the BelEUROS model. It was shown that by taking into account the effect of spatial representativeness of monitoring data model validation results can be improve, especially in urbanized and industrialized regions. Overall, the positive effect is estimated to be more than 15%.

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THE MODELLING SYSTEM SUPPORTING PIEMONTE REGION YEARLY AIR QUALITY ASSESSMENT.

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ABSTRACT
Regione Piemonte is in charge to elaborate air quality information to be transmitted to the European Commission on a yearly basis. From 2005 this institutional activity is supported by a modelling system capable to simulate air pollutant emission, transport, diffusion and chemical transformation, to provide concentration fields on a hourly basis and to compute all the indicators required by EU legislation. Computation results are provided as maps, tables and point values at measurement sites. Gridded emissions are computed from different emission inventories available for Piemonte and neighbouring Italian Regions and foreign countries. Meteorological fields are reconstructed assimilating ARPA Piemonte observations within ECMWF analyses background fields. Pollutants concentration fields are computed by the chemical transport model FARM. The model performance verification shows good results for all pollutants with exception of PM_{10}, for which underestimation of winter peak values is observed. Investigation is ongoing to improve particulate modelling in adverse winter conditions.

1. INTRODUCTION
The EU Air Quality Framework and Daughter Directives (1996/62/EC; 1999/30/EC, 2000/69/EC and 2002/3/EC) oblige the Member States to divide their territories in homogeneous zones and assess air quality within them, taking into account the different pollutants' limit and threshold values. The national implementations of EU Directives delegated to Italian Regions the air quality assessment (AQA) responsibility. Local observations are the basic method to determine pollutants' concentrations at the sampling sites, but measurements cannot cover the whole territory and the spatial generalisation of measured levels is not straightforward. These limits are particularly evident for areas characterised by varying topography and landscape and where atmospheric chemistry has a major role in determining ground level concentration of pollutants. To cope with the cited difficulties the Framework and the Daughter Directives introduced the possibility of using modelling for air quality assessment and management. The use of air quality models can improve the capability to map space distribution of pollutant concentrations at different scales (from regional background to city scale). The reliability of models has to be verified, comparing their results with measurements, to be able to quantify their accuracy. From 2005 ARPA Piemonte has built a three-dimensional modelling system capable to simulate air pollutant emission, transport, diffusion and chemical reactions, to estimate concentration fields for the main atmospheric pollutants (CO, NO_X, SO_2, PM_{10}, $PM_{2.5}$, O_3, and benzene) on a hourly basis over the whole Region territory. The modelling system is completed by post-processing tools to compute all the indicators required by EU air quality legislation. Concentration fields and air quality indexes are provided in form of maps, tables and point value to support the fulfilment of Regione Piemonte institutional duties and to facilitate the comparison of modelled concentrations with observations from the air quality monitoring network.

2. MODELLING SYSTEM AND COMPUTATIONAL METHOD
The AQA modelling system is built around four main components (Figure 1): the emissions processing system EMMA (ARIANET, 2005); the diagnostic meteorological model MINERVE (ARIA Tech., 2001); the interface module GAP/SurfPRO (Finardi et al., 2005), for the estimation of atmospheric turbulence and dispersion parameters; and the Eulerian chemical transport model FARM (COST728, 2006). The modelling system has been conceived to realise the best possible reconstruction of local air quality on the basis of the available information about emissions, meteorology and air quality, through the application of state-of-the art modelling techniques. Gridded emissions are computed on hourly basis starting from different emission inventories (high resolution regional INEMAR inventory for Piemonte and Lombardia, national CORINAIR inventory for the remaining Italian regions and EMEP for foreign countries). Hourly emission rates of the desired species are estimated by a pre-processing system allowing flexible space disaggregation, time modulation and non-methanic hydrocarbon speciation of inventory data related to point, line and area sources. Meteorological fields are reconstructed assimilating ARPA Piemonte meteorological network observations (including surface measurements and vertical soundings) within background fields obtained by ECMWF analyses. Eddy diffusivities and deposition velocities are evaluated using parameterisations based on the surface energy balance and similarity theory. The atmospheric pollutant concentration fields are finally obtained driving the chemical transport model FARM by means of boundary conditions provided by continental runs of the chemical transport model CHIMERE, from the INERIS Prev'Air service

(http://prevair.ineris.fr) and Regione Piemonte air quality monitoring network observations. The modelling system works on a computational domain of 220x284 km^2, covering the whole Piemonte and Valle d'Aosta Regions, part of Liguria, the eastern part Lombardia (including Milan urban area) and portions of France and Switzerland (Figure 2). The air quality model computational grid has an horizontal resolution of 4 km and 12 vertical levels, spanning the lower 3500 metres of the atmosphere.

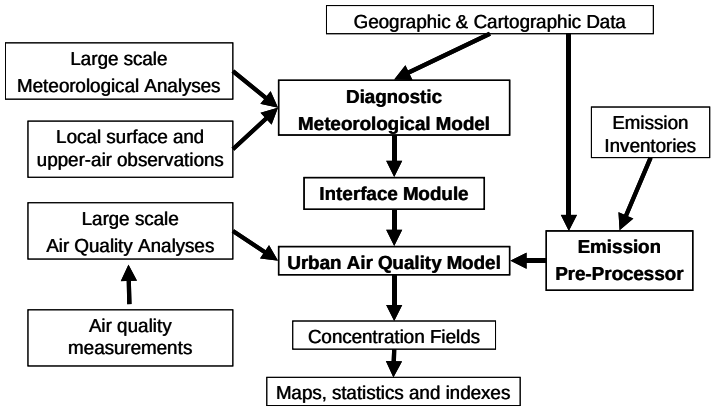

Figure 1. Piemonte Region air quality assessment modelling system architecture.

The modelling system has been built and optimised performing year 2004 AQA and successively applied for year 2005. The air quality model results have been compared with measurements provided by the air quality network stations operating over the whole region territory.

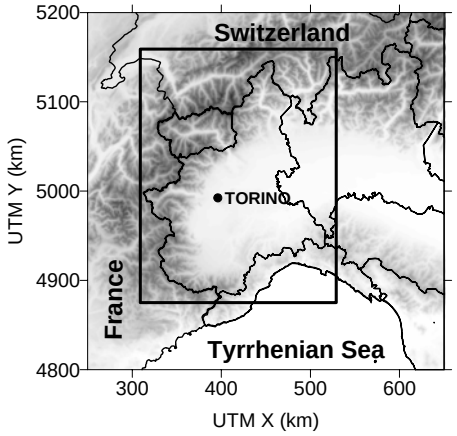

Figure 2. Piemonte Region air quality assessment modelling system computational domain.

3. YEARLY AIR QUALITY ASSESSMENT ANALYSIS AND VERIFICATION

Hourly average concentration fields have been processed to calculate air quality indexes and compare them with values obtained from air quality monitoring sites, in order to verify the accuracy of model results as prescribed by current legislation for the different pollutants. The comparison considered each monitoring station having at least 90% of valid data over the whole year, and it took into account the space representativity of the monitoring site and model simulation resolution. Particular interest is here addressed to results obtained for those pollutants which are responsible of the largest number of legislation threshold exceedances in the region, i.e. PM_{10}, nitrogen dioxide and ozone. The accuracy requested to model results for these pollutants is reported in Table 1 (cfr. respectively 1999/30/EC for PM_{10} and NO_2 and 2000/69/EC for O_3). Examples of results are illustrated in Figure 3 by scatter plots of PM_{10} and O_3 measured vs. simulated concentrations. Vertical and horizontal lines stand for limit values threshold, while the area between diagonal lines represent acceptable accuracy. The analysis of ozone indicators shows satisfactory results, with good accuracy in reproducing measured levels, for both eight-hours and hourly averages, at rural and urban background stations. Minor problems can be detected in simulating of very low concentration levels, but it has to be taken into account that measurements accuracy is generally lower too for low concentrations. Figure 3 shows two scatter plots for the eight-hour average concentrations at Buttigliera d'Asti (rural background station, bottom left panel) and at Novara-Verdi (urban background station, bottom right panel).

The assessment of ozone concentration levels highlights that limit values are exceeded in many areas, particularly in the flat region, with the exception of the main urban areas.

Table 1. Modelling accuracy as provided by European and Italian legislation.

	PM_{10} %	NO_2 %	O_3 %
Hourly averages	--	50 ÷ 60	50
Daily averages	Not defined	50	--
Eight-hour averages	--	50	50
Yearly averages	50	30	--

Figure 3 (top left panel) summarizes results obtained for PM_{10} yearly averages. The stations where underestimation makes model accuracy fall below the requested 50% value represent traffic related areas in middle size cities which can be hardly described by the model at the resolution of 4 km. The daily average values at Torino-Consolata station are reproduced with acceptable accuracy for most of the year (Figure 3, top right panel), even if values larger than 100 μg/m^3 are underestimated. PM_{10} is considered the most critical pollutant not only in Piemonte region but in the whole Po Valley. Emissions from large urbanized areas together with industries and road traffic cause the occurrence of high concentration levels during most of the winter period, when persistent low wind conditions prevail. Therefore, it is of particular importance a properly reproduction of particulate concentration.

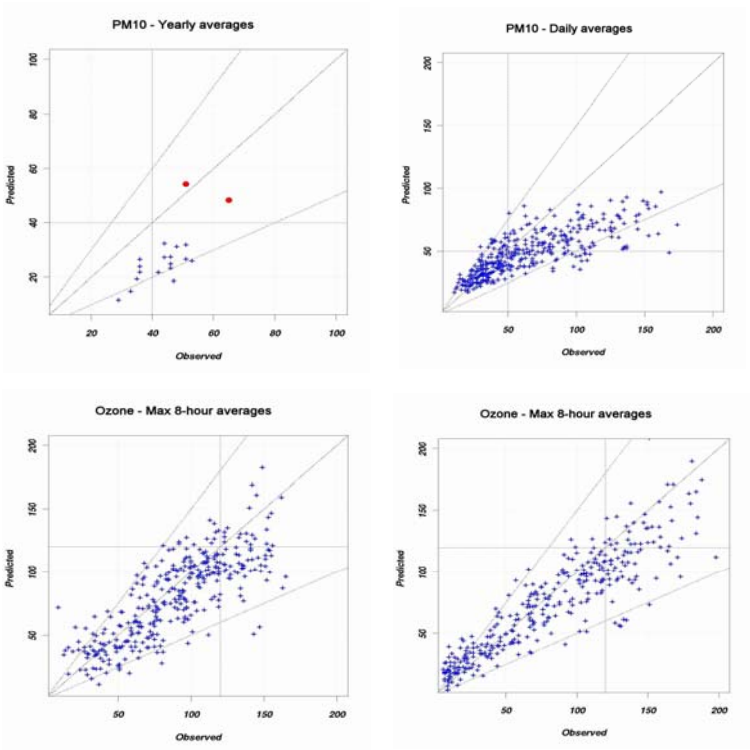

Figure 3. Scatter diagrams for: PM_{10} yearly average concentrations (top left; dots indicate monitoring sites located inside Torino); PM_{10} daily average concentration at Torino-Consolata station (top right); O_3 maximum 8-hour average concentrations at Buttigliera d'Asti (bottom left) and Novara Verdi (bottom right) stations. All concentrations are indicated in μg/m^3 and refer to year 2005.

The requested accuracy level for both daily and yearly averages of PM_{10} concentrations (Table 1) are accomplished in most of the measuring sites, even though a clear underestimation can be noticed for stations located outside the city of Torino. Differences between measured and predicted values can be ascribed to a certain underestimation of emissions outside the metropolitan area of Torino, particularly for what regards wood burning emissions from house heating systems. Moreover, the modeled background concentrations are lower than values usually recorded in the Po Valley indicating the difficulty to simulate large scale PM accumulation phenomena.

The regional air quality monitoring network observations show that NO_2 limits for hourly and yearly averages concentrations are exceeded mainly inside the Torino metropolitan area. These condition is confirmed by model simulation results (Figure 4). The model accuracy remains within the requested limit for most monitoring sites. Some underestimation of peak concentration has been detected during adverse meteorological conditions, when local features cannot be properly reproduced by the modelling system at the operational resolution.

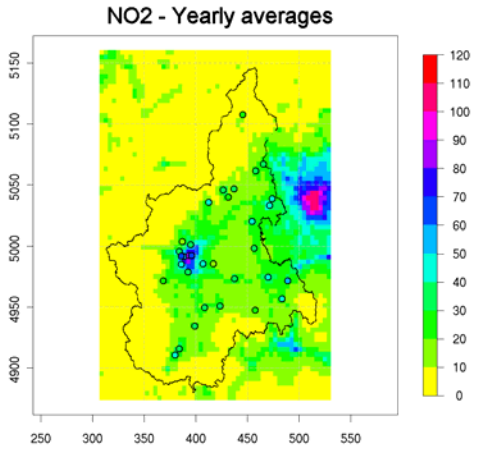

Figure 4. Simulated and observed NO_2 yearly average concentration ($\mu g/m^3$) during year 2005. Coloured dots represent measured values at the monitoring sites.

4. EVALUATION OF INDICATORS AND THRESHOLDS REQUIRED BY THE EC

The air quality assessment must refer to territorial units (municipalities for Piemonte Region) that are needed to define a proper classification of pollution levels on the basis of measured and/or modelled concentrations. The possibly needed action plans to improve air quality have to refer to those territorial units. Model simulation results are produced as gridded fields, where every grid cell provides a value representative of the average features of the portion of land covered. A GIS based method has been developed to assign model results to the territorial units. Superposing concentration fields to thematic cartography, the grid points overlapping each municipality are identified. The distribution of concentration values of grid points falling within each municipality is then computed. The 90° percentile of grid points values is selected as representative of concentration level in each territorial unit. This choice allows to be conservative in the air quality assessment of the whole municipality, excluding peak values representative of very small portion of land. To identify the presence of exceedances of air quality limits, the maximum of grid concentration value is considered for each territorial unit.

5. CONCLUSIONS

The Piemonte Region AQA modelling system has been evaluated comparing modelled concentrations with measurements from stations located in different geographic, topographic and land cover environment. Good results have been obtained for NO_2 and O_3 concentrations. The space structure and time variation of concentration fields are satisfactorily reproduced, yearly average and statistical distribution of concentrations are compatible with measured values. PM_{10} concentrations have been reconstructed with accuracy within acceptable limits at most station locations, even if underestimation has been detected during wintertime, when peak concentrations recorded during air quality episodes have not been captured. Further investigation is required to clarify the reason of this difficulty, while improvement of emission description is in progress. The positive results of model verification, together with the experience of using modelled concentration fields for AQA, induced Regione Piemonte to implement the modelling system as a permanent AQA tool and ARPA Piemonte is now in charge to perform modelling applications on a yearly basis.

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ANALYSIS OF INDUSTRIAL-URBAN NO_x SAMPLING DATA

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ABSTRACT

An analysis of a four-station NO_x survey conducted near an industrial area and its surroundings located in the Greater Buenos Aires is presented. Total variance (V_{ij}) at site "i" and wind direction "j" is broken down into three parts: an error component that represents variability attributable to the sampling and analysis methods, a zonal component due to urban-regional contribution and a local component originated by the variability of emissions from sources in the vicinity of each sampling station. The percentage of total variance that can be attributed to the error component is between 1.2% and 12.6%. Depending on wind direction and monitoring site, the zonal component contributes with 3.2% to 73.2% of total variance. The largest contribution of local component is 98.7% (site 1), 95.8% (site 2), 65.9% (site 3) and 93.9% (site 4) of V_{ij}.

1. INTRODUCTION

The cities and their surroundings are by nature concentrations of humans, materials and activities. They therefore exhibit both the highest levels of pollution and the largest targets of impact. Air pollution control depends upon adequate current data and projections describing concentrations of the important pollutants within the control region. In most of industrialised world urban air pollution is now monitored routinely. Often the data are based on one or a few stations, placed at critical sites and thus represent microenvironments. It should also be taken into account that coverage of stations is different for different countries (Mage et al, 1996), and that average values can therefore be differently biased.

The air quality in the city of Buenos Aires (Argentine) has been the subject of several studies carried out during the last years, using data obtained from different measurement surveys of pollutants in urban air (Bogo et al., 2001, 2003; Venegas and Mazzeo, 2000; Mazzeo and Venegas, 2002; Mazzeo et al., 2005; Bocca et al., 2006). In the Greater Buenos Aires, very few air quality measurements have been made (Fagundez et al., 2001, JICA-SAyDS, 2002).

This paper presents an analysis of multiple-station air quality data collected in an industrial-urban area. Measurements of hourly NO_x concentrations were made at four stations in an industrial area (IA) and its surroundings located in the Greater Buenos Aires, SE Buenos Aires city. Assuming that actual data arise from a zonal contribution (at an urban-regional scale), a local scale contribution and errors due to sampling and analysis methods, the contribution of these components to pollutant variability is estimated.

2. METHODOLOGY

When several observing sites are used to characterize the air quality in an area, the variations in the resulting data represent a mixture of instrumental error, local (transient) phenomena and medium scale variability in air pollutant concentrations. The best use of the field data results when estimates are available concerning the relative magnitudes of the observing errors, local phenomena and regional-scale process.

For this discussion the actual concentration value (X_i) observed at monitoring site "i" is assumed to arise from an urban-regional contribution (zonal) (Z_i), a local contribution (L_i) arising from sources located nearby the measuring site, and errors (e_i) due to measurement and analysis methods:

$$X_i = Z_i + L_i + e_i \quad (1)$$

With the aid of Eq. (1) the variance (V_i) of concentration data X_i can be expressed in terms of defined zonal ($V(Z_i)$), local ($V(L_i)$) and error ($V(e_i)$) contributions to the variance V_i:

$$V_i = V(Z_i) + V(L_i) + V(e_i) + 2Cov(Z_i,L_i) + 2Cov(Z_i,e_i) + 2Cov(L_i,e_i) \quad (2)$$

where $Cov(Z_i,L_i)$, $Cov(Z_i,e_i)$ and $Cov(L_i,e_i)$ are the co-variances between the zonal and local contributions, between the zonal and error contributions, and between the local and error contributions, respectively. Assuming that error contributions are random (i.e., uncorrelated with the regional and local variabilities), Eq. (2) reduces to:

$$V_i = V(Z_i) + V(L_i) + V(e_i) + 2Cov(Z_i,L_i) \quad (3)$$

3. RESULTS

Three months of hourly NO_x concentrations registered during working days at four sites (i=1,2,3,4) located in an industrial area (IA) and its surroundings are analysed. The area of interest (3km^2) is located in the Greater Buenos Aires (Figure 1) on the west coast of the de la Plata River, SE the city of Buenos Aires. At this site, the de la Plata River is 46km width and it can be considered that wind blows "clean air" from the river (65% of the time). There are fuel storage tanks and two refineries in the IA with point sources that emit 5.2ton-NO_x/day. There are also a port entrance and several docks with a traffic of approximately 420 ships/month (cargo boats and passenger ships). An elevated freeway with a mean traffic volume of 60000veh/day runs west the IA. There are also two Thermal Power Plants in the area. The large one is located north and emits 55.4ton-NO_x/day, the small one is next site 2 and emits 5.1ton-NO_x/day.

Figure 1. Monitoring site locations

In general, diurnal variation of NO_x registered during working days follows diurnal traffic flow variation, as can be seen in Figure 2. Site 3 shows the lowest concentration values. At this site there is very low influence of mobile sources (except for the relative morning peak). On the other hand, site 1 shows the highest NO_x concentrations. At this site, concentration levels result from local emissions of mobile sources (mainly tank trucks) and ships (oil tankers and cargo boats) and from the point sources of the Thermal Power Plants and the refineries.

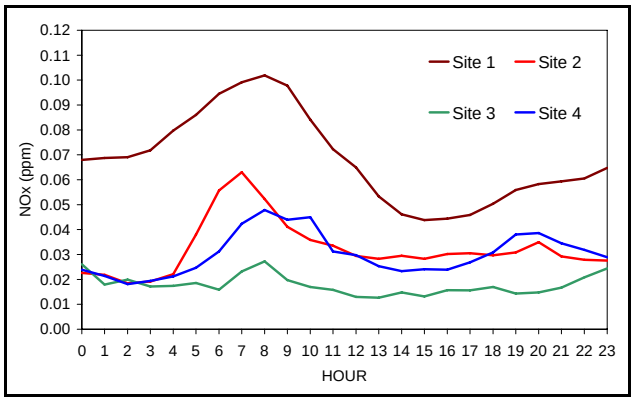

Figure 2. Diurnal variation of average NO_x at each site.

The variation with wind direction of mean values and total variances of NO_x hourly concentrations at each monitoring site, are shown in Figures 3 and 4, respectively. The largest variance (3.55E-03ppm^2) is observed at site 1, for wind direction NW. This result may be associated to the variability caused by mobile source emissions (road traffic and boats) passing near the monitor or at the docks.

