# European Community Respiratory Health Survey II Final Report of Work Package 6: PM<sub>2.5</sub> assessment in 21 European Cities of ECRHS II

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# Abstract

The follow-up of a cohort of adults from 29 European centres of the former ECRHS I (1989-1992) will examine the long-term effects of exposure to ambient air pollution on the incidence, course, and prognosis of respiratory diseases, in particular asthma and decline in lung function.

The purpose of this report is to present the annual mean  $PM_{2.5}$  mass concentrations in the participating centres and to describe the methodology and the European-wide quality control programme for the collection of  $PM_{2.5}$  in the ECRHS II. Furthermore it determines whether the ranking of the centres varies if  $PM_{2.5}$  mass concentrations are considered season by season and examines the association of  $NO_2$  with  $PM_{2.5}$  in the participating centres.

Since PM<sub>2.5</sub> is not routinely monitored in Europe, we measured PM<sub>2.5</sub> mass concentrations in 21 participating centres to estimate 'background' exposure in these cities. A standardised protocol was developed using identical equipment in each centre (EPA WINS impactor and PQ167 from BGI, <u>www.bgiusa.com</u>). Filters were weighed in a single central laboratory. Sampling was conducted for seven days per month for a year.

Annual mean  $PM_{2.5}$  mass concentrations varied substantially, with Iceland reporting the lowest value (3.7 µg/m<sup>3</sup>) and northern Italy the highest (44.9 µg/m<sup>3</sup>). We have developed a standardised procedure, appropriate for  $PM_{2.5}$  exposure assessment in a multi-centre study. We expect ECRHS II to have sufficient variation in exposure to assess long-term effects of air pollution in this cohort. Any bias due to variation in the characteristics of the chosen monitoring location (for example proximity to traffic sources) will be addressed in later analyses.

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# 1 Introduction

The follow-up of the European Community Respiratory Health Survey (ECRHS I (Burney et al. 1994) and ECRHS II (European Community Respiratory Health Group 2002)) population, ten years after the first cross-sectional assessment, will allow study of the effect of cumulative long-term environmental exposure on disease incidence and development. There are a large number of study centres increasing the ability to account for potential confounding by area specific characteristics. One major problem, however, is that to date Europe has no common, standardised, publicly available air pollution monitoring network. Knowledge of ambient concentrations that can be used as a proxy for human exposures is a prerequisite for investigation of the long-term effects of air pollution. The overall approach to assigning long-term air pollution exposure in the ECRHS II study is described in the grant. Briefly, we collated existing fixed site monitoring data of the past 20 years, asked for exposure relevant information, and estimated the current annual mean of fine particles (PM<sub>2.5</sub>). PM<sub>2.5</sub> is considered to be a particularly important indicator of health relevant aspects of air pollution. In Europe, however, PM<sub>2.5</sub> is currently not routinely measured. Therefore, ECRHS developed and implemented a PM<sub>2.5</sub> monitoring scheme across 21 participating centres to derive an annual mean (see Figure 1). This is the first time that PM<sub>2.5</sub> mass concentrations have been measured in a large number of cities in Europe, using a single standardised protocol during 12 months.

The association of short-term exposure to ambient air pollution and acute health effects (e.g. symptoms, medication use, decline in lung function, hospital admissions, daily mortality rates) has been extensively investigated in the last 10-20 years, particularly in the US and Europe (Katsouyanni et al. 1997; Holgate et al. 1999; Sunyer et al. 2000; von Klot et al. 2002 (in press)). In contrast there are only a few studies which investigate the long-term effects of cumulative, lifetime exposure to air pollution. These studies are expensive and, ideally, require the follow-up of the same subjects, over their lifetime. To date there are only four cohort studies published three of which have been conducted in the US with the main emphasis on air pollution and life expectancy (Dockery et al. 1993; Pope et al. 1995; Abbey et al. 1999). The only European cohort study in this field has used mortality as an outcome (Hoek et al.