The contributions of the three components to total variance of NO_x concentration data at each monitoring site have been estimated for eight wind directions. The zonal component contributes with an "urban background" concentration if the wind passes through the urban area before reaching the study area, otherwise, this component can be considered negligible. Therefore, it is not unreal to consider that $Cov(Z_i,L_i)$ is negligible in Eq.(3). Furthermore, if observations are grouped according to wind direction, the variance (V_{ij}) of observations at site (i) registered for wind direction (j) is given by:

$$V_{ij} = V(Z_i)_j + V(L_i)_j + V(e_i) \quad (4)$$

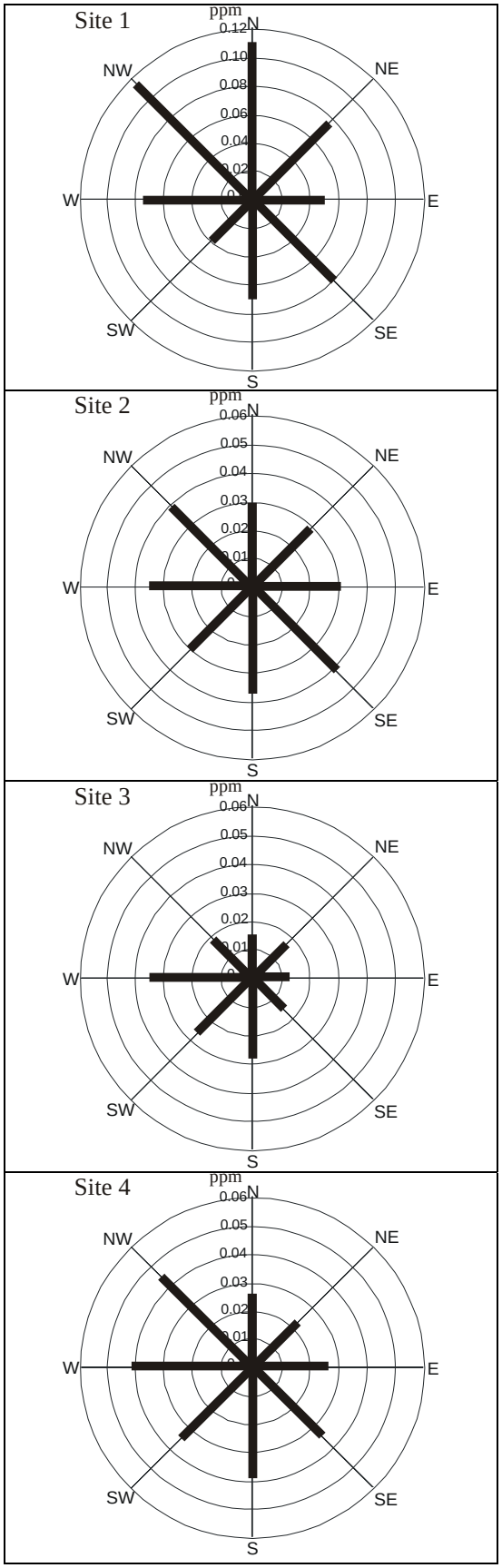

Figure 3. Variation of mean NO_x concentration with wind direction at each monitoring site

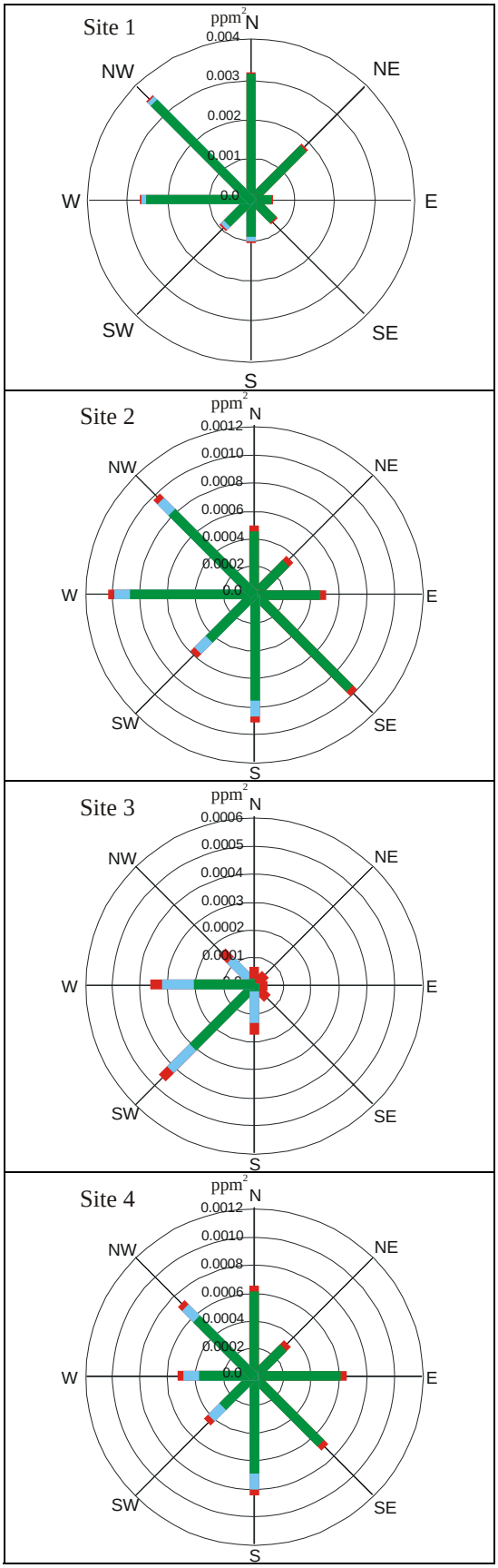

Figure 4. Variation of the variance of NO_x concentrations with wind direction at each monitoring site. $V(L_i)_j$, $V(Z)_j$ and $V(e)$ contributions.

The smallest mean concentration value (1.3E-02ppm) can be found at site 3 with eastern wind (Figure 3). These observations also show the smallest V_{ij} =4.2E-05 ppm^2 (Figure 4). During these conditions the wind blows clean air from the wide river towards site 3. For this reason, this value has been assigned to V(e) (assumed the same for all monitoring sites and wind directions). For wind directions coming from the river (j=N, NE, E, SE), the contribution $V(Z_i)_j$ has been considered negligible at all monitoring sites, as there are not any urban-regional emissions upwind. Analysing the values for S, SW, W and NW wind directions, the smallest mean NO_x concentration (1.9E-02ppm) and the smallest V_{ij} (=1.56E-04ppm^2) can be found at site 3 (for NW). As this site shows very low influence of local sources, it is assumed that these low values are associated to zonal (urban-regional) and errors contributions. In this way the value $V(Z_i)_j$=1.14E-04ppm^2 (j= S, SW, W, NW) is obtained. The main sources of urban-regional pollution for the study area are the city of Buenos Aires and the Greater Buenos Aires. Both areas show similar variability caused mainly by emissions from traffic so it is assumed that the zonal component is similar for all sites and depends only on wind direction, $V(Z_i)_j=V(Z)_j$. Finally, for each site and wind direction, the values of local contribution $V(L_i)_j = V_{ij} - V(Z)_j - V(e)$ are estimated. The contributions $V(L_i)_j$, $V(Z)_j$ and V(e) are indicated in Figure 4.

Except for site 3 and depending on wind direction, the percentage of total variance that can be attributed to V(e) is between 1.2% and 12.6% of V_{ij}. Depending on wind direction and monitoring site, urban (zonal) contribution to V_{ij} varies between 3.2% and 73.2%. Largest $V(L_i)_j$ are responsible for 98.7% (site 1), 95.8% (site 2), 65.9% (site 3) and 93.9% (site 4) of $V_{ij.}$. These high contributions of local component to the variance are mainly associated to mobile sources (depending on the monitoring site, may be from traffic flow at the streets nearby, the freeway and/or the boats at the docks).

4. CONCLUSIONS

This paper presents a statistical approach to the analysis of multiple air pollution sampling data in an industrial-urban area. Total variance (V_{ij}) at site "i" and wind direction "j" is broken down into three parts: a zonal ($V(Z)_j$) component due to urban-regional contribution (assumed similar to all sites), a local component ($V(L_i)_j$) originated by the variability of sources in the vicinity of each sampling station and a measurement component (V(e)) that represents variability attributable to the sampling and analysis methods. Three months of hourly NO_x concentrations registered during working days at four sites in an industrial area, located in the Greater Buenos Aires and its surroundings are analysed. The error contribution to the total variance in most field data series is significant (between 1.2% and 12.6%). Depending on wind direction and monitoring site, urban-regional contribution to V_{ij} varies between 3.2% and 73.2%. Largest $V(L_i)_j$ are responsible for 98.7% (site 1), 95.8% (site 2), 65.9% (site 3) and 93.9% (site 4) of $V_{ij.}$.

5. ACKNOWLEDGEMENTS

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SIMPLE CURVES FOR ESTIMATING NITROGEN DIOXIDE IN INDUSTRIAL PLUMES.

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ABSTRACT

Regulators need to assess non-linear contributions of industrial sources to secondary pollutants. The UK Environment Agency therefore funded advanced modelling. The Met Office NAME III Lagrangian stochastic dispersion model was used to model an idealised large point source plume. Eulerian chemistry was used. A plume with NO and NO_2 was dispersed in a model atmosphere with uniform meteorology and background concentrations of O_3 and VOCs. Simple tools are needed to screen out many permit applications for which advanced modelling is not appropriate. This talk proposes simple curves for screening NO_2 in industrial plumes, using our model results, comparing them with empirical functions for urban measurements of the ratio NO_2:NO_x that are used in the UK, as screening tools in LAQM. Simple screening curves to indicate the likely range of NO_2/NO_x for industrial plumes may be useful as a Regulatory screening tool, before advanced, costly modelling is used.

1. INTRODUCTION

Nitrogen dioxide NO_2, ozone O_3 and volatile organic compounds VOCs challenge local air quality management (LAQM), AQEG, (2004). Regulators need simple tools that can be used to assess the non-linear contributions of industrial sources to secondary pollutants. This complex problem requires advanced modelling. Such models need evaluation of their suitability for this regulatory work. The UK Environment Agency therefore funded work to assess the possible use of advanced models. Thus two scenarios, an idealised plume and pollution episodes, were modelled using Models3-CMAQ and NAME III respectively (Yu et al., 2007). The Agency also funded work to review the literature on NO to NO_2 conversion (Middleton et al., 2007), and propose a practical methodology to estimate the NO_2/NO_x ratio in plumes. Simple tools are needed to screen out many permit applications for which advanced modelling is not appropriate. This work to devise simpler regulatory approaches for NO_2 forms the basis of the present paper. This talk proposes simple curves for screening NO_2 in industrial plumes, using our model results.

Concentrations downwind of large sources are affected by meteorology, influencing plume rise, advection, and dispersion. Photochemical production of secondary pollutants depends on light intensity, a function of solar elevation and cloud cover. Reaction rates in the plume depend on precursors: NO and NO_2 from the source and O_3 and VOCs in the surrounding atmosphere. Janssen (1986), Janssen et al. (1988), highlight the importance of inhomogeneous mixing in reacting plumes. Near-source, NO_2 formation is diffusion limited, whilst further down-wind it becomes asymptotic to the photo stationary state, depending upon O_3 concentration, photolysis rate J, and rate of reaction k. The problem is complicated by background VOC concentrations, important if an industrial plume mixes into polluted air masses. The Met Office NAME III Lagrangian dispersion model was used to model an idealised scenario. Model particles disperse stochastically, representing in-homogeneous mixing. Chemistry is then simulated in an Eulerian grid, before the next dispersion step. A large point source plume containing NO and NO_2 was dispersed in a model atmosphere with uniform meteorology. Background concentrations of O_3 and VOCs were systematically varied. The modelling setup is described in Middleton et al. (2006a, 2006b), and Yu et al. (2007). Here we derive simple enclosing curves to encompass the range of NO_2/NO_x ratios seen in our idealised model results. We then compare our curves for the industrial plume NAME III model results with empirical curves for the ratio NO_2/NO_x that are used in the UK, as screening tools in LAQM. Such urban curves have summarised extensive urban monitoring data: Derwent and Middleton, 1996; Dixon et al., 2001. We conclude that simple screening curves to indicate the likely range of NO_2/NO_x for industrial plumes may be useful as a Regulatory screening tool, before advanced, costly modelling is used. Application of advanced modelling tools to Regulation of plumes requires further work.

2. MODELLING SET-UP USING NAME III

The **N**umerical **A**tmospheric-Dispersion **M**odelling **E**nvironment, NAME III, is a Lagrangian particle-trajectory model designed to predict the atmospheric dispersion and deposition of gases and particulates. NAME III is summarised by Middleton et al. (2006a, 2006b) and references cited therein. Model particles carrying pollutants, NO, NO_2, represent releases from a tall stack (200m). Particles disperse by stochastic or random walk processes, and advect following the flow field. The random walk modelling means that inhomogeneous plumes can be modelled, as recommended in Janssen (1986), Janssen et al. (1988), but the chemistry is then solved in an Eulerian grid between successive Lagrangian model dispersion steps. The NAME model can be used to simulate emissions and atmospheric reactions over many scales. In this study, we use a small model domain whose size is limited to minimise the very large number of model particles needed to initialise the background pollutants. This is a special case, for the model usually operates in a

standard latitude/longitude co-ordinate system. Here, as in Middleton et al. (2006a,2006b) and Yu (2007) we used a uniform wind of 4 m s^{-1} from the West, a neutral boundary layer (heat flux 0) with depth 564 m and clear skies. A mid summer diurnal cycle for solar elevation meant the change in chemistry between day and night was represented. When studying the plume chemistry the simplified meteorology made it easier to examine the results. Turning of the wind with height may be switched off as in most of our idealized runs, or on. Wind turning is important for realistic modelling of large plumes. The source emitted NO_2, NO and SO_2 at constant rates of 50, 950, and 8000 g s^{-1} respectively. The primary NO_2 was thus the commonly adopted value of 5% of emitted NO_x. In addition to source model particles, we use very many background particles. These supply the reactants O_3 and VOC to combine with the NO and NO_2 from the plume. By carrying the model O_3 and VOC on all the model particles we allow advection, stochastic dispersion, and more realistic interaction of O_3 and VOC with the NO_x plume. Total background VOC concentrations were varied for each run as 5, 10, 20, 50, 75 ppb. Background O_3 concentrations were 10, 20, 50, 100, 150 ppb, varied independently of VOC. Overall 25 combinations were modelled in a series of runs. These will be used to illustrate the results. The concentrations are seen fluctuating from one grid square to the next because of the stochastic nature of this Lagrangian model. With larger numbers of model particles, smoother fields could be obtained, but require much more memory and computer time.The chemistry scheme is summarised by Middleton et al. (2006a, 2006b) and Yu et al. (2007), and references therein, including the ratios of the 7 volatile species that constituted our VOC background. Model runs are also listed there.

3. PLUME RESULTS

Figure 1(a) shows NO_2 formation in the plume at mid-afternoon, and 1(b) has the corresponding removal of O_3 near the stack and its subsequent formation further downwind, reaching a concentration well above background. These plan views of concentrations are the lowest 100 m of the atmosphere to 1/2 stack height.

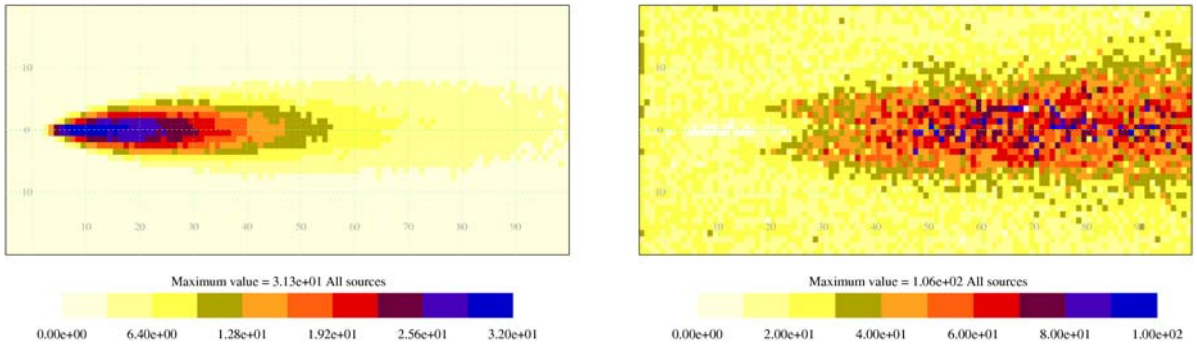

Figure 1(a) *Left*: Run 27: NO_2 in the plume at daytime (1500 local time) with background of 20 ppb O_3 and 50 ppb total VOCs. Figure 1(b) *Right*: Run 27: O_3 removal from background near the stack by titration with NO; also O_3 formation further downwind by photochemistry in the plume at daytime at 1500 hours with background of 20 ppb O_3 and 50 ppb total VOCs. The background O_3 was initialised at 20 ppb, was reduced by the NO plume, then increased by photochemistry after ~30 km to O_3 ~ 106 ppb.

Figure 2(a) shows the same situation at midnight, with diffusion limited formation of NO_2 revealed by the larger concentrations of NO_2 towards the plume edges. The corresponding O_3 field shows no O_3 formation at night, just O_3 removal, consistent with the lack of light.

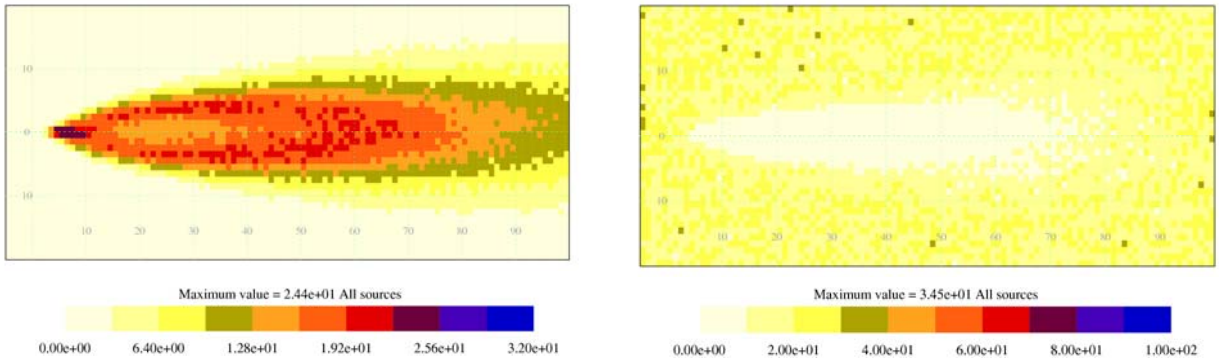

Figure 2(a) *Left*: Run 27. NO_2 in the plume at midnight (2400 local time) with the same background of 20 ppb O_3 and 50 ppb total VOCs. as above. Figure 2(b) *Right*: Run 27 O_3 removal from background near the stack and out to ~80 km away by titration with NO in the plume at midnight (2400 local time) with same background of 20 ppb O_3 and 50 ppb total VOCs as above. There is no increase in O_3 within the domain; also the depleted O_3 background extends much further from the stack, from ~4 km to ~80 km.

4. SIMPLE PLUME NO_2 SCREENING CURVES

Having emulated an inhomogeneous plume of NO (95%) and NO_2 (5%) mixing into a polluted atmosphere with chemical reactions at night and day, we give a methodology that might serve as a Regulatory screening tool. Existing simple methods found in the literature (Middleton et al., 2007) were hitherto based on analyses of measurements for a given site over several years of hourly monitoring data e.g. Derwent and Middleton (1996); Dixon et al. (2001). With this background, in analysing urban data, the present study analyses results from the NAME III runs. The urban data were analysed taking successive points along the time series; here we have no such time series. The non-linearity in the NO_2/NO_x curve is an inherent feature of the chemistry. Therefore here the usual time sequential sampling at a single point (the 'monitoring station') was replaced by spatial sampling over the whole NAME III downwind modelled domain (some 100 by 40 squares) for a single model time step. This amounts to a transformation between a time domain for many successive observations at one point, and a spatial domain at many points sampled for a single model time interval. We hypothesise that the chemistry was robust to the transformation between temporal sampling at a point (the air moves past the monitor) and spatial sampling at a time step (the 'monitor' or 'sampler' moves over the entire plume area, with points further downwind having been emitted earlier in time). We demonstrate below that this alternative method of spatial rather than temporal sampling is indeed remarkably similar in terms of its practical application in leading to a possible screening approach. Philosophically, this means that our hypothesis of robustness in chemistry towards spatial vis a vis temporal sampling appears justified.

The talk presents results generated by the spatial sampling over the NAME III modelled domain at 3-h time intervals. In the scatter plots (to be presented) of NO_2 versus NO_x for the NAME III output data, each point represents the concentrations from the grid points downwind and cross wind from the idealised point source. Each point is a 3-hour average for a 1 km × 1 km 100 m deep cell. We also show the associated scatter plot of the ratio NO_2/NO_x, versus NO_x. When analysing the NAME III scatter plots, it was found that polynomials needed several terms if they were to capture the general nature of the results, but these curve fits proved to be unreliable – they were prone to go off to infinity, or plunge towards large negative values. Neither of these behaviours was realistic for the ratio, which by definition has the range 0.0-1.0. Polynomial curve fitting was thus abandoned, even though it had proved useful in the earlier urban studies. The method suggested below now uses the concept of enclosing curves to obviate the polynomial curve fitting problems.

At each of the 105 × 40 1 km × 1 km squares in the pollution domain, we sampled the NO_2 and the NO_x and calculated the ratio NO_2/NO_x. Points were tested against the proposed enclosing curves. The number of points falling into each category, such as above or below the curve, was counted, and expressed as a percentage of the total number of points in each scatter plot. The aim was to assess the fraction of model results in the scatter plot whose NO_2/NO_x ratio was enclosed by the curves, or outside them. Space precludes these results being tabulated here (see Yu et al., 2007). In many of the runs (but not all) there were 85% or more points falling between the two curves presented below.

The talk shows two widely used empirical functions for urban data. We have plotted the night and day enclosing curves from NAME III, and by trial and error we found that if we use a background ozone concentration of 50 ppb, they would enclose the urban monitoring curves. This means that there is a strong similarity in the general form of the 3-hourly NAME III model results for the elevated plumes and in the empirical functions that represent urban NO_2 and NO_x 1-hourly monitoring data. This is a very interesting result, for it strengthens confidence in the idea that simple curves can be used to represent the NO2/NOx ratio in large industrial plumes, and that they bear a good similarity in general behaviour to the much more extensive urban data sets. Provided it is accepted that the simple day and night curves should be used as conservative screening tools (and not as best fits to the data – no formal curve fitting was used in their derivation – just analysis of the general asymptotic behaviour that seemed necessary), then this opens the way for a simple method to assess industrial plumes, before using complex models. Furthermore, the day and night enclosing curves are sensitive to background ozone concentrations, which means that future trends in the ozone climate can be easily accommodated by the curves. All the regulators need do is specify the ozone concentration to be used in the two curves. This could be increased year on year if necessary.

Finally, we also suggest that there is the possibility of using daily mean forecasts of ozone together with the curves for process operators to assess their likely NO_2/NO_x ratios on a daily basis when operating large combustion plant.

It was decided to adopt functions that would have the desired asymptotic behaviour. This is to approach unity at small NO_x, and to approach zero at large NO_x. After some experiment the following form was arrived at:

At night, the upper curve is $Y = \frac{[O_3]}{([NO_x]+[O_3]/4)}$ with the constraint that it does not go above unity, so we require $Y = 1.0$ when $[NO_x] \le 3[O_3]/4$. In the day, the lower curve is $Y = \frac{[O_3]}{([NO_x]+[O_3])}$ and this never goes above unity with $[NO_x] > 0$, which is always true for a meaningful NO_x concentration. The night curve is the upper one, larger NO_2/NO_x, because at night there is no light and the NAME III model turns off the photolysis of NO_2. This means the ratio Y = NO_2/NO_x is greater at night than day, for the same NO_x concentration. This is clearly seen in the NAME III scatter plots. In using these functions for day and night the same background for O3 is used as was defined in the background domain for the NAME III model runs. We have plotted a series of these curves, Yu et al. (2007), increasing the background ozone concentration through values of 20, 35, 50, 75, 100 and 150 ppb in turn. The curves show a well behaved response to increases in background ozone. If they have the same ozone background, then the form of the curves is such that they never cross each other, converging towards zero at vary large NO_x concentrations.

5. CONCLUSIONS

This work was funded by the Environment Agency to apply advanced modelling to the challenges raised by Regulation of secondary pollutants in large industrial plumes. Having developed an idealised modelling scenario we have found that polynomials through the modelled results for NO_2/NO_x were not reliable as they were sometimes poorly behaved. Instead we propose that enclosing curves to define the likely upper (night-time) and lower (day-time) ratios of NO_2/NO_x can encompass many (but not quite all) of our model results. They included day and night values as well as a range of background O_3 and VOC concentrations seen in these model runs. These simple curves represent a possible approach for initial screening of permit applications for plumes liable to form NO_2. At high NO_x concentrations it is a conservative approach to use the larger of the NO_2/NO_x ratios from the new enclosing plume curves, or the traditional urban curves.