2001). The long-term effects of air pollution on morbidity have been studied little and most of the available evidence relies on cross-sectional comparisons (Ackermann-Liebrich et al. 1997; Martin et al. 1997; Zemp et al. 1999; Sunyer 2001).

The purpose of this report is to describe the full Work Package 6 of ECRHS II, i.e. methodology of and quality control for the WP6 PM<sub>2.5</sub> protocol, and to present the results of measurements made during the period June 2000 until November 2001.

# 2 **Objectives**

According to the original grant proposal, Work Package 6 has the following objectives and deliverables:

#### **Objectives:**

- To estimate at each fieldwork centre a 12-months mean outdoor  $PM_{2.5}$  concentration
- To describe levels of PM<sub>2.5</sub> at each fieldwork centre
- To create a database of summary statistics of  $PM_{2.5}$  for a 12 month period in each of the fieldwork centres

#### Deliverables

**Deliverable Number 2** Standardised protocol for measurement of  $PM_{2.5}$  with SOP for devices and filters and sheets for recording of data

Deliverable Number 9 Pan-European database of annual mean PM<sub>2.5</sub>

**Deliverable Number 10** Short report on assessment of quality of data collected using the standardised protocol

Deliverable Number 12 Short report of distribution of PM<sub>2.5</sub> across Europe

#### Milestones and expected results

 Information on exposure to PM<sub>2.5</sub> available for incorporation into pan-European research database

This report corresponds to Deliverable Number 10 and 12, and is a prerequisite to understand Deliverable Number 9 (data base).

# 3 Methods

The objective of the measurements was to collect sufficient information to derive valid annual mean  $PM_{2.5}$  mass concentrations in 21 centres, and compare monthly and seasonal patterns across participating centres. The rest of the 29 centres joining the follow-up of ECRHS participate only in the collection of the historic air pollution data.

To achieve this, the protocol had to

- be applicable in all research centres which covered nine different languages;
- use identical and affordable equipment from only one manufacturer
- have equipment and procedures that were simple, transparent and error resistant
- allow centralised quality control
- use one laboratory for the weighing and handling of filters
- collect PM<sub>2.5</sub> in a form suitable for later physical and chemical characterisation.

In order to develop a method which is appropriate for a multicentre study, we drew on the experience of the EXPOLIS study (Jantunen et al. 1998; Koistinen et al. 1999) (Air Pollution Exposure Distributions of Adult Urban Populations in Europe). We adopted the same equipment and SOPs (Standard Operating Procedures) as used for the microenvironmental measurements in EXPOLIS. The manufacturer's instruction manual and the US EPA guidelines (US EPA 1998) were further adapted to the requirements of ECRHS II. Further information is available from www.ecrhs.org.

## 3.1 Equipment

All centres were equipped with a Basel  $PM_{2.5}$ -Sampler from BGI (www.bgiusa.com (BGI Incorporated 1998), see Fig. 2 ). The equipment contained an EPA-WINS

impactor (EPA Well Impactor Ninety-six, BGI), a 47 mm filter holder (BGI) for Gelman Teflo filters (R2PJ047, 47 mm, 2  $\mu$ m pore size), a Graseby-Andersen PM<sub>10</sub> inlet and a PQ100 pump (BGI). The Basel PM<sub>2.5</sub> sampler was a special version of the weatherproof portable PM<sub>10</sub> sampling system with a rigid tripod and was adapted for ECRHS II by BGI. The EPA-WINS was a single jet well impactor designed to sample particles with a 50% cut-off size of 2.5  $\mu$ m aerodynamic diameter at a flow rate of 16.7 L/min (US EPA 1997), (Research et al. 1996). Whatman fiberglas filters (32 mm 1820032) and silicone oil (Dow 704) were used for the impaction surface. Oil and filter were replaced after a maximum of 96 operating hours. Tetra Gun Grease (FTI Inc., ordered via BGI) was used for greasing the "O" rings.

The PQ pump was equipped with a microprocessor-controlled timing and mass flow adjustment system (1.0-25 L/min,  $\pm$ 5%). The pump was programmed and calibrated to 16.67 L/min at 20° C and 1013 mbar. It used a normal electricity supply (120 or 240 VAC, 50 or 60 Hz) or an internal 12 Volt lead acid battery enclosed in a weatherproof case, which was further protected by a plastic cover. A bubble Mini-Buck calibrator M-30 was used for regular air flow controls.

For NO<sub>2</sub> assessment NO<sub>2</sub> passive sampling tubes (Burri 1991) from Passam AG, Switzerland (www.passam.ch) were attached as near as possible to the  $PM_{2.5}$ monitor in a position protected from wind and rain. They smampling period covered the same 2 weeks during which  $PM_{2.5}$  was sampled (7 days, see protocol).

## 3.2 Weighing Procedure

All pre- and post-weighing of Teflon filters was conducted in one central laboratory in Aarau, Switzerland by one technician. For the gravimetric analysis a 1  $\mu$ g sensitivity microbalance (Mettler-Toledo MT5) with automatic data transfer to a PC was used. The weighing room was maintained at 22 °C (±1°C) and relative humidity held at 50% (± 5%). Humidity, temperature, and air pressure were constantly monitored. Since the laboratory blank filter fluctuated by only ±5  $\mu$ g during the whole period of May 2000 to Jan 2002, no buoyancy correction (Koistinen et al. 1999) was applied.

The filters were conditioned in the weighing room for between 16 and 48 hours prior to weighing. Tweezers were used to handle the filters by the 4mm rubber rim - and

filters were numbered with a pen on this rim. Each filter was then deionised on both sides using a Multistat deionizer (Haug Biel, Switzerland) prior to weighing. The preweighed filters were placed in a filter cassette in a plastic box (provided by BGI) before they were sent by priority mail to the fieldworkers. After exposure, filters were stored in a refrigerator at +4 C° and were sent back monthly in a single batch to the weighing laboratory (by priority mail) at the end of the 14 day measuring period. The weighing procedure for the loaded filters was identical to that used for unloaded filters, after which the filters were stored in PetriSlides for 47 mm filters (PDMA04700, Millipore Corp.) in a refrigerator at +4°C. In accordance with US EPA guidelines (US EPA 1998), exposed filters were stored for a maximum of 10 days at room temperature prior to weighing. If weighing could not be conducted within 10 days they were transferred to +4° C for a maximum of 30 days.

## 3.3 PM<sub>2.5</sub> and NO<sub>2</sub> Sampling Procedure

Based on the SOP, an English language instruction manual was developed which also contained pictures to help non-English speaking fieldworkers (see www.ecrhs.org). All deviations from the protocol were recorded. There were several levels to the quality control programme so that every stage of the collection procedure was monitored. At least one fieldworker per centre was trained by the coordinating centre in one of nine one-day workshops which included practical and written exercises.

## 3.4 Measurement Schedule

Fig. 3 shows the sampling schedule for all centres with the planned measuring dates for the whole study period June 2000 - November 2001. Sampling was conducted over a 24 and 48 hour period on weekdays and weekends, respectively. The starttime was always midnight Each month, six filter samples, representing seven days of measurement (= 168 hours), were generated, and the seven days were distributed over a two-weeks measuring period. From these six samples a monthly mean concentration was calculated taking into account the different pump sampling times, (i.e. the monthly mean concentration is the time-weighted average concentration of the six measurements). In this way it was possible to use information from filters which had not been exposed for 24 or 48 hours due to technical problems with the pump. Thus, some means are based on less than seven days' data and in these cases the effectively sampled hours are calculated as a percentage of the planned hours (see Tab. 1 and others). The value 100% indicates values are based on 168 hours per month of sampling.

Only one individual was employed to conduct the measurements in each centre and therefore capacity for staff cover during weekends and official holidays was limited. Therefore measurement during these periods was minimised and the two week gap between measurement periods allowed fieldworkers to plan their holidays. In addition centres closely located were able to share equipment.