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ON THE IMPACT OF PCDD/Fs EMITTED FROM A 2006 LANDFILL FIRE NEAR THESSALONIKI, GREECE

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ABSTRACT

Landfill fires raise significant air quality issues, as a broad range of toxic pollutants are produced and emitted into the atmosphere. In July 2006, such an incident occurred at the 'Tagarades' waste disposal site, which serves the Thessaloniki urban area in Northern Greece. Mathematical models are applied on a 48×48 km^2 domain for simulating the atmospheric dispersion and deposition of PCDDs and PCDFs which were emitted during this incident. Approximately 17 g I-TEQ of PCDD/Fs were estimated having been emitted during the landfill fire and about 10% of this amount were assessed having been deposited on the ground of the affected area.

1. INTRODUCTION

The present work aimed at assessing atmospheric dispersion and deposition of PCDD/Fs emitted from a major landfill fire at the Tagarades waste disposal site, near the city of Thessaloniki in Northern Greece (Figure 1). This site, located 23 km from downtown, is operating since 1991, currently receiving over 2300 Mg waste per day. The fire broke out on July 14th, 2006 in a filled part and over an area of 15,000 m^2; it lasted 16 days during which about 57,500 Mg of waste were burned, raising significant public and political concern. Relatively few studies have been conducted on PCDD/F amounts emitted from landfill fires: In an experimental study of two landfill fires in Finland (Ruokojarvi et al. 1995), measured PCDD/F concentrations in the working air during the active fire varied from 51 to 427 pg TEQ/m^3. On the other hand, Swedish pilot tests in landfill fires (Bergström and Björner, 1992) showed much higher concentrations of PCDD/Fs, i.e. between 66 and 518 ng(TE-Nordic)/m^3. Some studies have also been addressing the concentration of PCDD/Fs in the soil of and around landfills and open dumps: Martens et al. (1998) measured 1,144 ng I-TEQ PCDD/Fs per kg at soil samples of 'Kouroupitos' open dump in the island of Crete in Greece, whereas Minh et al. (2003) examined soil samples of open dumps in the Philippines, Cambodia, India and Vietnam and measured maximum PCDD/F concentrations of 1,700 ng I-TEQ/kg.

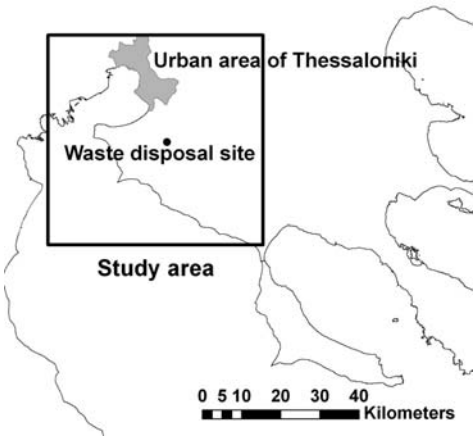

Figure 1. Locations of the Tagarades waste disposal site and the city of Thessaloniki.

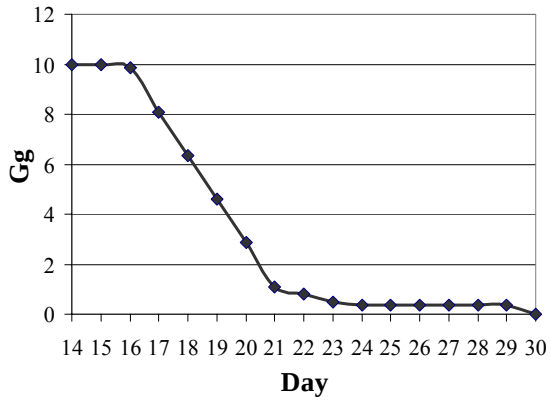

Figure 2. Assumed temporal variation of the waste burning rate.

For estimating PCDD/F emissions in the investigated Tagarades incident, the total amount of burned waste and its burning rate had to be estimated initially, a task that proved very difficult and still involves a large degree of uncertainty. The research team had to resolve to field surveys and interviews with involved personnel, in order to quickly approximate the open-burning rate, as this work was initiated and partly performed while the incident was still on-going: Thus, for the first three days (July 14-16), a steady rate of 10,000 Mg/day was concluded; due to intensified fire-extinguishing efforts, this rate was estimated to gradually decrease in a linear fashion and by July 21, it was concluded that it had reached 1,250 Mg/day. Over the next four days (July 21-24), this rate decreased further by 300 Mg/day and after July 24th and up to complete extinguishment (July 29th), the burning rate was considered as constant at 350 Mg/day (Figure 2).
For the estimation of PCDD/F emissions, emission factors were used although, due to differences in the composition of deposited waste and in combustion conditions, relatively very little data on PCDD/F

emissions from landfill fires were found. Concerning TEQ from burning of domestic and similar wastes in uncontrolled conditions, this seems to vary considerably from under 10 μg I-TEQ/Mg to over 4,000 μg I-TEQ/Mg, whereas for the purpose of the current study a value of 300 μg I-TEQ/Mg was finally used as emission factor according to UNEP (2001).

The meteorological analysis for the period of July 14-29, 2006 was conducted by the 3D meteorological model MEMO (Moussiopoulos et al., 1993). MEMO is a non-hydrostatic, prognostic mesoscale model which is used for simulating mesoscale air motion and inert pollutant dispersion, over complex terrain at the local-to-regional scale. The model domain covered an area of 48×48 km^2 using a dense grid with a 0.5 km resolution. MEMO's results were compared to the meteorological measurements of 6 nearby stations through statistical analysis of pertinent time series, in order to estimate the efficiency of the model in different hours of the day. Furthermore a statistical analysis of wind characteristics of the area was conducted, based on data from both meteorological measurements and the three-dimensional calculated wind fields, with emphasis on the local circulation. The orography dataset used in the simulation was based on the SRTM 90m database, while meteorological input data were obtained from upper air soundings in the airport area.

The simulation of PCDD/Fs dispersion in the area of study was performed using the MARS dispersion model (Moussiopoulos et al., 1995). MARS is a 3D local-to-regional scale Eulerian dispersion model for reactive species which solves atmospheric-diffusion-reaction equations. Processes of emission, dispersion, transformation and deposition of pollutants are calculated by solving a system of parabolic partial differential equations on a staggered grid in terrain-influenced co-ordinates. Meteorological input data such as wind speed in x- and y-direction as well as temperature, turbulent kinetic energy, surface roughness, Monin-Obukhov length and friction velocity were provided by MEMO.

In the current study, PCDD/Fs were considered as chemically inactive. MARS was also applied on the same 48×48 km^2 grid. Deposition of PCDD/Fs was calculated by assuming a dry deposition velocity of approximately 0.5 cm/s for PCDD/Fs (Shih et al., 2006). The simulation regarded the landfill fire as a point source which started to emit simultaneously with the outbreak of the fire, so the initial and lateral boundary conditions of MARS for PCDD/Fs where considered equal to zero. For simulating the dispersion of PCDD/Fs during the incident, the emission height was also required, as it varied depending on factors such as fire radius, plume temperature, amount of burned waste, stability conditions etc. For the estimation of the emission height, the FIREPLUME model (Brown et al., 1999) was used in this work.

2. SIMULATED WIND FIELDS

The greater area of the Tagarades site is characterized by a generally complex terrain with considerable roughness variations. The area morphology is crucial for the formulation of local circulation patterns, favoring air mass channeling phenomena and resulting to NW – SE winds towards the Vassilika valley. In case of weak synoptic conditions, more intense local circulation patterns develop, such as valley – mountain winds, as well as the sea breeze of Thermaikos bay. In essence, weak synoptic conditions that dominated during the incident favored the influence of local circulation in wind mass transfer.

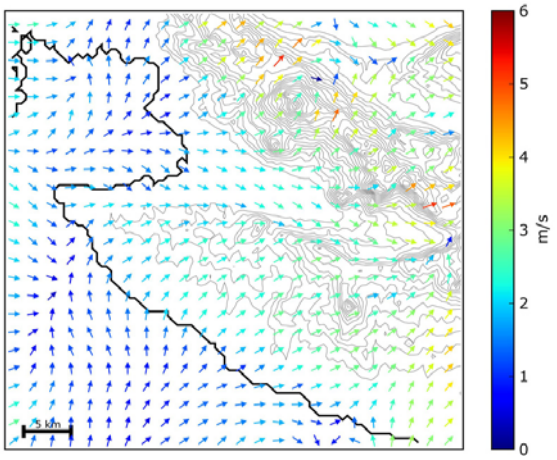

Figure 3. Wind flow simulation (July 19, 10:00 LST) by the model MEMO.

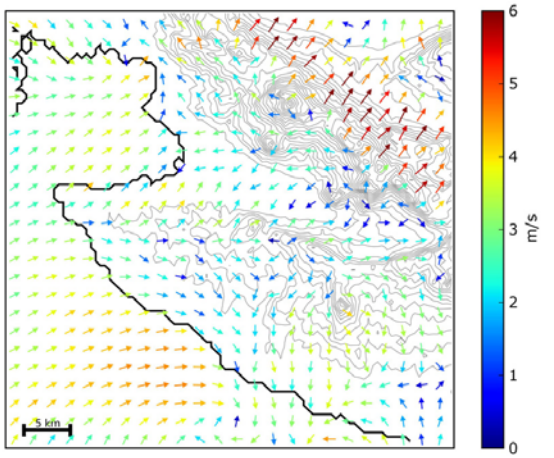

Figure 4. Wind flow simulation (July 19, 22:00 LST) by the model MEMO.

Daily fluctuation of wind flow calculated by MEMO shows that the development of sea breeze, combined with local anabatic and katabatic flows, resulted in a significant transfer of air mass to the east and southeast, with penetration of the flow to the Vassilika valley especially pronounced during midday. On the other hand,

transfer of air mass from the landfill fire towards the city of Thessaloniki was not favored in general during the period under study.
Figure 3 illustrates a representative daytime flow during the incident which indicates the sea breeze inland penetration through the Vassilika valley. Figure 4 illustrates the prominent nocturnal katabatic winds on the slopes of the area's hills and mountains. Statistical analysis of MEMO results showed that the model was able to correctly reproduce the main features of the local flows and the daily profiles of wind speed and direction at the station locations.

3. DISPERSION AND DEPOSITION OF PCDD/F

According to the calculations, during the first three days of the incident, when the fire was most intense, the emission of PCDD/Fs amounted to approximately 3 g/day. As the fire was gradually put out, these emissions declined accordingly, reaching 0.1 g/day during the last days of the incident. In all, approximately 17 g I-TEQ of PCDD/Fs were estimated having been emitted over the entire incident. Figure 5 illustrates the simulation results of PCDD/F dispersion as a map, depicting the mean values of PCDD/Fs concentration for the incident's period. It can be concluded that the areas mostly affected by the fire are located south and southeast of the fire source. In a radius smaller than 5 km from the source (which includes the populated sites of Ag. Paraskevi, Souroti, Tagarades and K. Scholari), average atmospheric concentration exceeded 150 fg I-TEQ/m^3. For the areas of Triadi, the airport, Galatista and Sozopoli, calculated concentrations were marginally larger than 20 fg I-TEQ/m^3. The mean ambient PCDD/F concentrations for selected locations of the studied area are shown in Table 1. Measurements of PCDD/F concentrations on emitted particulate matter (PM_{10}) were conducted by the Environmental Pollution Control Laboratory, Department of Chemistry of Aristotle University of Thessaloniki, for the period 22 – 28 July 2006. These indicated values of approximately 260 fg I-TEQ/m^3 for Ag.Paraskevi and 6 fg I-TEQ/m^3 for Triadi. It must be noted that there are no EU limit values for the PCDD/Fs concentrations in ambient air; nevertheless the limit value in Germany is 150 fg I-TEQ/m^3 (Laenderausschuss für Immissionsschutz, 1993).

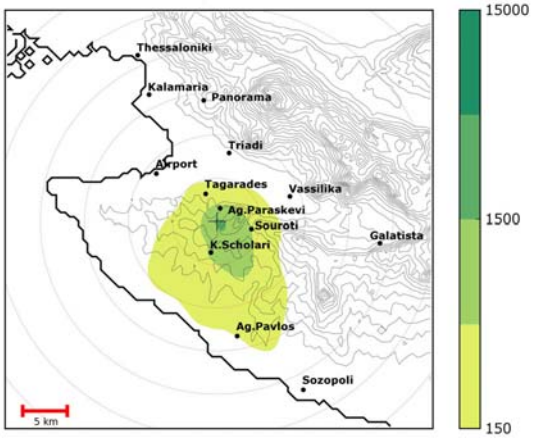

Figure 5. Mean PCDD/F ambient concentration (in fg I-TEQ/m^3) for the period of 14-29 July, 2006.

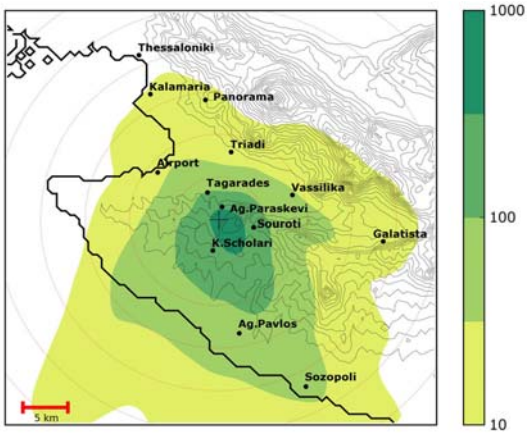

Figure 6. Average daily PCDD/F deposition (in pg I-TEQ/m^2/day) for the period of 14-29 July, 2006.

Table 1. PCDD/F ambient concentration and deposition in the period 14-29 July,2006.

Area	Mean PCDD/F ambient concentration (in fg I-TEQ/m^3)	Average daily PCDD/F deposition (in pg I-TEQ/m^2/day)
Vassilika	24	35
Triadi	22	21
Souroti	182	200
Airport	36	24
Kalamaria	9	11
Panorama	10	12
Tagarades	190	113
Ag.Paraskevi	482	267
Ag.Pavlos	17	69
K.Scholari	214	243

Average daily deposition of PCDD/Fs is illustrated in Figure 6. For Ag. Paraskevi and K. Scholari the average deposition value during the period of the incident was approximately 250 pg I-TEQ/m^2. Galatista, Triadi, the airport, Panorama and Kalamaria were less affected, as the mean values of daily deposition remained under 30 pg I-TEQ/m^2, while for the city of Thessaloniki the maximum daily deposition value was even smaller (approximately 8 pg I-TEQ/m^2). As resulted from the daily deposition maps, in 4 out of the 16 days which were studied, deposition of PCDD/Fs exceeded 10 pg I-TEQ/m^2 in the greater Thessaloniki area. Although no standards have yet been established on the deposition of dioxins, Durif (2001) recommends the aforementioned target value of 10 pg I-TEQ/m^2/day. The deposition of PCDD/Fs for selected places of the area under study is shown in Table 1. As a means to verify the total PCDD/Fs deposition calculated with MARS, the mass balance of the area under study was estimated. According to the calculations, 10% of PCDD/F mass was deposited to the ground of the study area, a result which is in accordance to pertinent studies (Lohman and Seigneur, 2001).

4. CONCLUSIONS

During the studied 2006 landfill fire at the 'Tagarades' waste disposal site, significant PCDD/F amounts were emitted to the air. According to the conducted simulation, which was based on the natural characteristics of the plume and the meteorological conditions during the landfill fire, the biggest part of the emitted PCDD/Fs was dispersed in distances outside the study area. Proportionally larger quantities were transported towards the less populated areas at the south-southeast of the site, while transfer of air mass towards the city of Thessaloniki was (thankfully) not generally favored during the incident. Air is the main distribution pathway for PCDD/Fs and according to the study, dioxin concentration reached and exceeded the recommended safety limits in areas adjacent to the site. Similarly, dioxin deposition which exceeded the recommended safety limits was calculated having occurred in an area of about 100 km^2 around the incident location.

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DETECTING CHANGES IN NO_2 AND NO_X AIR QUALITY IN VALENCIA (SPAIN)

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ABSTRACT

This paper presents the joint application of the low-pass KZ filter and regression models to analyse temporal changes of NO_2 and NO_x concentrations at two sampling sites of an urban area. The long-term fractions (periodicities greater than 1 year) represent a very small amount in the overall variability of the original time series. This component may be attributable to long-term changes in emissions (traffic) and meteorology. The seasonal and short-term components are more important fractions in the hourly air pollution series. NO hourly concentrations are used as traffic density indicators. The daily cycle of hourly data mainly depends on traffic density. The impact of traffic density on changes between days, weeks and months is higher than the effect of meteorology variability. The prediction of daily maxima using this methodology is less accurate than the prediction of hourly concentrations in a period without traffic congestion.

1. INTRODUCTION

Air quality is a field of major concern in large cities. This paper analyses several years of ambient nitrogen dioxide (NO_2) and oxides of nitrogen (NO_x) data at two monitoring sites with different traffic density. The stations are located in Valencia. The distance between them is 2.6 Km. Valencia is the third largest city in Spain and is a typical Mediterranean city in terms of its urban structure and climatology. Air pollution in Valencia mainly derives from motor vehicle exhaust emissions, with industrial pollution and heating playing a smaller part.

Recent studies have shown in this city, a significant association between an increase of 10 $\mu g/m^3$ in NO_2 pollution (24 hour mean and hour maximum) and asthma, measured as a relative risk of emergency visits (Tenias et al., 1998). Daily levels of NO_2 are also associated with cerebrovascular and cardiovascular admissions. Furthermore this pollutant is precursor of other secondary pollutants that are related to photochemical smog and acid rain.

2. METHODOLOGY

The two sampling stations are P. Silla and Viveros. At Viveros station the meteorological records were provided by the National Institute of Meteorology (wind speed ws, wind direction wd, temperature t, solar radiation sr). The Viveros geographic coordinates are W0º21'59" N39º28'50" and its altitude is 11 m. This site, located on city outskirts at the beginning of the twentieth century, have been since 1970 progressively enveloped by urban growth. The automatic station at Viveros was established in the year 2002. The P. Silla site geographic coordinates are W0º22'52" N39º28'05" and its altitude is 11 m. This site is located at a higher traffic density point due to the proximity of a motorway access. Therefore pollution changes at P. Silla are more dependent on traffic density than at Viveros. Besides pollutants concentrations, P. Silla also monitors meteorological variables (ws, wd, t, relative humidity rh, pressure p, sr). The period analysed in P. Silla is 1st January 2001- 31st July 2006 and in Viveros is 1st January 2002- 31st July 2006. Hourly and daily data are available at a web page of the local government (www.gv.es). The analyses and plots have been obtained using the language and environment R (R Development Core Team, 2006).

This study assesses hourly and daily air quality changes of NO_2 and NO_x using the low-pass Kolmogorov-Zurbenko filter ($KZ_{m,p}$) (Rao and Zurbenko, 1994; Rao et al., 1995). This method has been shown to be an effective way to analyse the temporal components and to assess the influence of meteorological conditions on atmospheric pollutants. This assessment allows to study the relative importance and the contribution of meteorological variables and emissions to pollutants variability. The hourly data set at P. Silla shows 15.33 % of missing values (hours with at least 1 component missing); on a daily basis, this percentage is 11.7%. At Viveros, the missing rates are 41.26% (hourly observations) and 44.11% (daily data). When computing daily maxima and means, each case was considered to be valid when at least 50% of the data necessary for each observation was available, otherwise it was considered a missing value. The KZ filter can be applied to data sets with missing rates up to 30% without problems (Iasonos and Zurbenko, 2002). A recent paper (Capilla, 2007) on ozone hourly data has shown that this method is quite robust to higher missing rates.

3. RESULTS AND DISCUSSION

The results of this study show that annual levels of NO_2 have exceeded the limit values for human health within the Directive of the European Union (1999/30/EC), in three years of the studied period, at the sampling station with the highest traffic density (P. Silla). The annual averages of NO_x concentrations have been above the limit value for the protection of vegetation at the two sampling sites during the whole period.

The daily cycle mainly depends on traffic variability. There are sharp NO_2 and NO_x concentration peaks associated with hours of traffic congestion. Figure 1 shows the daily cycle of NO_x at P. Silla. Similar patterns are observed for this pollutant at Viveros, and for NO_2 at both stations. The highest value of NO_x is observed between 8 and 11 a.m. During the weekends, NO_2 and NO_x concentrations at these hours are clearly lower, and their highest records are monitored between 19 and 23 hours.

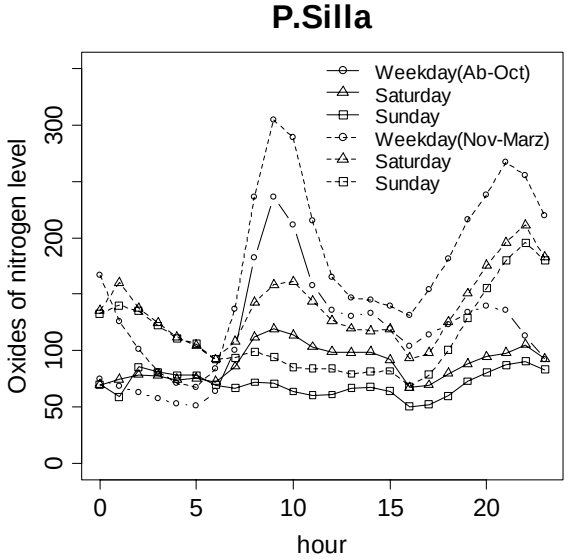


Figure 1. NO_x daily cycle at P. Silla

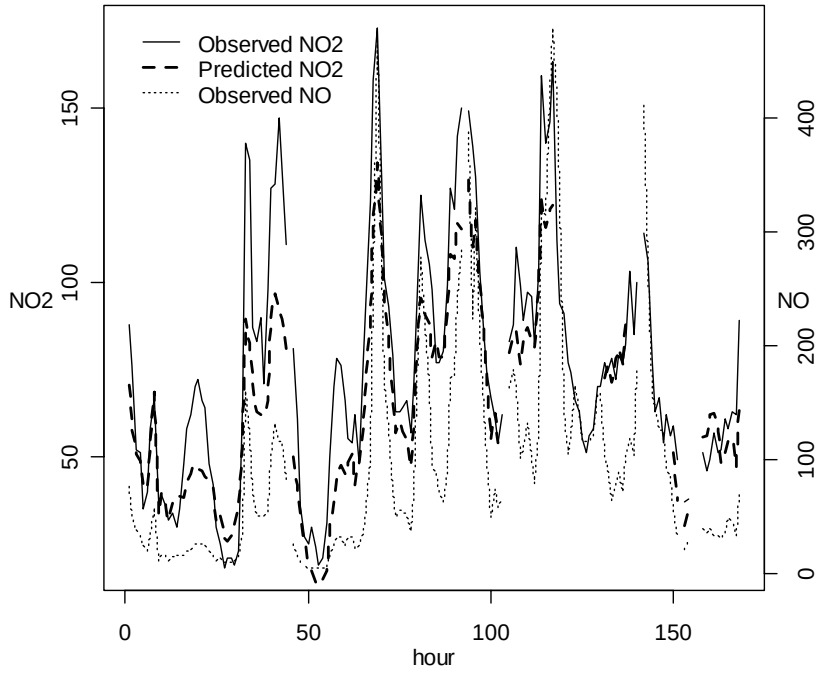
The original time series consisting of hourly averages of NO_2 and NO_x concentrations were splitted into long-term, seasonal and short-term components. The package 'kza' for R was applied (Close and Zurbenko, 2005). The long-term fractions (periodicities greater than 1 year) represent a very small amount in the overall variability of the original time series. This component may be attributable to long-term changes in emissions (traffic) and meteorology. The seasonal and short-term components are more important fractions in the hourly air pollution series. Seasonality has been analysed as periodicities between 24 hours and one year (differences between days of the week, including the weekend effect, and between months). The seasonality is more important at Viveros (45.6% for $\log(NO_2)$ and 45% for $\log(NO_x)$) than at P. Silla (42.1% for $\sqrt{NO_2}$ and 43.2% for $\log(NO_x)$). The daily cycle has a greater contribution at P. Silla (46.6% for $\sqrt{NO_2}$ and 47.3% for $\log(NO_x)$) than at Viveros (43.2% for $\log(NO_2)$ and for $\log(NO_x)$).

The meteorological effects on periodicities above 24 hours and on the daily cycle, were estimated from filtered series using stepwise regression. At P. Silla the percentage of variance of $KZ_{14,3}\sqrt{NO_2}$ explained by the effects of meteorology periodicities above 24 hours is 29.2%. This percentage is higher (40.2%) for $KZ_{14,3}\log(NO)$. For Viveros station, the variability of $KZ_{14,3}\log(NO_2)$ explained by candidate meteorological variables is 31%. The effects of changes above 24 hours in meteorology represent 29.3% of the total variability of $KZ_{14,3}\log(NO)$, and a similar percentage of the variability of $KZ_{14,3}\log(NO_x)$. Using meteorologically adjusted NO as an indicator of traffic density, a regression model was applied to approximately estimate the traffic influence on meteorologically adjusted periodicities above 24 hours in transformed NO_2 and NO_x. The percentage of variability explained by NO periodicities on pollutants changes above 24 hours is 47.3% for NO_2 and 47.5% for NO_x at P. Silla. At Viveros, this effect represents 44.5% of changes above 24 in $\log(NO_2)$ and 43.8% in $\log(NO_x)$.

Periodicities above 1 year unexplained by meteorological variables and NO represent a small fraction (range from 6.8% at P. Silla to 2.2% at Viveros). The influence of other variables not included in the analysis represents 16.7% and 9.3% of the variability of $KZ_{14,3}$ filtered pollutants at P. Silla, and 30.4% and 18.2% at

Viveros. The variability of the daily cycle explained by daily variations of meteorology is very low at both stations (ranged from 5.7% at Viveros to 11.7% at P. Silla). At P. Silla the percentage of variability of pollutants daily cycle explained by NO changes, is 52.7% for NO_2, and higher for NO_x. A similar value is obtained at Viveros (53.7% for NO_2).

Figure 2 plots the predictions (dotted line) and observations (dashed line) of NO_2 at P. Silla for the week 8th-14th December 2003 (in this year the limit value for the protection of human health was exceeded). The predictions were obtained by summing estimated values of periodicities above 24 hours and of the daily cycle. The NO observations are also plotted. The main cause of the uncertainties of the predictions might be that the study method did not include real traffic data (not available at this station, NO concentrations were used as a traffic density indicator). Therefore the predictions underestimate the NO_2 peak values and represent 59% of the total NO variability observed at P. Silla from 2002 to 31st July 2006.



The daily maximum values of these pollutants were also filtered to study periodicities above and below 31 days. Maxima changes above 1 year represent a small amount of total variability in the study period (2001-2006 at P. Silla and 2002-2006 at Viveros). The influence of meteorology on changes above 31 days (differences between months and years) and on the monthly cycle of daily maximum NO_2 is small than on the same changes of daily NO_x maxima at the two sampling sites. The candidate meteorological variables and NO concentrations (traffic indicator) jointly explain a percentage of variability of daily maximum NO_2 between 32.9% (P. Silla) to 53.06% (Viveros). These variables explain a higher percentage of variability of log-daily maximum NO_x values. The monthly cycle is less dependent on meteorology than periodicities above 31 days. However, the traffic density influence, analysed using hourly NO concentration as an indicator, is higher on this cycle than on changes above 31 days. The accuracy of daily maximum predictions is lower than the accuracy of hourly means, specially at P. Silla, where the daily cycle is more dependent on traffic density. As mentioned above, the approach makes worse predictions of peak concentrations due to the lack of traffic accurate information.

4. CONCLUSIONS

Six and five years of hourly NO_2 and NO_x have been analysed with the joint application of the low-pass KZ filter and regression models. Two sampling sites at an urban area of a Mediterranean city were considered because both pollution and meteorology data were available at them. The sites present different

traffic densities. Although there was no traffic density data at the sites, hourly NO concentrations were used as traffic indicatives.

The results of the study show that annual levels of NO_2 have exceeded the limit values for human health within the Directive of the European Union, in three years of the study period at the sampling station with the highest traffic density. NO_x limit values for the protection of vegetation have been exceeded at the two sampling sites during the whole study period.

The pollutants daily cycle are clearly dependent on traffic density. The meteorology influence on changes between days, weeks and months is higher than on changes between hours. However, the traffic influence on these changes is more important than meteorology variability effects. Hourly pollutants predictions are worse for hours with traffic congestion.

A more detailed analysis of daily maximum NO_2 and NO_x periodicities, reveals that in the study period there has not been significant maxima changes between years. The meteorological conditions variables and NO concentrations jointly explain a higher percentage of variability of daily maximum NO_2 at the site with lower traffic density. Maximum NO_2 and NO_x monthly cycles are less dependent on meteorology than maxima periodicities above 31 days. Daily maximum predictions are less accurate than hourly mean predictions, specially at the site with higher traffic density due to the lack of this type of information. Future research will concentrate on the application of alternative methodologies, on the analysis of the influence of missing rate on the performance of the methods, and on the use of real traffic data to predict pollutants concentrations.

5. ACKNOWLEDGEMENTS

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POM CHARACTERIZATION AT A PERI-URBAN SITE IN CENTRAL ITALY: THE RB-MIUR-2001 PROJECT "CHEMICAL INDEXES OF AMBIENT TOXICITY"

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Abstract: The knowledge of main composition of organic fraction associated to suspended particulates provides suitable information about nature and relative intensity of sources of ambient pollution. In that perspective, a three-year study is conducted in the metropolitan region of Rome, Italy. Measurements are made at two sites, the former semi-rural and the second in downtown.

1. Introduction:

The toxic potency of numerous organic compounds associated to aerosols is consolidated. Nevertheless, only a restricted number, overall PAH, are usually investigated in ambient air. For that concerns Italy in particular, the sole benzo(a)pyrene BaP is quoted by legislation aimed to preserve air quality. As a consequence, in Rome only measurements of this compound have been performed till now, except short campaigns conducted by our group in association with ISPESL and ISS, two Technical Departments of Italian Ministry of Health. The organic fraction of aerosols (POM) is a very complex matrix, whose composition is strictly dependent upon the nature and strength of sources. Thus, by looking the percent composition of homologues within the *n*-alkane group or the relative abundance of resolved and unresolved components of the non-polar fraction it is possible to assess the relative impact of biogenic and anthropogenic emissions, the petrogenic in particular. In order to fulfil the gap about the knowledge of POM composition and sources in the metropolitan region of Rome, a dedicated study was started in July 2005 and will be concluded in December 2007.

2. Experimental:

The study is conducted at two sites; the former, Montelibretti (RM, Central Italy) is a semi-rural area located around 30 km NE of Rome centre, while the second sampling site is the city garden of Villa Ada, assumed by the Civic Network as representative of background pollution in Rome. At Montelibretti the measurements started in July 2005, and in downtown Rome in June 2006. Groups and individual congener exploiting different polarity/neutrality properties as well as carcinogenic/mutagenic potency are investigated. Our concern is focused on *n*-alkane (C_{18}~C_{36}), PAH (from fluoranthene to dibenzopyrenes), and fatty acid (C_{12}~C_{30}) groups. In addition, two compounds (nicotine and caffeine) are monitored among "highly-polar" pollutants. Spot measurements include also Nitrated-PAH (from nitrofluorenes to nitrochrysenes) and dicarboxylic alkanoic and aromatic acids. A set of releases are expected from this study, exceeding the "environmental significance" of the crude concentration data sets. In particular:

- through the distribution patterns of *n*-alkanes and *n*-alkanoic acids, the relative impacts of anthropogenic/ biogenic sources would be discriminated, also distinguishing contributions arising from high vegetation and soil (Figure 1); at this respect, the CPI_{25} rate calculated from *n*-alkane concentrations is diagnostic, since it is equal to 1 in petrogenic emissions whilst, for vegetation-related source, it can exceed 10;
- dicarboxylic alkanoic acids as well as native phthalic acid provide information about the rate of oxidation processes occurring in charge of reactive hydrocarbons;
- PAH would provide an index of aerosol-associated toxicity, as well as further information concerning the emission source nature and the influence of atmospheric reactivity (see Figure 2);
- the two highly-polar compounds are tracers of tobacco and cooking-related sources;
- by comparing the concentrations of nitro-PAH it is possible to evaluate the relative importance of day- and night-time degradation rates of pollutants and also the specific impact of diesel vehicle emission.

Collection of suspended particulates is conducted by means of two sequential samplers, each provided of two independent, mass-flow-controlled gas lines. The former one is dedicated to collect the "pulmonary" fraction (PM_{10}), the second to "fine particles" ($PM_{2.5}$). Samples are collected daily onto inert membranes, grouped accordingly to $PM_{2.5}/PM_{10}$ mass ratio (which, over season-like periods, is strictly related to dominating meteo-climatic situation) and chemically processed to evaluate the target compounds. For this purpose, the organic particulate is extracted by refluxing in soxhlet, then fractionated by alumina column

chromatography and finally analyzed by HRGC-MSD. Organic acids require a pre-treatment, comprising conversion into propyl esters by means of propanol in the presence of BF_3.

Figure 1. GC-MS profile of non-polar aliphatic fraction (comprising *n*-alkanes) associated to airborne particulates. A: particulates collected in downtown Rome; B: sample collected at Montelibretti.

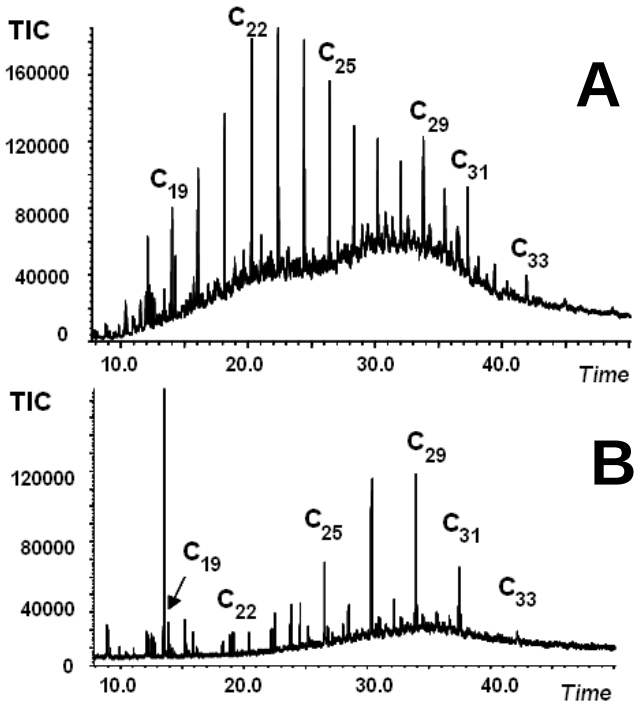
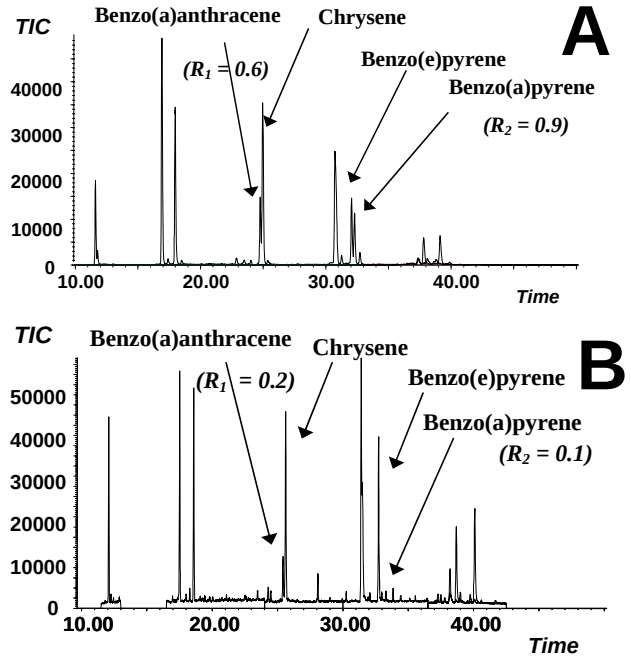


Figure 2. GC-MS profile of PAH fraction associated to airborne particulates collected in downtown Rome: A: winter sample, representative of fresh emission; B: summer sampling, aged particulate showing heavy degradation of reactive benz(a)anthracene and benzo(a)pyrene.



3. Results:

The concentrations of organic pollutants measured in the air of Montelibretti are reported in a parallel poster communication. However, they do not account for the main release of this study. Indeed, although the research is still running, however some remarkable insights could be drawn till now, e.g.:

- the *meteo-climatic approach* for grouping days and daily samples seems more promising than that based upon the year time, in the perspective of investigating sources and their relative impact;
- the Montelibretti site is poorly affected by the pollution coming from downtown Rome, whilst local *fresh* sources seem to give raise to detectable amounts of air contaminants;
- high stability conditions induce quite high concentrations of air toxicants (e.g., PAH) also in that rural area;
- both season-dependent and semi-continuous sources affect the Montelibretti site.

4. Acknowledgments:

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SITUATION AND TENDENCIES IN AIR QUALITY IN A NORTH EUROPEAN MEDIUM-SIZED TOWN

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ABSTRACT

This study is targeted at comprehensive model-based overview of air quality in Tartu, the second largest town of Estonia (100 000 inhabitants). The database of sources consists of (1) regular statistical data on industrial point sources, (2) emissions from streets, based on traffic counting and modelling results and (3) gridded emissions from domestic heating, derived bottom-up(questioning of inhabitants). The urban-scale model AEROPOL was applied to atmospheric dispersion calculations (concentrations of SO_4, NOx, CO and particulate matter (TSP)). First time this type of study was carried out in Tartu 1999. The second investigation based on emission data from 2002-2005, aims at correcting the shortcomings made in the first one (more precise and comprehensive questionnaire about domestic heating) and chasing the tendencies in air quality due to changes in the use of fuels and traffic flows.

1. INTRODUCTION

The urban air pollution is a result of complicated pattern of different emission sources. Although transport vehicles are important sources everywhere, the other emissions vary greatly with geographical location of the town. This study is focused at Tartu – the second largest town of Estonia (100 thousand inhabitants). The results presumably have more general value, because Tartu is a rather typical medium-sized town for the countries with transitional economy in the northern part of Europe. Study by Houthuijsa et al. (2001) at that kind of East-European cities has shown high levels of pollutants with large changes between seasons, caused by local heating.

The main factor affecting the air quality in most cities – street traffic has been dramatically increasing in nineties and slight tendency of growth still exists. The street network was not designed for so heavy traffic, thus traffic jams are frequent. On the other hand, the technical improvement of cars prevents severe worsening of air quality due to transportation – only slight increase of NOx concentrations is observed near the main streets.

As Tartu is not an industrial town, the industrial emissions have never been a big problem there. Situation has even improved during recent 15 years, because small coal-fired boiler houses are replaced with a few large ones burning natural gas, wood and peat. Due to high stacks and good purification their impact on local air quality is marginal.

Emissions from local heating are typical for the region. About 1/3 of people are living in residential areas without central heating. Besides wood, natural gas and light fuel oil a remarkable amount of coal is still used for heating of private houses. Dense dwelling areas with multi-flat wooden houses built in the 19th and beginning of the 20th century are situated close to the town centre. Most of these houses still have stove heating, resulting in high emissions of CO and particulate matter per area.

Earlier comprehensive model-based study of air pollution in Tartu, including all the sources mentioned above (base year 1998), was carried out by Kimmel & Kaasik (2003). Besides general success, two problems were addressed: (1) traffic counting was relatively scarce, leaving the total emission from streets uncertain; (2) it was difficult to estimate the peak levels of emissions from local heating due to unknown simultaneity factor of heating the furnaces. This study is aimed to clarify these drawbacks, as well as to find out the tendencies in emissions and resulting airborne concentrations during recent years.

2. METHODOLOGY

The general methodological principle of the emission inventory is the bottom-up approach. Although the top-down approach is easier usually and includes fewer uncertainties, it can hardly be applicable in this study. First, high urban-scale spatial resolution requires the same resolution of emission data. Second, it is hard to follow the consumption of fuels for residential heating in top-down manner. That concerns especially the wood, because private forest owners sell a large part of it directly to the residents.
The emission inventory of industrial (incl. central heating) sources was based on governmental database of air pollution sources (reference year 2002).

The street emission data are based on the traffic counting and modelling by Stratum AS. Rush hour emissions are estimated from traffic flow and speed, applying the formulae by Finnish Meteorological Institute (Härkönen et al., 2001, Karppinen et al., 2000). The yearly average traffic flows were estimated, dividing the

afternoon rush hour (4-5 p.m.) ones with coefficient 2.4 that was recommended by the Nordic Council of Ministers, 1984 and validated by authors for randomly selected streets of Tallinn (standard deviation 10%, systematic difference only 2%).

The inventory of local heating emissions is based on the questioning of inhabitants carried out in 2004. 200 households were randomly selected out of a several thousands. Data about yearly consumption and maximal hourly consumption of fuels, nominal capacity of heating devices (if available) and typical times of heating were collected. The questionnaires were grouped by the dwelling areas (32 areas in total), amounts of used fuels were scaled with the numbers of inhabitants in the areas, getting the estimation for fuel consumption in each area as a result.

It was found out that typical time of heating of a wood-fired stove is about 2 – 3 hours (once or twice per day). The domestic boilers for heating are used for longer time, but not permanently, either. To avoid the overestimation of peak-level emissions from domestic heating due to non-simultaneous heating of furnaces, a coefficient of simultaneity was derived. Assuming the Gaussian distribution of heating times near the most typical one, the influence of partial overlapping was approximated with a multiplicative coefficient *S* that was calculated as

$$S = \frac{\Delta t}{\sqrt{\pi / 2}\sigma + \Delta t},$$

where σ is the standard deviation of the distribution of heating times and Δt is the average heating time.
The emissions from domestic heating were gridded, assuming uniform distribution within each dwelling area. The dispersion calculations were made with the AEROPOL model. AEROPOL is a Gaussian plume model, enabling point, line and gridded area sources (main features, see Kaasik & Kimmel, 2004). The computations were performed at 9×7 km grid with resolution 40 m. To calculate the yearly average concentrations, the local wind rose and long-term climatic average weather conditions were applied. To estimate the maximal concentrations, the worst dispersion conditions – ground-based inversion and 10 m wind velocity 0.5 m/s – were applied.

3. RESULTS AND DISCUSSION

3.1. Emission Inventory

The simultaneity coefficient for domestic heating was found 0.43 as average for the town, varying from 0.31 to 0.53 for different dwelling areas. Thus, the effect of non-simultaneity of heating to the top-level emissions is very significant. According to the questionnaire, the most intense heating occurs during cold wintertime in workdays from 4 to 7 p.m. The afternoon rush hour of traffic fits exactly within the same time. According to that, the highest emissions from boiler houses occur in cold weather as well. Thus, the maximal short-term emission from the town must be nearly equal to the sum of peak level emissions from industrial enterprises (mainly central heating), domestic heating and transportation vehicles. The overview of total emissions is given in Tables 1 and 2. An example of gridded emission data for the AEROPOL model is given at Figure 1.

Table 1. Yearly emissions from Tartu, tonnes per year

	SO_2	NO_X	CO	PM
Industrial	31	16	34	220
Transportation	-	418	1436	8*
Domestic heating	61	124	1018	1326
TOTAL	92	558	2488	1554

* only exhaust particles

Table 2. Maximal peak-level emissions from Tartu, g/s

	SO_2	NO_X	CO	PM
Industrial	139	78	310	60
Transportation	-	32	109	1*
Local heating	13	18	112	204
TOTAL	152	128	532	265

* only exhaust particles

The total emissions of all studied pollutants have decreased since 1998 (see Kimmel & Kaasik, 2003). The most dramatic, nearly seven-fold decrease occurred with sulphur dioxide emissions. The main reason is the

transition of central heating from coal and heavy fuel oil to wood, peat and natural gas. Although the same tendency occurs partially in domestic heating, the sulphur dioxide emissions from private furnaces are dominating now. Transportation has retained its leadership in NOx and CO emissions. Domestic heating is increasing in its share in particulate matter emissions, as purification of industrial emissions is becoming more efficient.

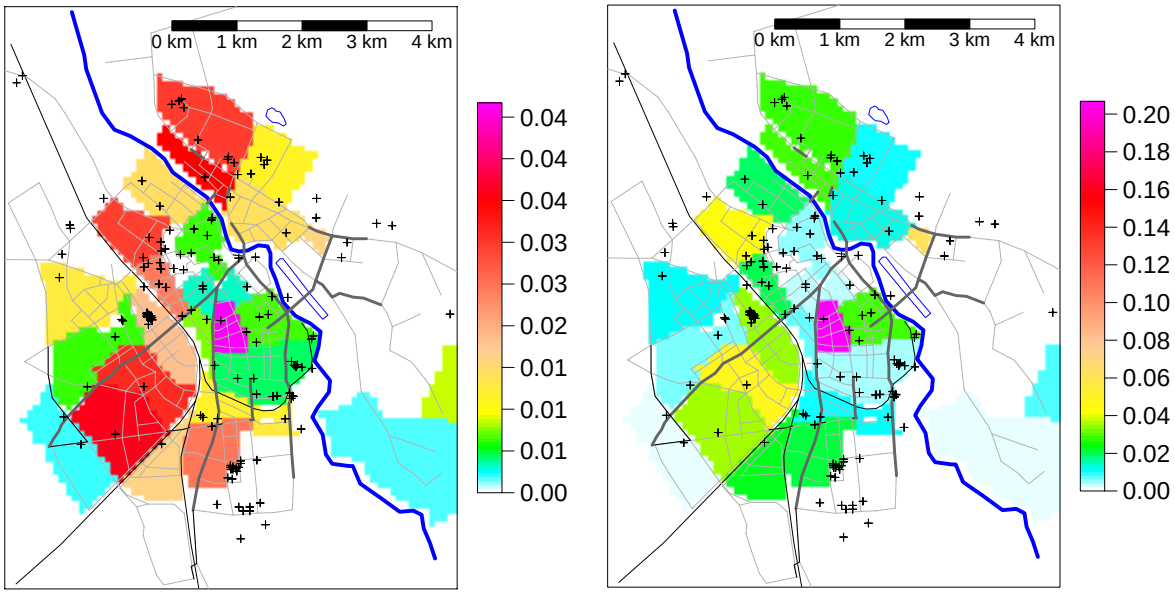

Figure 1. Point sources of air pollution emissions (black crosses), main traffic streets (more than 1000 vehicles/h at rush hour, heavy dark-grey lines) and local streets (light grey lines). Colours indicate the yearly average emissions from domestic heating (g/s per ha): particulate matter (left) and carbon monoxide (right).

3.2. Modelled Concentrations

The model calculations show that, in agreement with its share in the emissions (Table 1), the emissions from transportation are heavily prevailing in the distribution of nitrogen oxides and also dominating in the distribution of carbon monoxide (Figure 2). It must be kept in mind, however, that in the case of vehicles only exhaust emissions from the engines are included. The bigger and greatly varying emissions from dry street surface can easily make concentrations much higher (Brunekreef & Forsberg, 2005), as measured near main crossings in Tartu.

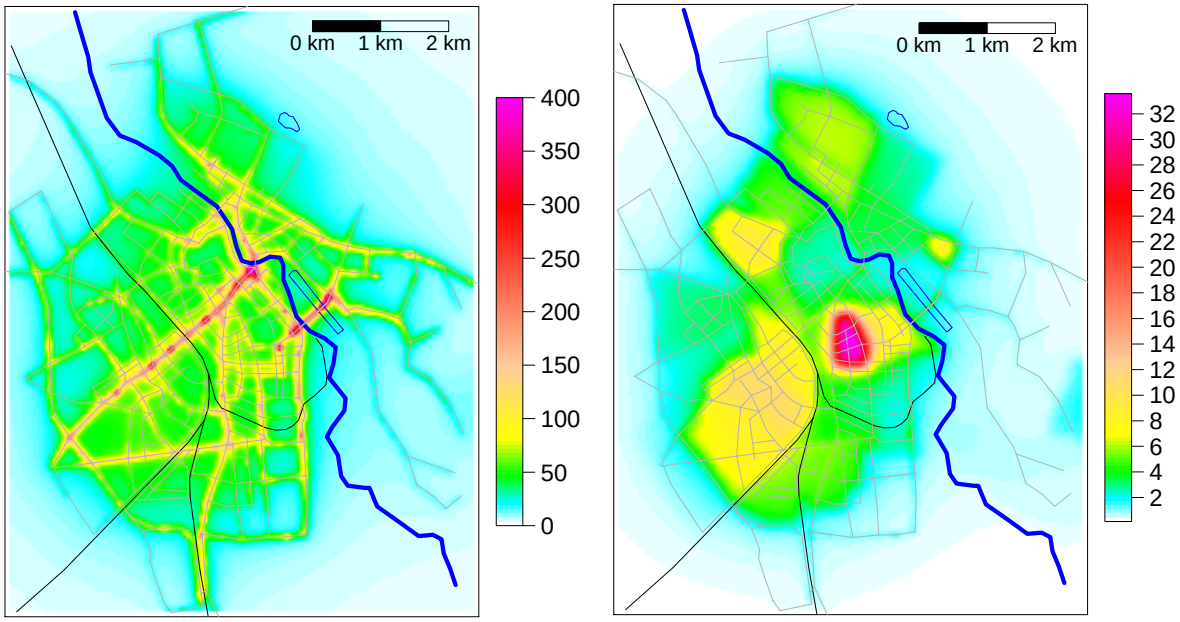

Figure 2. Modelled yearly average concentrations of carbon monoxide (left) and particulate matter (right).

The concentrations of NOx and CO, mainly induced by street transportation, are found nearly stable during last 8 years (see Kimmel & Kaasik, 2003). The concentrations of particulate matter and SO_2 remain to be

induced mainly by domestic heating, but we cannot evaluate the tendencies in strict quantitative terms, because the assessment methods have been improved much in comparison with earlier study. The model calculation suggests that the yearly average limit value of PM10 (20 $\mu g/m^3$) may be exceeded in a small densely built-up area near the city centre (see Figure 2). Some uncertainty exists on this question, because the fraction of PM10 in the TSP emissions is not known exactly.

4. CONCLUSIONS

It was found that the economic transition and restriction of environmental norms is improving the air quality in a Northeast-European town in general. In this region the moderate transition to wood heating, both in private cottages and boiler houses, is a progressive tendency, because more polluting fuels (coal, heavy fuel oil) are replaced and local forest stands are sufficient for permanent supply. In some densely built-up areas the domestic heating may cause problematic situations in respect to normative limits of PM10.

The emissions and resulting pollution levels of NOx and CO from street transport are nearly stable despite the excessive increase of the number of private cars, because of catalytic converters etc. technical improvements. It is expected that in close future the air quality near main traffic streets stay stable or will even improve, because the number of cars per capita is reaching the level of most developed industrial countries.

This study has demonstrated the reasonability of bottom-up approach in the emission inventory aimed at dispersion modelling of urban pollution.

5. ACKNOWLEDGEMENTS

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ASSESSMENT OF THE SPATIAL VARIABILITY OF AMBIENT NO_2 CONCENTRATIONS WITHIN THE URBAN AREA OF ATHENS

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ABSTRACT

Three techniques for spatial interpolation, namely inverse distance weighting, radial basis function and kriging, were applied to data from the network of ambient monitoring stations of the Hellenic Ministry for the Environment, Physical Planning and Public Works. The performance of these methods was evaluated using mean annual data for 2002. The kriging and the radial basis function techniques gave a better mapping of the concentrations, with the kriging technique providing a somewhat smoother and more conservative representation of the concentration distribution. Using these interpolation techniques the location and coverage of the monitoring stations of the Athens network was examined. Finally, the spatiotemporal characteristics of an air pollution episode observed in the winter of 2002 were also investigated, using air pollution and meteorological data together with the kriging interpolation procedure.

1. INTRODUCTION

Several studies have shown that air pollution levels observed in urban areas, where many sources of airborne pollutants are concentrated, have a significant impact on human health. In order to monitor the levels of atmospheric pollution in urban areas of Europe and to assess the attainment of the Air Quality Standards set by the EU, monitoring networks have been developed in major European cities. However, concentrations of atmospheric pollutants exhibit spatial variability within urban conurbations, resulting to different levels of exposure among the population. Concentration measurements of air pollutants are usually conducted at a limited number of ambient monitoring sites at each metropolitan area and a monitoring network is not possible to cover the whole urban setting. In this respect, interpolation techniques can constitute a useful tool for optimally locating a network of air pollution monitoring stations and for assessing exposure at different locations within an urban area in support of pollutant exposure models (Hopkins et al., 1999).

Risk assessment studies have shown that both short and long term exposure to NO_2 can induce effects to the human health and that given the role of NO_2 as a precursor of other pollutants and as a marker of traffic related pollution, there should be benefits for the public health from keeping low NO_2 levels in the atmospheric air (European Commission, 1997). The First Daughter Directive of the EU Framework Directive on Air Quality Assessment and Management sets limit values for hourly and annual mean concentrations of NO_2, to be achieved by 1 January 2010. The annual NO_2 standard is established at 40 μg m^{-3}, while the hourly standard is set at 200 μg m^{-3}, as a value not to be exceeded more than 18 times per calendar year. The high traffic density in the Athens area has resulted in exceedances of both NO_2 air quality standards (e.g. Mavroidis et al, 2006).

The main purpose of this study is to use monitoring network data to apply and evaluate spatial interpolation procedures to assess NO_2 concentrations at different locations within the urban area of Athens. The study focuses initially on the mean annual NO_2 concentrations for 2002 in order to examine the NO_2 distribution in the Athens area and to evaluate different spatial interpolation techniques. Then, a selected episode period during the winter of 2002 is examined, when high NO_2 concentrations were recorded in the Athens area. Using monitoring network data together with the kriging interpolation technique, the spatial and temporal characteristics of the episode are examined.

2. METHODOLOGY

The Hellenic Ministry for the Environment, Physical Planning and Public Works (YPEHODE) has developed in the mid-80's a network of ambient monitoring stations, to cover the need for a systematic analysis of meteorological and air quality observations from the Athens area. Each monitoring station is measuring certain criteria air pollutant concentrations and meteorological data at different characteristic locations within the urban conurbation of Athens. Nitrogen oxides concentrations are measured using a HORIBA APNA-360 analyzer, which provides NO, NOx and NO_2 concentrations, the latter by subtracting electronically NO from NOx. In the examined area 16 such monitoring stations were in operation in 2002 (Figure 1a).

Initially, mean annual NO_2 concentrations for 2002 are examined, which is a year that a complete set of good quality data is available from the Athens monitoring network. Three different interpolation procedures are applied to predict mean annual NO_2 data between monitoring stations in the Athens urban area, using the Surfer v.7 surface mapping software. These techniques are:
a) *Inverse distance weighting:* this interpolation technique incorporates the spatial variation of the data set in calculating the interpolation estimates. The interpolating surface is a weighted average of the data points and the weight assigned to each data point diminishes as the distance from the interpolation point to the data point increases. A power parameter (set equal to 2 in the present case) controls how the weighting factors drop off as distance from a data point increases.
b) *Kriging:* this is a geostatistical gridding method that attempts to express trends that are suggested in the data. It is an optimizing interpolation technique and can be custom fit to a data set by specifying the appropriate variogram model. It incorporates anisotropy and underlying trends in an efficient and natural manner.
c) *Radial Basis Functions:* these are a diverse group of data interpolation methods. The Multiquadric method is used here, since it is often considered to be the best method in terms of the ability to fit the data and to produce a smooth surface. The functions that can be specified are analogous to variograms in Kriging. These functions define the optimal set of weights to apply to the data points when interpolating a grid node.

In the second part of the paper an NO_2 air pollution episode is identified, covering the period 29-31 January 2002. The spatial and temporal characteristics of the episode are examined using air quality monitoring data together with the kriging interpolation technique.

3. RESULTS AND DISCUSSION

The Athens monitoring network includes both stations located in the inner urban centre of Athens and Pireaus that are affected by busy traffic arteries located in their vicinity, as well as stations located further away from major roads of the inner urban circle, and therefore tend to represent the pollution climate over larger residential areas where the population is exposed to pollutant concentrations. Since the present paper aims mainly at investigating the spatial variability of the NO_2 concentrations in the residential urban area of Athens, the monitoring stations located at "hot spots" are excluded from any further analysis. These include four stations, namely Patisiwn, Aristotelous, Athinas and Piraeus – 1, noted respectively as "ΠΑΤ", "ΑΡΙ " , "ΑΘΗ" and "ΠΕΙ-1" in Figure 1a.

Using the 2002 mean annual NO_2 data from the remaining 12 monitoring stations, the three interpolation techniques are applied and the results are presented in Figure 1b, 1c and 1d for kriging, radial basis function and inverse distance weighing respectively. The results suggest that the kriging and radial basis function techniques present a smoother and better distributed map of the NO_2 concentrations over the Athens area, while the inverse distance weighing technique generates "bull's-eyes" surrounding the position of observations within the mapped area. Furthermore, the kriging technique appears to provide a somewhat smoother and more conservative representation of the concentration distribution over the Athens area. Since these distributions influence substantial segments of the population, more specific examination of the interpolation techniques performance could include the use of error-related statistical methodologies. The contours from the kriging and radial basis function techniques also suggest that higher concentrations are observed on an "axis" moving from south west to north east, which is indeed a characteristic of the atmospheric pollution in the Athens area, related to the prevailing winds and the topography of the Athens basin. Furthermore, very low concentrations are mapped in the east / northeast sector. This is due to the small number of monitoring stations located in this sector, together with the fact that these stations monitor very low concentrations. It is therefore suggested that more stations are probably required to satisfactorily cover this area and that the existing stations should be more appropriately located, in order to monitor population exposure representative of the greater residential area covered by these stations. Finally, the contours suggest that when the traffic affected stations of the inner urban circle are not included in the mapping, there are still quite large residential areas where the mean annual EU standard for NO_2 is exceeded.

The spatial and temporal characteristics of an air pollution episode during the winter of 2002 is also examined. For this episode period, data are not available for two of the monitoring stations used for the examination of the annual concentrations, namely Pireaus-2 ("ΠΕΙ-2") and Thrakomakedones ("ΘΡΑ"). Figure 2a shows the development of this episode during the period 29-31/1/2002 in four of the examined stations (for reasons of economy), while Table 1 shows the main characteristics of the episode at each of the 10 stations, for the two days when it was more intense (30 and 31 January 2002).

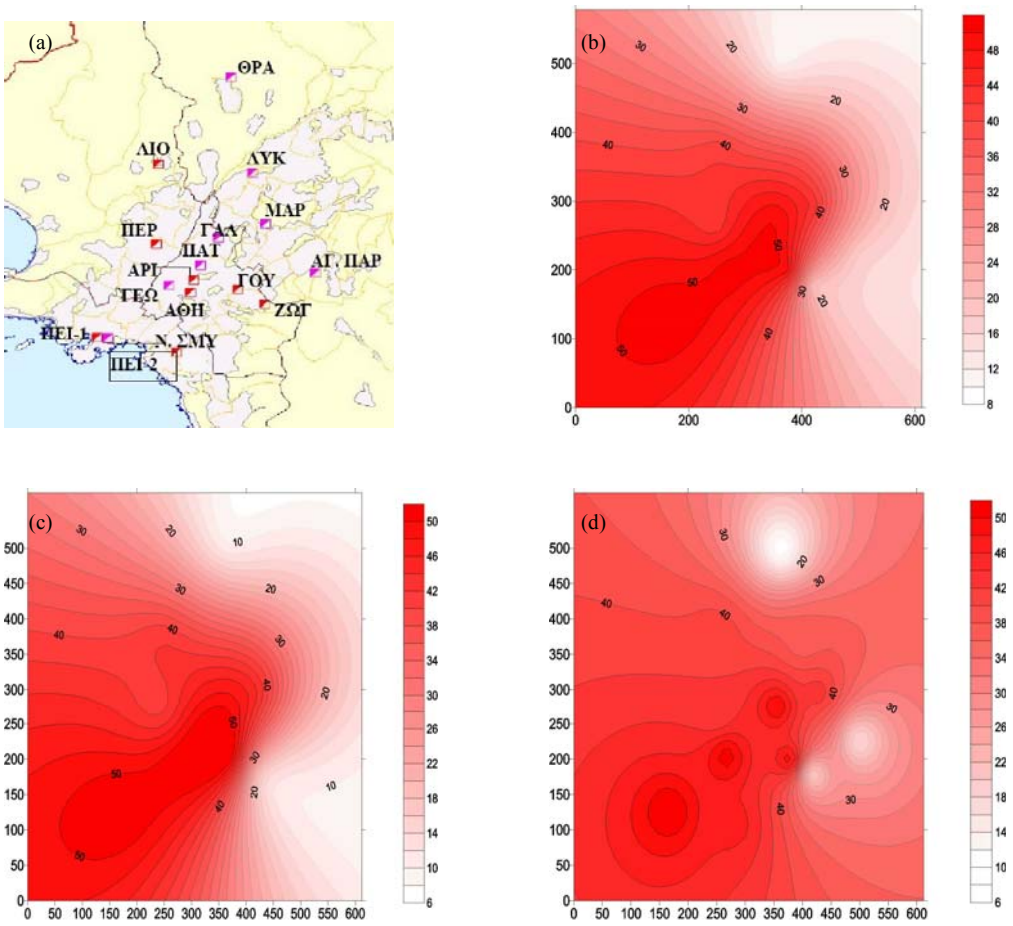

Figure 1. (a) Map of the Athens area with the monitoring sites. (b), (c) and (d) Contours of annual mean NO_2 data for 2002 using the kriging, radial basis function and inverse distance weighing interpolation techniques respectively.

Table 1. Characteristics of the air pollution episode

Monitoring station	Date of maximum	Time of peak concentration/day		No. of 200 μg m^{-3} exceedances	
		30/1/2002	31/1/2002	30/1/2002	31/1/2002
Peristeri - ΠΕΡ	30/1	13:00	13:00	2	1
Marousi - ΜΑΡ	31/1	11:00	13:00	1	2
Geoponiki - ΓΕΩ	30/1	11:00	11:00	1	2
N.Smirni - Ν.ΣΜΥ	31/1	10:00	13:00	0	1
Liosia - ΛΙΟ	31/1	9:00	9:00	1	3
Lykovrisi - ΛΥΚ	30/1	12:00	13:00	1	0
Zografou - ΖΩΓ	31/1	10:00	15:00	0	0
Galatsi - ΓΑΛ	31/1	11:00	12:00	1	2
Goudi - ΓΟΥ	31/1	10:00	15:00	0	0
Ag.Paraskevi - ΑΓ.ΠΑΡ	31/1	10:00	13:00	0	0

Figure 2 (b), (c) and (d) shows the NO_2 contours using the kriging technique at three characteristic hours of the episode (6:00-7:00 on 30/1/02, 10:00-11:00 on 30/1/02 and 12:00-13:00 on 31/1/02, the first representing an onset hour of the phenomenon and the last two representing hours of maximum concentrations at each day) in order to provide the spatiotemporal development of the NO_2 concentrations in Athens during this episode.

The contours suggest that higher concentrations are observed on a central "axis" located from south west to north east, similar to the case of mean annual concentrations, with the maximum concentrations moving on this axis as the episode proceeds. This is in agreement with the meteorological data, since the wind direction for both days at the peak concentration hours (10:00 to 16:00) ranges between 200 and 250 degrees. Another observation is that at the earlier hour of the 30th January, where concentrations have not yet increased significantly, concentrations are distributed in a smoother pattern over the Athens area. However, at the hours when maximum concentrations are observed (10:00-11:00 on 30/1/02 and 12:00-13:00 on 31/1/02), and especially at the second day of the episode (Figure 4d), concentrations are less evenly distributed and there are sites where clear maxima are observed.

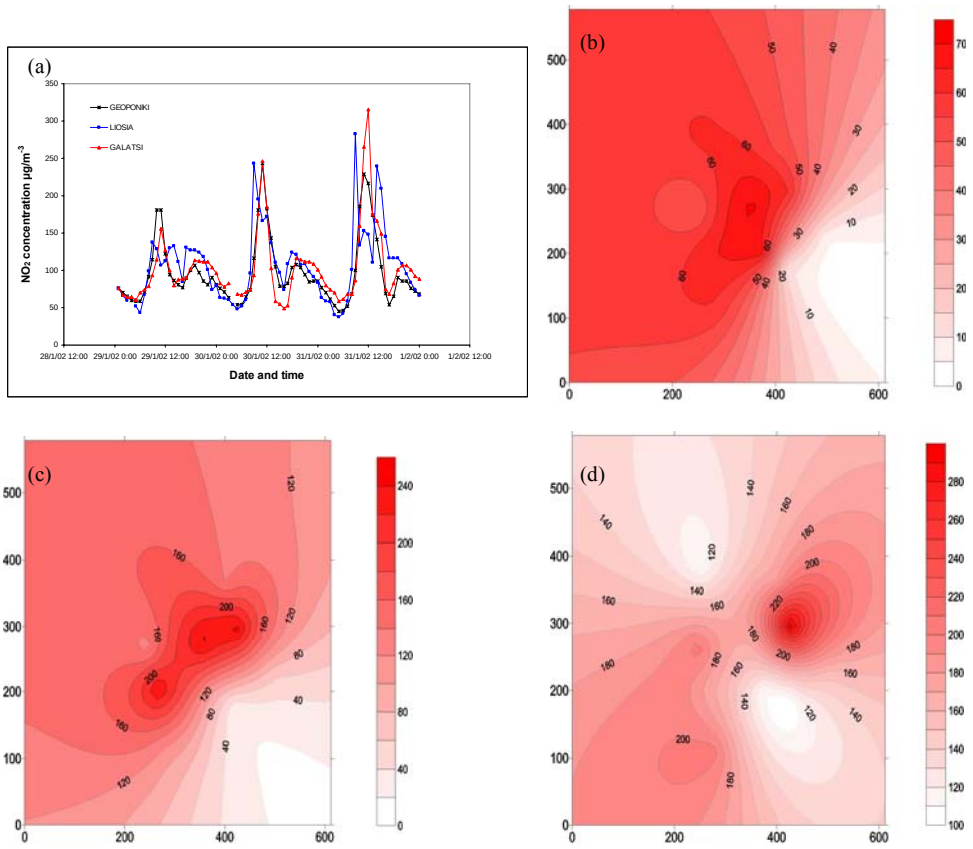

Figure 2. (a) Development of the air pollution episode at 3 monitoring stations. (b), (c) and (d) Contours of hourly NO_2 data for the hours 7:00 on 30/1/02, 11:00 on 30/1/02 and 13:00 on 31/1/02 respectively.

4. CONCLUSIONS

The kriging and the radial basis function techniques performed satisfactorily in mapping NO_2 concentration distributions over the Athens area, while the inverse distance weighing technique generated "bull's-eyes" surrounding the position of data points. Furthermore, the mapping results showed that higher concentrations are observed on an "axis" moving from south west to the north east, while more and appropriately located stations would be needed to satisfactorily cover the eastern / north eastern - part of the urban area of Athens. The kriging technique was also applied using hourly concentrations and provided some indication of the spatiotemporal characteristics of an air pollution episode on a small time scale. It should be noted though that mean annual concentrations have a much more statistically stable nature than hourly concentrations, especially for NO_2 which presents increased spatiotemporal variations. In this respect, it may be useful to examine more episodes – and more hours during an episode - of increased NO_2 concentrations in future work.

5. ACKNOWLEDGEMENTS

The authors wish to acknowledge the Hellenic Ministry for the Environment, Physical Planning and Public Works for providing the data from the network of ambient monitoring stations.

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AMBIENT AIR QUALITY OVER THE INDUSTRIAL CITY OF YANBU (SAUDI ARABIA)

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The ambient air quality assessed in Yanbu was marked with an increasing concentrations trend. The sulfur dioxide had yearly increase of 4.0ug/M3. The lowest annual average was recorded (10.2 ug/M^3) in 1986; whereas the highest (95.8 ug/M^3) in 1997. The highest daily average (254.4ug/M^3) was in 1993. The percentage frequency distribution indicated a clear shift of the mode from the low range of (26 to 39ug/M^3) in 1984 to (52 to 65ug/M^3) in 1992 and (65 to 79ug/M^3) in 1996, means a clear increase in modes with years. The nitrogen oxides had increased by three-fold, with less than 17ug/M^3 in 1985 and with 60ug/M^3 in 1988. The highest daily average concentration of NO_x (215.6ug/M^3) was in 1998, with the hourly average reaching to 729.3ug/M^3 in 1986. The annual average of ozone had any trend in the last 20 years. The current yearly average of CO is 0.6mg/M3 with the highest daily average (12.2mg/M^3) in 1994 and with the highest hourly average (16.0 mg/M^3) in 1998. The increase in the sulfur dioxide levels was attributed to the increased industrial activity of YANBU such as the installation of new plants, increased capacity of the refineries and an increase in the unregulated sources. A significant proportion NO_x emissions was from boilers and heaters of 29 MW capacity. The background data was successfully used in revising air quality standards and for prediction of the ground level concentrations.

1. INTRODUCTION.

The environmental regulations of Yanbu were developed, by incorporating the salient features of US Environmental Protection Agency (EPA) federal regulations (40 CFR), as applicable for the local needs of Yanbu. The increase of industrial activity had inadvertently increased the amount of pollution loads emitted into the Yanbu atmosphere. The Royal Commission had developed a network of ambient air quality stations to monitor and assess the ambient air quality (Anon, 1991). At present the air quality is being monitored at four locations, with continuous analyzers for Sulfur dioxide (SO_2), Oxides of nitrogen (NO_x), Carbon Monoxide (CO) and Ozone. The Air Quality Monitoring Station (AQMS) #1 is located between the industrial area and the residential area. During the winds from the south-south-west and east directions, this station can monitor the combined pollution load from the industrial area to the community area. During the predominant westerly winds this station can record the background concentration from the residential area. The Air Quality Monitoring Station #2 is located near Luberef-II plant, on the side of Yanbu-Jeddah highway. This location is suitable for recording the combined effect of all industries on the air quality, during the predominant westerly winds. Due to its close proximity to highway traffic, the air quality at this station may record the vehicular pollution also. The Air Quality Monitoring Station #4 is mainly a background/control station. It is located at a residential area upwind to the major sources. Localized sources or long-range transport of pollutants from industries could affect the ambient air quality of this station. The Air Quality Monitoring Station #5 is located at the southern part of the city, between NGL plant and Red Sea. The terrain is open and flat. The ambient air quality data collected from this station is an indicator of the emission magnitude at Yanbu.

2. DATA COLLECTION METHODOLOGY

All the ambient air quality-monitoring stations were provided with continuous monitoring equipment from M/s. Monitor Labs, USA . The fluorescence property of SO_2 on exposure to UV radiation is utilized for the detection of SO_2 in UV Fluorescence Sulfur Dioxide analyzer. This instrument can monitor SO_2 up to 10ppm with a lower detection limit of 0.001ppm and time lag of 20 seconds. The property of NO during reaction with ozone is made use in Chemi-luminescence Oxides of Nitrogen analyzer. The instrument measures the concentrations of NO, NO_2 and NO_x simultaneously. The instrument can monitor the NO_x up to 10ppm with a lower detection limit of 2ppb and with lag time 10 seconds. The ozone levels were monitored with Dacibi ozone analyzers. This instrument can measure ozone up to 1 PPM with an accuracy of 3%.

3. RESULTS AND DISCUSSION

3.1 Ambient Air Quality

Stringent enforcement of regulations and proper planning of development strategies are essential for arresting this increasing trend of pollution in the area. The concentration levels of most of the

pollutants in Yanbu had increased considerably. The sulfur dioxide concentration in YANBU had increased about three times at all the stations. The lowest annual average concentration was recorded at station #4 (10.2 ug/M^3) in 1986 whereas the highest annual average was recorded at station #5 (95.8 ug/M^3) in 1997. The concentration level of Sulfur dioxide is not showing any changes with month at any of the four stations.

Table-1: Highest daily and hourly average concentrations of Sulfur dioxide

St	Daily Maximum ug/M^3	Jul. day and year of daily max.	Hourly Maximum ug/M^3
1	229.9	233, 1991	515.6
2	254.4	23, 1993	635.6
4	152.5	250, 1998	474.7
5	223.5	251, 1997	534.7

The daily maximum concentration had attained the highest level (254.4ug/M^3) at station #2 in 1993. The hourly average concentration was also recorded the highest level at station #2 in 1998. The yearly average of sulfur dioxide concentration level had increased to the present level (70ug/M^3) from a concentration level of <30ug/M^3 in 80's. The growth rate of sulfur dioxide levels at Yanbu is about 4.0ug/M3 per annum. Obviously the increase in the sulfur dioxide levels was due to the increased industrial activity of Yanbu. Installation of new plants like LUBEREF, increased capacity of the refineries (SAMREF and Yanbu Refinery) and the increase in the unregulated sources, which were using fuel oil, were the contributing factors for the increased sulfur dioxide emissions. The sulfur dioxide emitted to the atmosphere was mainly from the combustion of fuels containing sulfur, catalyst regeneration and incineration of hydrogen sulfide. The major sources of SO_2 in the YANBU area were Samref, RC Power plant, and Yanbu refinery. The estimated emission of sulfur dioxide from these facilities were [Samref: Boilers and heaters: 28.0, FCC Unit : 63.5 , Sulfur plant: 38.1 , Crude stack : 8.7 , Total =138.3 Tons/day] ; [Yanbu Refinery: Boilers and heaters: 3.4 , H_2S Incinerator 5.2 , Total = 8.6 Tons/day]; [RC Power plant : STGs31. 7 , GTGs: 8.2 , Total =39.9 Tons/day]. The contribution from other sources of sulfur oxides is minor in comparison with these major sources.

3.2 Nitrogen Oxides

Ninety percent of all NO_x emissions come from furnaces and combustion processes. Most of the NO_x emissions are in the form of Nitrogen monoxide (NO) and out of which about 80% is transformed into nitrogen dioxide in the atmosphere. Nitrogen oxides also have an important role in the formation of ozone. In Yanbu, the nitrogen oxide levels had increased during the last 15-years by about three-fold. The nitrogen oxide levels were less than 20ug/M^3 in 1984-85; which had increased to the present level of about 60ug/M^3. The yearly average concentration of nitrogen oxides had recorded the lowest value of 16.1ug/M^3 in 1985 and the highest value of 65.4ug/M^3 in 1997. The concentrations were little higher during winter than in summer. The highest daily maximum concentration of NO_x (215.6ug/M^3) was recorded at station #2 in 1998. The hourly average concentration of NO_x had reached a level of 729.3ug/M^3 (660ug/M^3 is the RC standard) during the year 1986. Since RC standard allows two exceedances in every 30 days, the above recorded concentration level was not considered as a violation of RC standards. The highest of the daily average and the hourly average recorded at the four stations were given in Table-2. Out of the total NO_x emissions, a significant proportion is expected from sources, which were not falling under scope of environmental regulations. The boilers and heaters with capacity less than 29 MW were not regulated.

Table-2: Highest daily and hourly average concentrations of Nitrogen Oxides

St	Daily Maximum ug/M^3	Jul. day and year of daily max.	Hourly Maximum ug/M^3
1	140.3	70, 1995	729.3
2	215.6	8, 1998	580.0
4	174.8	219, 1998	321.1
5	170.3	23, 1998	481.5

The major sources of NO_x in the region were from the following facilities: Power plant:12.0Tons/day, Samref: 7.6 Tons/day, Yanbu Refinery 10.1 Tons/day, Yanpet 5.8 Tons/day

3.3 Ozone

The concentration levels of ozone had not undergone any significant changes in YANBU area. This might be due to nitrogen oxides and reactive hydrocarbons availability in the lowest ambient air, prior to industrialization due to high vehicular movements. The ozone concentration levels had crossed the RC standard level of 235 ug/M^3 on many occasions, especially in the hot summer months. The annual average concentration level of ozone was not showing any trend. The highest hourly average of ozone recorded so far is 394.6ug/M^3 at station #5 in 1993. The highest daily maximum of ozone recorded (205.9ug/M^3) at station #1 in 1992. The highest of the daily average and the hourly average of ozone recorded at the four stations are given in Table-3.

Table-3: Highest daily and hourly average concentrations of Ozone

St	Daily Maximum ug/M^3	Jul. day and year of daily max.	Hourly Maximum ug/M^3
1	205.9	133, 1992	277.4
2	201.0	209, 1995	331.6
4	146.9	216, 1995	217.3
5	190.9	73, 1998	394.6

Restricting the emission of nitrogen oxides and hydrocarbon into atmosphere can control the formation of ozone in the ambient air. The hydrocarbons are entered into the atmosphere through natural sources such as swamps, marshes or other water bodies, which generate methane. The petroleum handling operations were the main source of hydrocarbon in the industrial sector. The other sources of hydrocarbon in Yanbu were fugitive emissions from storage tanks, process areas of refineries and petroleum related industries and loading operations in marine terminal.

3.4 Carbon Monoxide

The highest of the daily average and the hourly average of carbon monoxide recorded at the four stations are given in Table-4. The highest daily average concentration (12.2mg/M^3) was recorded at station #1 during the year 1994. The highest hourly average (16.0mg/M^3) was recorded at station #2 during 1998.

Table-4: Highest daily and hourly average concentrations of Carbon Monoxide

St	Daily Maximum mg/M^3	Jul. day and year of daily max.	Hourly Maximum mg/M^3
1	12.2	126, 1994	14.1
2	8.9	88, 1994	16.0
4	8.1	89, 1993	11.6
5	9.7	246, 1986	11.4

The CO emissions from entire Yanbu areas were not estimated. Road transport might probably, contribute the major part of these emissions.

4. FREQUENCY DISTRIBUTION OF AMBIENT AIR QUALITY

4.1 Sulfur Dioxide

The frequency distribution of sulfur dioxide showed that the mode concentration with an increasing trend with years. For the purposes of presentation, four years data from 1984 with an increment of four years (1984, 1988, 1992 and 1996) were used. The percentage frequency distribution for the station-1 showed a clear shift of the mode from the low range of 0.01-0.015ppm in 1984 to 0.02-

0.025ppm in 1992 and 0.025-0.030ppm in 1996. This increasing trend might be due to the increase in the over all industrial activity in the area and associated fuel oil consumption. Most of the years, a double band structure was observed in the frequency distribution. These double band structures might be due to two predominant wind directions in the region. Similar trends were observed at station-5 also. At station-5, the modal ranges for the years 1984, 1988, 1992 and 1996 were <0.005ppm , 0.005 to 0.01ppm , 0.015 to 0.02ppm and 0.025 to 0.03ppm respectively.

4.2 Nitrogen Oxides

At station-1, the modal range for the years 1984, 1992 and 1996 were 0.005 to 0.01ppm , 0.02 to 0.025ppm and 0.03 to 0.035ppm respectively; which means a clear increase in modes with years. The range was similar forall other stations. At station-4, the modal ranges for the years 1984, 1988 and 1996 were<0.005ppm , 0.01 to 0.015ppm and 0.015 to 0.02ppm respectively. At station-5 the corresponding modal ranges were <0.005ppm , 0.01 to 0.015ppm , 0.015 to 0.02ppm and 0.03 to 0.035ppm respectively. The concentrations at stations 1 and 5 were showing higher values. The concentration at station-2 was widely spread out and at station-4 it was centered near lower ranges.

4.3 Ozone

The frequency distribution pattern of ozone was not showing any significant changes with years. For the three years (1984, 1992 and 1996), the concentration was centered between 0.025 to 0.06ppm. The range was similar for all other stations. During the years (1995, 97), the stations 1 and 5 were showing higher concentrations as these stations fall in the close vicinity of major sources of hydrocarbon and NOx in the area. At station-2, the distribution showed a wider spectrum ranging from very low concentration level to higher ranges. The station-2 is acting as a background station during the night winds (land breezes) and at the same time falling in the downwind of the major sources during day winds (sea breezes). Hence this station is recording both lower ranges and the higher ranges.

4.4 Carbon Monoxide

The mode concentration of carbon monoxide had an increasing trend with years at station-4. The modal ranges of CO at station-4 were 0-0.5 PPM, 1.0-1.5PPM and 2.0-2.5PPM for the years 1984, 1988 and 1996 respectively. The distribution of CO levels at station-5 is also showing a similar trend, except for the year 1996. During the year 1996, the distribution shows a multi modal structure, with major accumulation in the range 0-0.5PPM and a secondary peak at 1.0-1.5PPM. During this year, a new CO analyzer with more sensitivity at lower ranges is installed at this station.

5. CONCLUSIONS

The studies indicated an yearly increasing trend in greenhouse gas emissions. The ambient air quality assessed in Yanbu was marked with an increasing trend in the concentrations of air pollutants. The sulfur dioxide had an yearly increase of 4.0ug/M^3. The nitrogen oxides had increased by three-fold, with less than 17ug/M^3 in 1985 and with 60ug/M^3 in 1988. The annual average of ozone had no trend. The current yearly average of CO is 0.6mg/M^3 with the highest daily average (12.2mg/M^3) and with the highest hourly average (16.0 mg/M^3). The increase in the sulfur dioxide levels was attributed to the increased industrial activity in YANBU. A significant proportion NO_x emissions was from the sources not falling under the scope of environmental regulations. The ozone concentration levels had crossed the RC standard level on many occasions. The background data was successfully used in revising air quality standards, selection of suitable pollution control devices and for prediction of the ground level concentrations, worst-case concentrations and most probable air quality.

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METHODOLOGIES AND RECOMMENDATIONS FOR EMISSION CALCULATION IN THE FRAME OF AIR4EU

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ABSTRACT
Identifying major emission sources in a detailed way requires the development of an emission inventory. Generating emission data for the operation of a CTM requires in addition accurate geographic source allocation and the analysis of temporal variations. Within the Air4EU project, methodologies of emission data generation were reviewed and recommendations for best practice were derived. Results were included in several project reports and compiled in the Milestone Report on Emissions and Data Needs (Pregger et al. 2007). The focus was on selected issues that were relevant for the Air4EU scope and priorities. A methodology review was done for emission calculation and characterisation and the generation of emissions in spatial and temporal resolution. A data review was done for selected topics dealing with emissions from two-wheelers, residential wood combustion and non-exhaust sources of road traffic as well as primary NO_2 emissions.

1. INTRODUCTION
An emission inventory which is able to provide accurate emissions with a high resolution in space, time and substances is a prerequisite for the operation of dispersion and chemistry transport models (CTM). The generation of emission data firstly requires the development of a detailed source inventory that leads to an identification of major sources. Furthermore the generation of emission maps requires a geographic source and emission allocation and the analysis of temporal emission variations. Within the Air4EU project (Air Quality Assessment for Europe: from Local to Continental Scale, supported by the EU 6th Framework Programme), methodologies of emission data generation were reviewed and recommendations were derived for best practice and the assessment and reduction of uncertainties. Results were included in several project reports (see www.air4eu.nl) and were compiled in the Milestone Report on Emissions and Data Needs (Pregger et al. 2007). The focus was on the following issues relevant for the Air4EU scope and priorities:

- Calculation of emissions, spatial and temporal disaggregation (mobile/stationary sources, substantial disaggregation, scenarios, applications, quality assessment, uncertainty analysis)
- Data review for selected topics: PM and benzene from two-wheelers, PM from residential wood combustion, non-exhaust PM emissions from road traffic, NO_2/NO_x ratio of road traffic emissions

2. REVIEW AND RECOMMENDATIONS
A methodology review was done to derive recommendations on basic requirements, best practice and further scientific research. Approaches and information used for emission calculation and characterisation as well as the generation of emissions in spatial and temporal resolution were considered. Data bases used for mobile and stationary sources were described and compared and basic user information about substantial disaggregation (NMVOC, PM, NO_x), the development of scenarios and about quality assessment/uncertainty analysis were provided. Uncertainties in emission data are strongly related to uncertainties in the source specific input data for emission calculations (activities, emission factors) as well as in the usage and availability of spatial allocation data and temporal profiles. The following issues were addressed with regard to emission inventories and were included in the Air4EU Recommendation Document (Denby et al. 2007) which was compiled as a guidance document for the spatial assessment of particulate matter (PM), nitrogen dioxide (NO_2) and ozone (O_3):

- General recommendations: transparent documentation, archiving, cross checking (source contributions, plausibility, completeness), external review (methodology, emission factors), data harmonisation (urban/regional/national), uncertainty analysis (at least qualitatively).
- Calculation of annual emissions: high level of detail for key sources, bottom-up approach if possible, accurate use of statistical data for top-down methodology, use of specific/local/industrial information for calculation and scenarios, consideration of NMVOC/NO_x/PM speciation for comprehensive air quality assessment.
- Spatial/temporal disaggregation: distinguish point/area/line sources, use of a geographic information system, use of industrial data for point sources and their effective emission height, digitised road net for main traffic, statistics for administrative units, land use for area sources, incorporate temperature dependence of temporal variation, use of monitoring data for validation/evaluation of emission data sets (spatial/temporal variation).

The following scientific recommendations were derived and included in (Denby et al. 2007) for urban scale:

- Clear and standardised requirements and guidelines for quality assurance/control of urban emission inventories
- More research activities in Europe dealing with the definition of adequate procedures for an error quantification and sensitivity analysis with regard to emission data in temporal and spatial resolution
- Scientific spatial and temporal analysis of model and parameter sensitivities in cooperation between scientific/city experts for modelling, monitoring and emission calculations
- Derive and use (real-world) emission factors for road traffic emissions that depend on traffic situations rather than on average speed, based on dynamometer measurements for different vehicle types, engine loads and driving conditions (real world driving cycles)
- Systematic verification of vehicle emission factors used in Europe in order to assess their reliability and improve accuracy
- Further examination of fugitive PM emissions, i.e. to carry out more size selective measurements at typical sites and a systematic analysis and determination of model parameters
- Identify and further examine significant PM emission sources in a systematic way in order to better characterise source contributions and their potential health impacts in urban areas
- Systematic analysis of the impact of technical and non-technical PM measures such as low emission zones, improving traffic flows, speed limits, gas engines or particle traps for public transport, street sweeping etc.

3. RESULTS FOR SPECIAL TOPICS CONSIDERED

The special topics of Air4EU led to a review and compilation of basic data for emission calculation and specification with main focus on particulate matter from abrasion and dust suspension due to road traffic, wood combustion and two-wheelers (see Pregger et al. 2007). Within a case study for the city of Rome, different approaches where applied to calculate non-exhaust PM emissions for a certain road. In addition, a data review was done for NO_2/NO_x ratio of traffic emissions in order to specify the increase of direct NO_2 emissions due to the implementation of diesel oxidation catalysts in modern diesel cars and diesel filter systems with passive regeneration for buses and other heavy duty vehicles.

Non-exhaust emissions from mobile sources are caused by brakes, tyres and road dust. In the Air4EU case study „Assessment of non-exhaust PM by road traffic in urban areas“ (Keuken et al. 2007) different approaches were tested for the quantification of road dust suspension on Magna Grecia road in Rome (Figure 1). Exhaust emission factors were selected according to the driving situation from HBEFA (UBA 2004) and COPERT III (Ntziachristos & Samaras 2000). Additional PM_{10} emission factors for gasoline cars, two-wheelers and tyre and brake abrasion were taken from literature (see Keuken et al. 2007). Non exhaust emission factors for road dust suspension were derived from (Duering et al. 2005), (Gehrig et al. 2003) and a modified formula based on (EPA 2003) and (Venkatram 2000). Measured German and Swiss emission factors for re-suspension may lead to an underestimation of PM_{10} emissions in Rome, probably due to different meteorological factors and different road and traffic conditions. The purely statistical US-EPA model is based on the observed correlation between measured emission factor (e.g. by tracer or upwind/downwind measurements) and the selected explanatory variables silt loading and vehicle weight. Even if the US EPA model is modified for normal road conditions (see Venkatram 2000) the US approach seems to rather overestimate road dust suspension in Rome. Assuming emission factors of 100 mg/km for PC/LDV and 550 mg/km for HDV results in a PM_{10}/NO_x emission ratio of ~0.15 to 0.185, similar to the monitored PM_{10}/NO_x ratio for road traffic of 0.15 to 0.17 (see Keuken et al. 2007).

Average NO and NO_x concentrations near roads in Europe decreased during the last decade while NO_2 concentration remained stable or even increased. One reason for this trend is an increase of direct NO_2 emissions. Diesel engines have a higher NO_2/NO_x ratio than gasoline engines. As result of the data review Figure 2 shows selected technology specific NO_2/NO_x ratios that demonstrate higher ratios for new diesel technologies. For diesel engines without oxidation catalyst or catalysed PM filter, the average NO content is usually > 90%. Engine-out NO_x portion contains typically some 5% of NO_2, but even higher than 20% of NO_2 may exist in raw exhaust at some running conditions. The implementation of oxidation catalysts results in NO_2/NO_x ratios of 20% to 50%. Even higher values are produced from vehicles equipped with diesel particulate filters, especially the passive regenerating CRT-systems for heavy duty vehicles (HDV). The expected combination of diesel particulate filters (CRT or PM cat) with NO_x removal (e.g. SCR) - as a result of EURO VI limit values - might reduce NO_2/NO_x ratios to the level of pre-EURO vehicles.

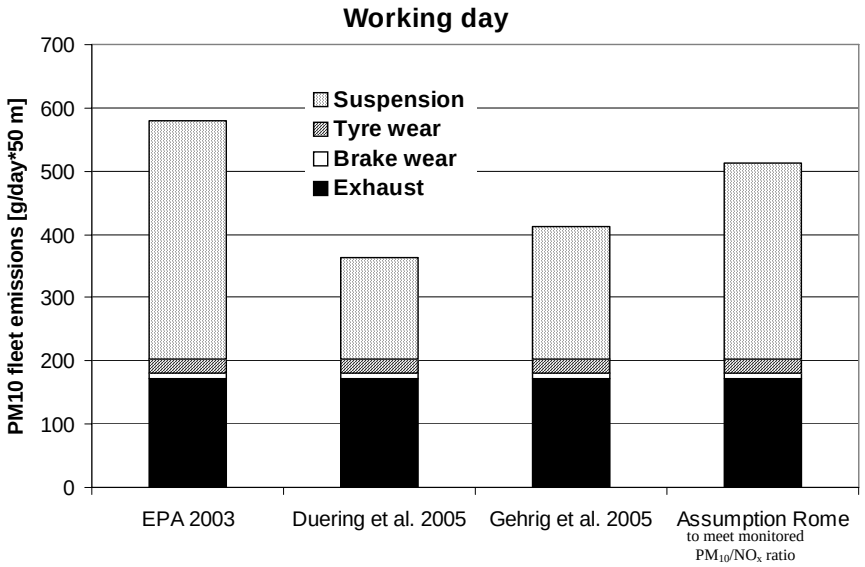

Figure 1: Calculated PM_{10} emissions from road traffic on Magna Grecia 2003: comparison of different approaches for road dust suspension

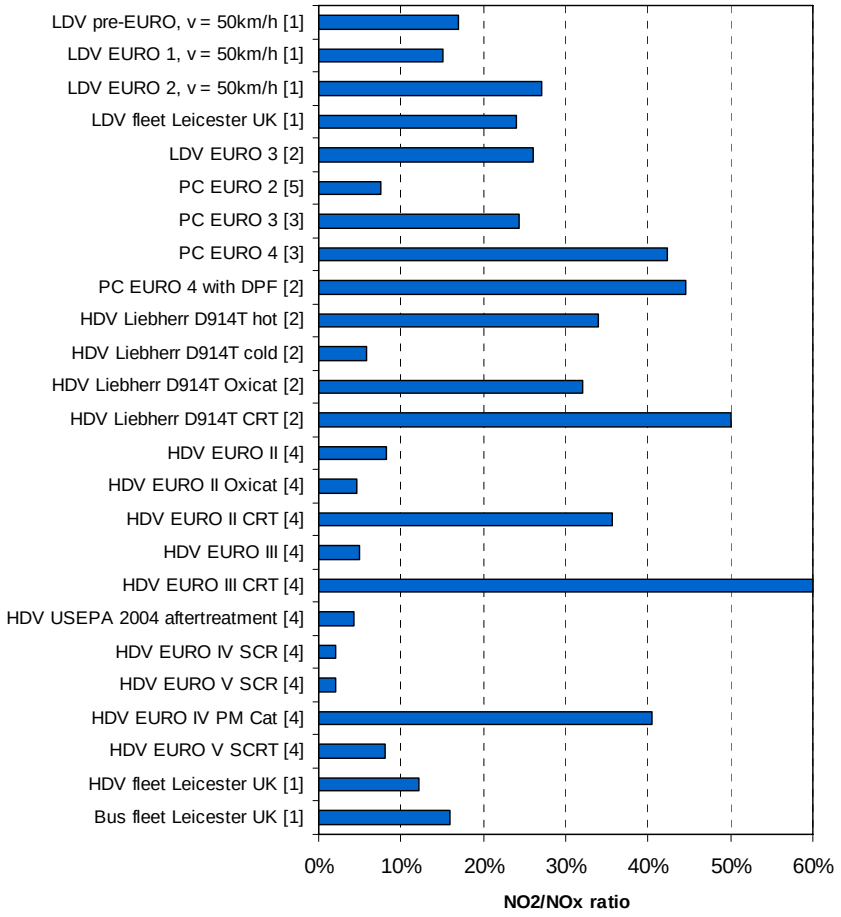

Figure 2: Measured average technology specific NO_2/NO_x ratios for diesel vehicles
Ref.: [1] Latham et al. 2001 [2] Czerwinski et al. 2006 [3] Petit 2006 [4] DC 2006 [5] AQEG 2006

4. CONCLUSIONS

Comprehensive reviews and compilations of relevant information were done within Air4EU to provide and assess methodologies and basic data for emission calculation. Numerous recommendations were derived

from this work defining basic requirements, best practices as well as scientific recommendations, referring to identified knowledge gaps and uncertainties of emission data generation. Results of data reviews for specific topics are provided in (Pregger et al. 2007) and can support emission calculations. An application of different non-exhaust emission factors confirmed PM_{10}/NO_x ratios derived from monitoring data and showed that in the case of Rome abrasion and road dust suspension is a significant PM source which was so far not included in air quality assessment. It is recommended to perform more research on factors affecting non-exhaust emissions such as traffic volume, speed, fleet, road type, dispersion and meteorological conditions.

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Comparisons of data assimilation methods in the framework of the AIR4EU project

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Abstract

Within the frame of the AIR4EU project, AIRPARIF chose to focus on data assimilation methods which allow improving air pollutant concentration estimation and reducing uncertainties from dispersion models.
Two data assimilation methods are currently used in AIRPARIF, that is anisotropic statistical interpolation and innovation intrinsically Kriging. In the Paris case study, we aimed for two objectives: first, to extend the assimilation processes to the new ESMERALDA forecast system and then to get an evaluation of the 2 techniques for ozone.
The two methods have been evaluated against measurements at monitoring stations. Standard deviation maps have been also plotted allowing us to visualize the spatial distribution of ozone concentration uncertainties for the two methods.

1 Background

Air quality assessment in Ile-de-France region is performed using different tools:

- A monitoring network covering all the region.
- An inter-regional emission inventory elaborated jointly with the ESMERALDA project partners.
- 2 integrated mesoscale modelling systems POLLUX and ESMERALDA.

1.1 Monitoring Network

In the frame of the AIR4EU project, we focused on ozone and used almost 80 monitoring stations (fig. 1)

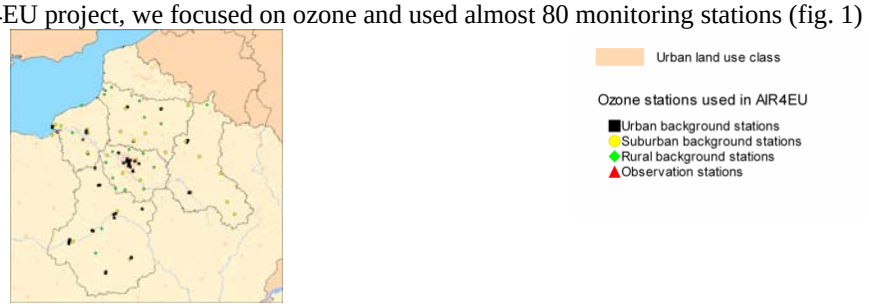

Figure1 : Ozone monitoring stations in ESMERALDA region used in the AIR4EU project

1.2 Integrated modelling systems

1.2.1 Emission Inventory

In the ESMERALDA project, the region partners shared also their experience and knowledge of their home region in order to elaborate a common emission inventory at 1km² resolution.

1.2.2 Air quality forecast systems

The two forecast systems use the chemical transport model CHIMERE developed by LMD/IPSL.
The POLLUX system uses as meteorological forcing, outputs from ARPEGE (Météo-France) while the ESMERALDA system has its own mesoscale meteorological interface, the MM5 model. The system is run daily and provides air quality forecasts from Day-1 to Day+2. CHIMERE is run over an inter-regional domain (9km resolution) and 6 regional domains (3km resolution). The mother domain uses as boundary conditions outputs from PREV'AIR, the French air quality forecast system run by INERIS over Europe with a 0.5° spatial resolution.

1.2.3 Data assimilation

In addition, AIRPARIF performs sequential data assimilation. The methodologies used for ozone and nitrogen dioxide maps have been developed within the frame of the PHD of Nadège Blond [1] "*Assimilation de données photochimiques et prévision de la pollution troposphérique*". The author has applied statistical interpolation using innovative method to model covariance matrices and innovation Kriging to Ile-de-France. Within this work, representativeness errors of measurement data are taken into account.
The AIR4EU Paris case study objectives were the following:

- To better understand and harmonize the assimilation processes.
- To adapt those techniques to ESMERALDA and produce concentration maps and uncertainties.

Within this project, ozone assimilation has been performed and evaluated for SI and Innovation Kriging. The selected period of interest is summer 2005.

2 Methodology

2.1 Presentation of the assimilation data techniques implemented in ESMERALDA in the framework of AIR4EU

Z^t, Z^b, Z^a, Y° respectively average real state, model output, analysis and observation vectors.

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$\varepsilon^b = Z^b - Z^t$ the model error and $\varepsilon^\circ = Y^\circ - HZ^t$, observation error (representativeness error).
H is the observation operator representing the interpolation for grid cells to the measure points
Innovation is difference between observation and model output at measure points

2.1.1 Statistical Interpolation

General formulation :

$$Z^a = Z^b + K[Y^\circ - HZ^b] \quad (1)$$

K is the weight matrix of the linear interpolation calculated by minimisation of error variance.
To calculate K, the model error covariance matrix (B) and the observation error covariance matrix (R) have to be calculated.
The general expression of the analysis can be finally written as follows:

$$Z^a(s_a) = Z^b(s_a) + \sum_{k=1}^{p} w_k(s_a)(Y^\circ(s_k) - Z^b(s_k)) \quad (2)$$

If the observations and the simulations are unbiased, the weights are solution of the following system:

$$\forall k = 1,...p = number\ of\ stations \quad \sum_{l=1}^{p} w_l(s_a)[B_{k,l} + R_{k,k}\delta_{k,l}] = B_{a,k} \quad (3)$$

s_a grid node, s_k and s_l measure points.

Estimation of the global bias (addition of model bias and observation bias) and covariance matrices:
The observation error is evaluated (eq. 4), by linear regression between innovations and model outputs:

$$E[\varepsilon^\circ(s_k)] - E[\varepsilon^b(s_k)] = E[Y^\circ(s_k) - Z^t(s_k)] - E[Z^b(s_k) - Z^t(s_k)] \quad (4)$$

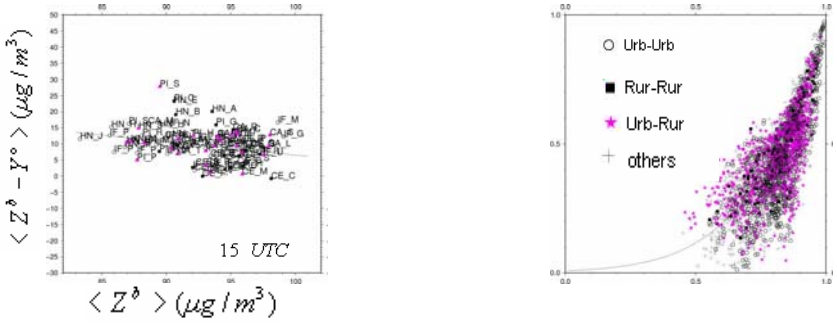

Figure 2: Bias at 15h UTC calculated over the summer 2005 - innovation correlations against observed concentration correlations

The $B_{a,k}$ terms are estimated by using *the simulated ozone concentration correlations.*

A correlation model is adjusted with the following function:

$$f_c(c) = \left(1 + \frac{|1-c|}{L}\right)\exp\left(-\frac{|1-c|}{L}\right) \text{ with } -1 \le c \le 1 \text{ and } L \ge 0 \quad (5)$$

B, is obtained by multiplying the correlation model f_c by the innovation standard deviations (fig. 2).
The analysis error variance is obtained by calculating:

$$\sigma_a^2(s_a) = B_{a,a} - \sum_{k=1}^{p} w_k(s_a)B_{a,k} \quad (6)$$

2.1.2 Kriging

Instead of observations (usually considered in ordinary Kriging), innovations are used to combine model outputs and observations, in the method implemented in AIRPARIF.
The general formulation for innovation Kriging is the following (where *X* is innovation):

$$Z^a(s_a) = Z^b(s_k) + \sum_{k=1}^{p} w_k(s_a)X^\circ(s_k) \quad (7)$$

Kriging and Statistical Interpolation formulations are very close. The only difference is in the estimation of the interpolation weights. In the Kriging method, they are calculated with statistics on *spatia*l data instead of *spatial and temporal* data in the Statistic Interpolation.
In this method, intrinsically Kriging is used. It consists to model semi-variances of differences between two innovations. The variogram is used to model spatial variations.
The weights of the linear interpolation are solutions of the following linear system:

$$\forall k = 1,...,K \ \sum_{l=1}^{p} w_l(s_a)[\gamma_{k,l} + R_{k,k}\delta_{k,l}] - \mu = \gamma_{k,a} \qquad \text{with} \sum_l w_l = 1 \quad (8)$$

s_a grid node, s_k and s_l measure points and μ Lagrange parameter.

J classes of distance are defined, each containing N_j values to calculate the theoretical variogram characterized by its range and sill (by minimisation procedure).
Finally, the weights of the interpolation and the analysis vector can be estimated. The analysis error variances can also be determined. The latter can be seen as kriging uncertainties:

$$\sigma_a^2(s_a) = \sum_{k=1}^{p} w_k(s_a) * \gamma_{a,k} - \gamma_{a,a} - \mu \tag{9}$$

2.2 Evaluation methodology

To evaluate the analysis method efficiency, analysis error statistics are calculated for each hour, that are standard deviation and bias. In order to do it objectively, Jackknife is used. Uncertainty maps have been also plotted. The assimilation methods have been tested for June 2005. The covariance error matrices, for the Statistical Interpolation, have been estimated using July, August and September 2005 data.

3 Results

3.1 Comparison with the model output at monitoring stations

Biases and standard deviations are, on average, much better for analyses (SI and Kriging) than for Simulations. Nevertheless, this is not true for all stations; actually, for stations with low initial bias, it could slightly increase. On the contrary, important biases are significantly reduced (figure 3).

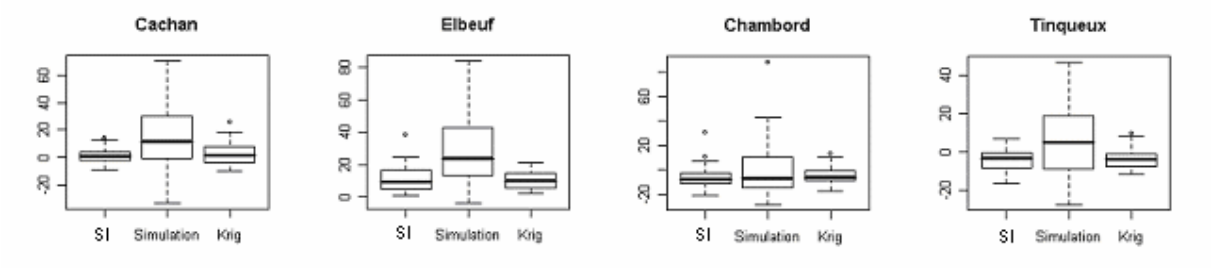

Figure 3: Error boxplot, for analyses and simulation the Cachan, Elbeuf, Chambord and Tinqueux stations at 15h UTC. SI on the left, simulation in the middle and Kriging on the right (Krig).

3.1.1 Comparisons between the data assimilation methods

In the following graphs, error biases and standard deviations for the 2 methods are compared.

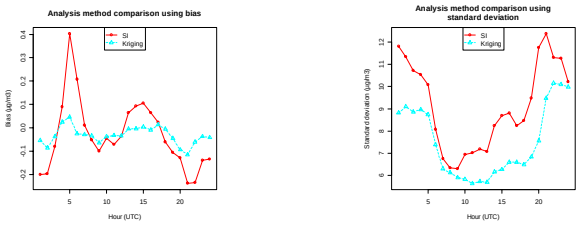

Figure 5: Hourly Biases and standard deviations - SI and Kriging on average over ESMERALDA ozone stations.

Biases and standard deviations are both better for Kriging at any hour of the day. These results are not surprising as the variograms used in the kriging method are adjusted every hour whereas in the Statistical Interpolation method, the covariance matrices are based on climatology. In order to compare the two methods on the whole grid, the error variance have been plotted for each method.

3.1.2 Example of the 20th June 2005

This day, the simulation forecasted a plume on *Centre* region (South of the domain). Observations made in the plume, were not so important. As shown in the next images, assimilation allowed to decrease the concentrations predicted in this area while preserving the simulation shape.

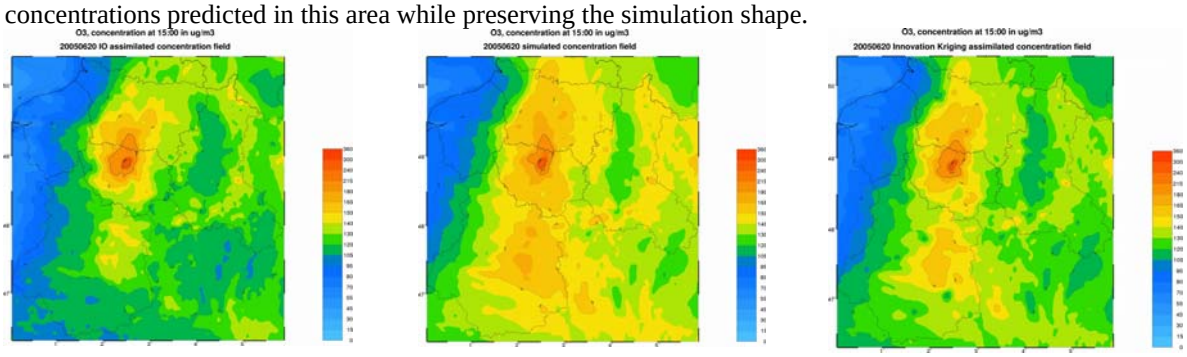

Figure 6: Analysed ozone concentration (µg/m^3), june,20,2005 at 15h UTC. SI, Kriging, Simulation

3.2 Uncertainty assessment

Standard deviations can be plotted for SI and Kriging, (equations 7 and 15). However, we must keep in mind that the assumptions made in the methods are different:

- In the SI method, standard deviations depend on the climatology of the selected hour: from one day to another, the error maps only differ with the number of valid measurement data.
- On the contrary, in the kriging method, the variograms are evaluated at every time step: error maps could be very different from one day to another depending on the concentration patterns.

Error Maps for the 20th June 2005 are presented in the next figure:

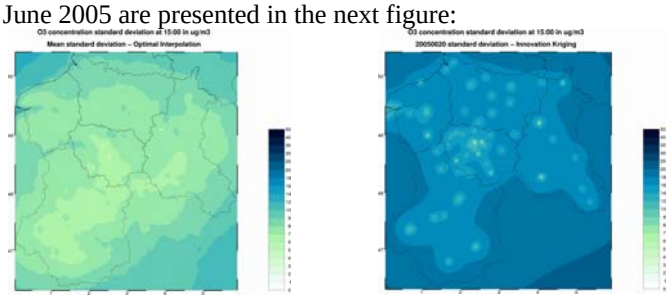

Figure 8: Standard deviation average for ozone concentration – Statistical Interpolation –Kriging.

At the stations, Kriging gave better concentration values than Statistical Interpolation, but the differences were very weak. However, over the whole grid, the SI error is clearly smaller. In the Kriging error maps, the uncertainty increase as we go away from the stations is linked with the variogram range. Between stations, errors strongly depend on the sill. This can be also illustrated by the correction maps for that particular day.

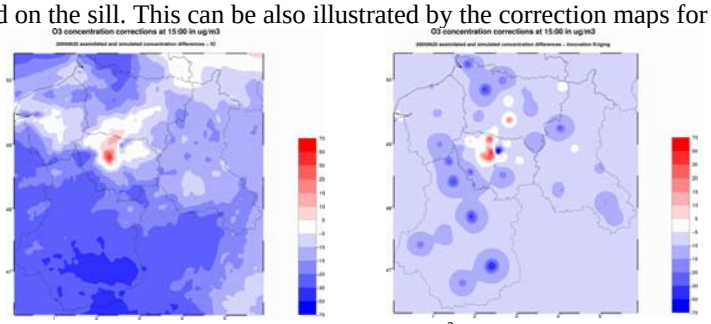

Figure 7: Simulated ozone concentration corrections (µg/m^3) – Statistical Interpolation – Kriging.

In the SI method, the concentration corrections are homogeneous on consistent geographical areas. On the contrary, concentration corrections are localised around the monitoring stations in the Kriging method. For pollutant like ozone, the correction maps for SI assimilation method seem to be much more consistent as correlations between stations and grid nodes are considered instead of distances

Nevertheless, further work should therefore be performed in order to evaluate whether these results are specific to the selected day. A study over June 2005 would be probably more representative.

It could be noticed that the spatial distribution of monitoring stations is very heterogeneous over the ESMERALDA domain and could explain the shape of the Kriging error maps.

4 Conclusion

4.1 Assessment of the case study

In the frame of the Paris case study, two analysis methods have been used to estimate, as well as possible, the real pollutant concentrations in the atmosphere. These methods use model outputs and observations.

The first one is the Anisotropic and heterogeneous Statistical Interpolation which is an adaptation of the classical Statistical Interpolation used in meteorology: it no more considers covariances as function of distances but correlations between stations. The second one is Intrinsically Kriging, a geostatistic method which is used in mining sciences.

A comparison between these methods and the model outputs concludes with a significantly better estimation of pollutant levels using data assimilation methods.

Concerning the differences between methods, even if Kriging seems to be better at measure points, Statistical Interpolation gives more spatially homogeneous results. Nevertheless, some additional work should be undertaken to compare precisely the two methods.

4.2 Improvements in assessment derived from the case study

This case study allowed us to get a better understanding of the SI and Kriging data assimilation methods. Moreover, assimilation seems to be a very promising tool for AIRPARIF needs:

- Annual statistics.
- Network optimization.

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FACILITATING THE COMPARISON OF URBAN AIR QUALITY – A NEW SET OF COMMON AIR QUALITY INDICES

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ABSTRACT

Air quality is a public concern, partly due to the "right to know" principle embodied in European legislation. Despite common European legislation, the way air quality is being interpreted and communicated differs considerably. Easy to understand and (internationally) comparable air quality information is scarce. The CITEAIR project facilitates the comparison of urban air quality in near real-time by introducing two products: a Common Air Quality Index (CAQI), implemented on a common website "www.airqualitynow.eu". Raw air quality data for Europe is available to specialists (Airbase, Ozone web) but this is not easily usable by the general public. To suit their needs, an international index and a dedicated website were developed. This paper describes the CAQI and its application to a number of European cities and presents the common website. The purpose of the CAQI and the website is not to replace more targeted local information. The value added is to provide, for the first time, a European and comparable picture of the air quality in near real-time.

The CAQI was designed as a compromise: hourly updates and easy to understand by the public (main target group), usable in many countries and air quality conditions, and scientifically reasonably rigorous. It is based on a review of existing indices and tested on city data. The CAQI is calculated both for city background and for traffic data. Three time scales are available: an annual, a daily and an hourly index. Annual indices are rare and its calculation differs from the common approach used in daily/hourly indices. The dynamic image arising from hourly updates of the website is considered necessary to attract repeated visits. The CAQI was first presented in 2005 and based on data from a limited number of cities. As participation in the website grows a need to review the PM_{10} part of the index became apparent. This paper describes the common index and pays particular attention to the PM_{10} sub-index and the new year average index.

1. INTRODUCTION

The Framework directive and daughter directives on air quality in the European Union force member states to monitor and report on their air quality and to (actively) inform the public. Over the past years a good number of cities and countries have started to display monitored or modelled air quality data on the internet. The fact that so much air quality information is available and that all EU countries face similar legislation makes it tempting to compare air quality in different cities. This however proves particularly difficult. The most widespread way to interpret air quality on websites is the use of an index to make measurement data more understandable for the general public. A review of existing websites and indices shows that the way air quality is presented and interpreted differs (e.g. Elshout and Léger, 2006; Garcia et al, 2002). Some of these differences are historical: the index existed before the EU regulations came into force; others are conceptual: the index is based on health and exposure criteria, e.g. the UK index (DEFRA). The fact that air quality problems (sources, meteorological conditions, etc.) tend to differ is also one of the reasons. The indices tend to be calibrated to the local situation, to make sure that there is some variation in the index from day to day (to entice repeated visits) and that the typical range of pollutant conditions occurring locally is being covered.

Technical websites on air quality (Airbase, http://air-climate.eionet.eu.int/databases/airbase/; Ozone web, www.eea.europa.eu/maps/ozone/welcome) with a side-by-side, uniform presentation of data are too complicated for the general public. To facilitate the international comparison of near real time air quality by the public the CITEAIR project (http://citeair.rec.org) has developed a common website www.airqualitynow.eu where cities can display their air quality information in near real time. The website uses a common air quality index (hourly, daily and annual) making sure that the interpretation is the same for all participating cities. The common air quality index (CAQI) does not aim to replace

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local indices where they exist. Users (the public, local and health authorities) are used to them, they have their own tailor-made logic to them, etc. A Europe-wide compromise such as the CAQI is not a serious alternative for the local role they perform. The added value of the CAQI is the possibility to facilitate, at a glance, international comparisons. The hourly and daily indices were first presented in Elshout et al (2005). This paper focuses on the recently added annual index and the review and fine tuning of the PM_{10} sub-index since the CAQI was first presented in 2005.

2. THE HOURLY AND DAILY INDICES

The elaboration of the hourly and daily index is described in Elshout et al (2005). The index is inspired by the EU air quality directives and is hence more stringent than the short-term exposure based indices like the UK index (DEFRA). Short-term exposure is rarely (fortunately!) a problem, so indices based on this type of exposure tend to trail at the good end of the scale with limited movement. The CAQI aims to support public information on air quality so it is deemed necessary that the index shows changing values to make the website attractive. So, the index is designed with a short time resolution (hourly for current observation data, and daily for yesterdays data) and a scale that covers the daily concentration variation. The index should be able to capture, for example, a rush hour. The CAQI is calculated and presented both for traffic and city background conditions. The indices were initially developed and tested on data from four project cities (Leicester, Paris, Rome and Rotterdam). A further test, using Airbase data, was done by Leeuw and Mol (2005). As the participation in the website grows (currently about 20 cities) and more pollution regimes have to be covered the index was reviewed. In particular the PM_{10} grid proved difficult. Firstly PM_{10} is monitored on two different time scales: most websites present only daily data or a moving 24 hour average. From a communication point of view this is not ideal: one cannot see the rush-hours and even incidents like a fire, or the fire-works celebrating New Year are impossible to observe (the effect is limited and the timing is delayed). Some networks do provide hourly measurements so at least two index grids are needed. Secondly, the pollution range of PM_{10} differs considerably across Europe making it hard to find a compromise. Lastly $PM_{10,}$ at the commonly prevailing concentrations seems to be the pollutant with the biggest impact on public health. Whereas some potential participants to the website found the earlier grid too strict, others argued that, in view of its health relevance, it was too lenient. A revision was needed.

THE PM_{10} SUBINDEX

Facing the reality that more cities tend to report moving 24-hour average concentrations it was decided to take the 24-hour average concentration as a starting point for the development of a grid instead of the maximum hourly concentration (as in the 2005 version). Secondly the grid was relaxed at the top-end. The previous index grid had regularly spaced 25 $\mu g/m^3$ intervals for each class and it ranged from ranging from 0 to >100 $\mu g/m^3$ for hourly concentration measurements. The current grid doubles the concentration in each class, like for example the NO_2 grid. See table 1. A daily average concentration of 50 $\mu g/m^3$ is considered the maximum value for "medium" pollution. This seems fair in relation to EU guidelines: days with average concentrations above 50 $\mu g/m^3$ should not occur too often. To assure some kind of consistency between the hourly concentrations and the daily averages a relation was established between the daily maximum hourly concentration and the daily average concentration. Using Airbase data from 52 urban and suburban stations for the years 2001-2004 this relation was found to be 0.55 on average. The full CAQI grid for hourly and daily observations is shown in table 1.

The relaxation at the top end of the scale puts the CAQI in line with the grid of for example Baden-Württemberg (Germany) but the grid remains stricter than for example the UK, Brussels and Paris PM_{10} sub-indices. Cairncross and John (2004) propose an index based on the Relative Risk that each pollutant poses. From their analysis it follows that health-wise there is a relation of 2.5 between the hourly NO_2 and daily PM_{10} concentrations. E.g. at the top end of the scale a NO_2 concentration of 400 $\mu g/m^3$ would correspond to a PM_{10} concentration of 160 $\mu g/m^3$. If that assessment is correct the current PM_{10} grid is still quite strict.

Table 1: Pollutants and calculation grid for the CAQI (2007)

Index class	Grid	Traffic				City Background					
		Mandatory pollutant			Auxiliary pollutant	Mandatory pollutant				Auxiliary pollutant	
		NO_2	PM_{10}		CO	NO_2	PM_{10}		O_3	CO	SO_2
			1-hour	24-hours			1-hour	24-hours			
Very low	0	0	0	0	0	0	0	0	0	0	0
	25	50	25	12	5000	50	25	12	60	5000	50
Low	26	51	26	13	5001	51	26	13	61	5001	51
	50	100	50	25	7500	100	50	25	120	7500	100
Medium	51	101	51	26	7501	101	51	26	121	7501	101
	75	200	90	50	10000	200	90	50	180	10000	300
High	76	201	91	51	10001	201	91	51	181	10001	301
	100	400	180	100	20000	400	180	100	240	20000	500
Very High*	> 100	> 400	>180	>100	>20000	> 400	>180	>100	>240	>20000	>500
NO_2, O_3, SO_2:		hourly value / maximum hourly value in $\mu g/m^3$									
CO		8 hours moving average / maximum 8 hours moving average in $\mu g/m^3$									
PM_{10}		hourly value / daily value in $\mu g/m^3$									

* An index value above 100 is not calculated but reported as "> 100"

3. A COMMON YEAR AVERAGE INDEX

Year average air quality indices are rare. However they are a useful indicator for non-experts, facilitating the comparison of cities at a glance. Comparing cities by their individual pollutant levels is difficult as one city might be better on one pollutant and worse on the other. In addition, some cities might monitor other pollutants than others. Such an index is a huge simplification but it does provide an easy way to make some kind of relative assessment on the position of one city to the other or for one city from year to year. A year average index can be devised according to a concentration grid in the same way as most short-term indices. Akkan et al (2004) propose such an index for Baden-Württemberg in Germany. This index uses long-term exposure (one year) health risks as a guiding principle for classifying air quality. Health (risks) being the main public concern, this is an interesting approach. Alternatively a "distance to target" principle can be used. One advantage of a distance to target index is that each parameter considered contributes to the index (unlike the principle where the worst parameter determines the index as in common in most other indices). A distance to target indicator calculates, for each pollutant, a ratio of how far the actual measurement is away from the target value, e.g. the limit value. The overall index is the average of the sub-indices. A distance to target index is based on policy targets or limit values. The limit values have important implications both for environmental policy makers and for the public. Besides, they also have a link to health risks: the European limit values generally relate to the recommendations of the WHO (2005). The distance to target concept is used for the year average index on www.airqualitynow.eu and described in this paper.

Like the hourly and daily indices, the Year Average Common Air Quality Index (YACAQI) is calculated for traffic and city background sites. The calculation of the sub-indices is detailed in table 2. Sub-indices are calculated for each pollutant by dividing the actual year average by the EU limit or target value. The overall city index is the average of the sub-indices for NO_2, PM_{10} (both year average and daily averages) and ozone for the city background index. For the traffic year average index the averages of the sub-indices for NO_2 and PM_{10} are being used. The other pollutants, if data are available, are used in the presentation of the YACAQI but do not enter the calculation of the city

average indices. The main reason is that not every city is monitoring the full range of pollutants. Furthermore for SO_2 we expect that the situation in different kinds of cities is very far apart, being no problem in most cities and a concern in others.

Table 2: Calculation basis for the year average index

Pollutant	Target value / limit value	Calculation
NO_2	Year average is 40 µg/m^3	Year average / 40
PM_{10}	Year average is 40 µg/m^3 Max. number of daily averages above 50 µg/m^3 35 days ≈ year average of 31 µg/m^3	Year average / 40 Year average / 31
Ozone	25 days with an 8-hour average value >= 120 µg/m^3	# days with 8-hour average >=120 / 25
SO_2	Year average is 20 µg/m^3	Year average / 20
Benzene	Year average is 5 µg/m^3	Year average / 5

PM_{10} enters the calculation twice as both limit value parameters tend to behave quite differently. If $PM_{2.5}$ regulation comes into force it will replace one of the PM_{10} sub-indices avoiding that the index will be dominated by particulate matter. The relation between the year average PM_{10} concentration and the number of days with a daily average concentration above 50 µg/m^3 was derived from Airbase data. This and other details on the elaboration of the CAQI and YACAQI, as well as sample applications, are available in Elshout and Léger (2006).

4. SUMMARY AND CONCLUSION

Despite common European legislation on air quality, the way cities and countries present and interpret their air quality differs considerably. To facilitate the comparison from one city to the other by local authorities and the general public a dedicated common website and a new set of indices were developed. The indices are calculated for traffic and city background conditions and for three time scales: hourly, daily and annually.

5. ACKNOWLEDGEMENTS

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APPLICATION OF DATA ASSIMILATION IN OPEN ROAD LINE SOURCE MODELLING

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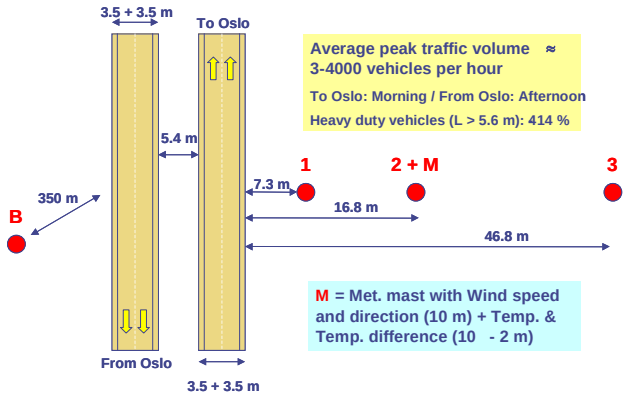
ABSTRACT

The main aim of this study is to investigate how data assimilation can be used to improve Gaussian open road line source models using local (roadside) air quality observations. The idea is to use assimilation to improve estimates of various selected model input parameters, in order to reduce the uncertainty and improve the accuracy of the model output concentrations. The study uses data from a measurement campaign at Nordbysletta, Norway in 2002. The results shows that by using hourly average observations of NO_x at one roadside station, we can improve hourly estimates of different model input parameters such as horisontal diffusivity and initial size of plume, which in turn improves the model concentration results significantly, as is shown by using cross validation, using data from two other stations.

1. INTRODUCTION

Nordbysletta is situated at about 60ºN and 11ºE in the municipality of Lørenskog in a north-easterly direction from Oslo. The area consists of a relatively flat area containing a single (approximately) 850 m long segment of roadway with 4 separate lanes with traffic (Fig. 1). During morning hours the traffic is mainly headed towards Oslo, while in the afternoon and evening most of the traffic is directed in the opposite direction towards the town of Lillestrøm. The average peak traffic volume during morning and afternoon rush hours is around 3-4000 vehicles per hour. During the period 1 January – 15 April 2002 a measurement campaign was conducted at the site (Hagen et al., 2003). The set up of monitoring stations for air quality and meteorology during this period is shown in Fig. 1.

Figure 1: The Nordbysletta 4-lane roadway with air quality monitoring stations (1-3), meteorological station (M), and an air quality background station (B) used during the campaign. Direction is 240º towards Oslo.



Stations 1-3 and B are air quality stations, measuring hourly average concentrations of NO_x at a height of 3.5 m above ground, while station M is a meteorological mast coinciding with air quality station 2. Stations 1-3 and M are all situated on a line placed approximately midway between the end points of the roadway segment considered, while station B is a background station, placed around 350 m from the roadway, and in the opposite direction. The 4-lane roadway geometry and distances to the stations are indicated in the figure. When the wind is headed towards stations 1-3 (1038 hours), the concentration at station B can be subtracted from the concentrations at stations 1-3, to form net observed concentrations of NO_x at stations 1-3 which can then be compared with corresponding model-calculated concentrations.

2. METHODOLOGY

The WORM Model

The line source model used in this study is the WORM model (Weak Wind Open Road Model), which is a newly developed integrated Gaussian line source model at NILU, somewhat similar to the CAR-FMI model

(Härkönen et al., 1996). The model calculates hourly average concentrations in a number of arbitrary receptor points based on local emission data and meteorology. For the purpose of this study the model has been set up to calculate hourly average concentrations of NO_x (only) for each hour during the campaign period at the local (roadside) monitoring stations 1-3 (Fig. 1). The model uses a new numerical procedure for calculating integrated concentrations in receptor points, based on Gaussian quadrature, which is highly accurate, also in situations when the wind blows parallel to the road, contrary to other numerical approximation methods typically used in Gaussian line source models, such as e.g., the one used in the CAR-FMI model.

Emissions and Meteorology

During the measurement campaign, traffic counting was performed locally on an hourly basis. For each hour the number of light and heavy-duty vehicles (with length > 5.6 m) were counted individually on each of the 4 lanes of the roadway. The heavy-duty vehicles constitute around 4-14% of the traffic volume on average. Based on these data, hourly emissions of NO_x are calculated using individual emission factors for the different vehicle classes primarily based on NILU's AirQUIS system (AirQUIS, 2005). For the model calculations performed here, the emission height above the terrain has been set to 1.5 m (tentatively), in order to account for the tailpipe emission height (0.5 m), and the roadway dam effect (1.0 m).

Data recorded at station M (Fig. 1) consist of hourly averaged values of wind speed and direction at 10 m above ground, temperature at 2 m, and temperature difference measured between 10 m and 2 m, the latter to indicate atmospheric stability (Hagen et al, 2003). A separate meteorological preprocessor (MEPDIM) is then used to calculate other derived meteorological parameters such as horizontal and vertical turbulence intensity, friction velocity, Monin-Obukhov length, Lagrangian time scales, and mixing height etc., on an hourly basis, based on standard boundary layer (M-O) similarity theory (AirQUIS, 2005). For the current version of the WORM model, a minimum value of horizontal plume diffusivity (sigma-v) equal to 0.5 m/s is used. Surface roughness at Nordbysletta is tentatively set to 0.25 m based on a Davenport & Wieringa site classification.

Data Assimilation

The data assimilation method used in this study is the SIR (Sequential Importance Re-sampling) method, which is a relatively new data assimilation method, based on a completely general Bayesian statistical framework (Box and Tiao, 1992). The recent FP6 Air4EU project milestone report M.5 describes this and other methodologies of data assimilation in connection with air pollution modelling.

After careful investigation of uncertainty and model sensitivity of different dispersion parameters it was decided to focus on horizontal diffusivity (sigma-v) and initial size of plume (sigma-z0) as two of the most important parameters to estimate on an hourly basis using available roadside observations of NO_x at station 2, leaving the other two stations (1 and 3) for cross-validation.

For the horizontal diffusivity σ_v, the Bayesian prior probability density function (PDF) is defined (tentatively) as a Uniform probability distribution function:

$$\pi_1(\sigma_v) = \begin{cases} \dfrac{1}{(2 - \sigma_v^0)} & \text{for } \sigma_v^0 < \sigma_v < 2 \\ 0 & \text{otherwise} \end{cases} \qquad (1)$$

where σ_v^0 denotes the standard Monin-Obukhov similarity theory calculated σ_v value, and where 2 m/s is used as a (tentative) upper bound for this variable in the prior PDF.

For the vertical initial size of plume parameter σ_{z0} (with $\sigma_{y0} = 2\sigma_{z0}$), the prior PDF is defined (tentatively) as a Gaussian probability distribution function:

$$\pi_2(\sigma_{z0}) = \frac{1}{\sqrt{2\pi}\sigma_0} \exp\left(-\frac{(\sigma_{z0} - \sigma_{z0}^0)^2}{2\sigma_0^2}\right) \qquad (2)$$

where the mean value σ_{z0}^{0} is calculated by the same semi-empirical equation for traffic-originated turbulence as used in the CAR-FMI model (Härkönen et al., 1996, Eq. 9), and with standard deviation σ_0 tentatively set to 0.75, i.e. using σ_{z0}^{0} ± 1.5 m as a 95% probability interval in the prior PDF.

A likelihood function is then defined as follows:

$$L(\sigma_v,\sigma_{z0} \mid c_2) = \frac{1}{\sqrt{2\pi}\sigma_c} \exp\left(-\frac{(c_2 - H(\sigma_v,\sigma_{z0})^2}{2\sigma_c^2}\right) \quad (3)$$

where c_2 is the net observed NO_x concentration at station 2, and where σ_c denotes the associated measurement uncertainty, which is here set to 7.5% relative error. In Eq. 3, H denotes an observation operator (which is the line source model operator itself) linking the data assimilation variables (σ_v and σ_{z0}) to the observed concentrations at station 2.

The posterior PDF can then be defined by using Bayes' theorem (Box and Tiao, 1992), multiplying together the prior PDFs π_1 and π_2 and the likelihood function L as follows:

$$P(\sigma_v,\sigma_{z0} \mid c_2) = k \cdot \pi_1(\sigma_v) \cdot \pi_2(\sigma_{z0}) \cdot L(\sigma_v,\sigma_{z0} \mid c_2) \quad (4)$$

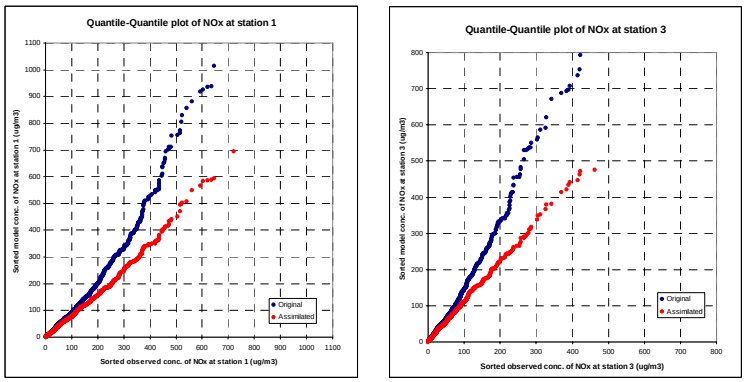
where k is simply a constant so that P integrates to 1. Assimilated values of σ_v and σ_{z0} are then defined (for each hour) as the mean (or expectance) values of the posterior PDF. For the calculations performed here P is approximated using N = 2500 randomly drawn values of σ_v and σ_{z0} from this posterior density.

3. RESULTS AND DISCUSSION

A model evaluation shows a good agreement between model-calculated and observed concentrations of NO_x during the campaign period with correlations in the interval 0.8-0.9, but with clear overestimation of model calculated values at all three stations during low wind speed and stable conditions (< 2-3 m/s).

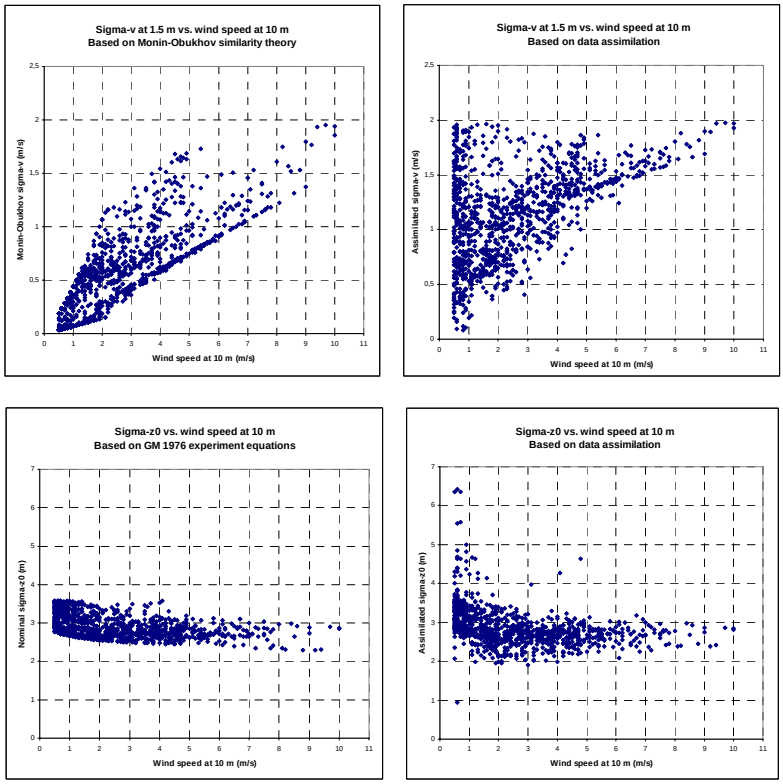
Results of improvement in the model calculated concentrations by using data assimilation are shown in the Q-Q plots in Fig. 2, showing observed and model calculated concentration percentiles at stations 1 and 3 respectively. The blue dots represent percentiles for the unassimilated model, while the red dots represents percentiles for the assimilated model. As can be seen from this figure, use of observed concentrations at station 2 and data assimilation improves the model concentration results significantly at the two remaining stations (1 and 3), especially regarding the high percentiles of the concentration distribution.

Figure 2: Q-Q plots of percentiles of net observed and model calculated hourly average concentrations of NO_x at station 1 (left) and 3 (right) before data assimilation (blue curve) and after (red curve). Unit: µg/m^3.



Estimated values of horizontal diffusivity sigma-v and initial size of plume sigma-z0 before and after data assimilation (as a function of wind speed) are shown in Fig. 3.

Figure 3: Sigma-v (m/s) calculated according to standard Monin-Obukhov similarity theory (top left), and estimated based on data assimilation (top right), and sigma-z0 (m) calculated according to the semi-empirical model of traffic-originated turbulence from CAR-FMI (bottom left) and estimated based on data assimilation (bottom right), as a function of wind speed at 10 m above ground.



As can be seen from Fig. 3, the largest adjustments of the two model parameters take place when the model overestimates the concentrations, in low wind speed (< 2-3 m/s) and stable conditions. Future use of data assimilation in conjunction with the WORM model will also include other important factors such as effective wind speed at plume height, and more advanced modelling and estimation of traffic-produced turbulence.

4. CONCLUSIONS

The study shows that Gaussian integrated line source models, like the WORM model, can clearly be improved by assimilation of local (roadside) air quality observations. Here, horizontal diffusivity and initial size of plume were estimated on an hourly basis using the SIR-method and NO_x observations at a single local (roadside) air quality station. This improved the model results significantly at the two remaining (roadside) stations, especially regarding the high percentiles of the model concentration distributions.

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