For the purpose of discussing seasonality 'winter' and 'summer' mean concentration are defined. The **winter** average of the four monthly mean concentrations from **November 2000-February 2001** (In one centre, Albacete, readings obtained from November 2001 were taken as a proxy for November 00). The **summer mean** concentration is the average of the four monthly mean values **May-August 2001**. The **annual mean** is calculated across the 12 monthly values. The schedule represents approximately 23% of all possible measuring days. If more than 12 monthly mean concentrations were available (some centres having extended the period of monitoring beyond the minimum 12 months) , the 12 means with the closest match to the period October 2000 - September 2001 were chosen, see Tab. 1.

Centres started monitoring between June and November 2000, and there was some variation to the sampling schedule due to practical constraints such as funding, personnel resources, or local holidays.

In order to discuss the difference between weekdays and weekends, mean concentrations were calculated of the 60 24 h-weekday filters (**weekday mean**) and of the 12 48 h-weekend filters (**weekend mean**).

The NO<sub>2</sub> tubes were exposed for 14 days during the  $PM_{2.5}$  measuring period. The tubes were usually opened on Monday (first day of  $PM_{2.5}$  measurement) or one day afterward. Thus, the monthly mean was derived from largely the same period, for both pollutants. Some deviations from the standard protocol occurred. In Barcelona, all NO<sub>2</sub> tubes were exposed for 13 instead of 14 days. In Huelva, tubes were opened

on Friday (approx. 3 days prior to the start of the  $PM_{2.5}$  measurement), thus covering 17 - 18 days. Among the other centres, 8% of the monthly NO<sub>2</sub> concentrations are based on either 13 or 15 days. Since the exact opening and closing times were recorded, all values correctly reflect the mean across the true measurement periods.

## 3.5 Sampling Location

Most participating centres were cities with at least 150'000 inhabitants (see Fig. 1). However, in Galdakao, Tartu and Umea only 30'000, 101'000 and 105'000 inhabitants, respectively, lived in the city centre. The location of choice for the PM<sub>2.5</sub> sampler was an official air monitoring station. The advantage was that these stations had a power supply, a clean laboratory for handling the filters and technical support. Furthermore, simultaneous measurements of other pollutants from other equipment at the site would be available. Where this was not possible, another suitable location was identified. In general, this decision was made in collaboration with the local air monitoring authorities. We obtained descriptive data, including pictures and maps about sampling sites. As Table 2 illustrates, 13 samplers were located at an existing fixed air monitoring station, and one sampler was located close (50 m in Umea) to a monitoring station. The distance to the nearest street was between 2 and 100 m. Monitors that were less than 15 m from the street were in Antwerp City (12 m), Basel (5 m), Huelva (10 m), Norwich (5 m), Pavia (6 m), Turin (2 m), and Verona (4 m). The sampling height (above ground level) was between 2 and 25 m. The terminology of 'background' and 'traffic' commonly used to describe sampler locations by monitoring agencies turned out to be based on different definitions and therefore has been disregarded. The distance between the two sites in Antwerp, which studied two discrete populations, was 11.5 km.

## 3.6 Quality Control

## PM<sub>2.5</sub> sampling system

The sampling system pump was designed to record sampling time, cumulative sample volume and stability of the pump. The sampling log data was downloaded to a PC using manufacturer's software. This identified shorter sampling times caused by

shutdown of the pump due to an overloaded filter, a power failure or a manual stop by the fieldworker. Downloaded pump data were sent monthly to the co-ordinating centre where they were checked immediately. Concentrations were not calculated for sampling times of less than 10 hours. The pump flow was checked at least three times during the study and at the end of the study. Deviations of up to  $\pm 5\%$  were accepted (BGI Incorporated 1998).

#### Blanks and Duplicates

For each measurement period one unexposed PM<sub>2.5</sub> filter per centre was used as a field blank filter. During the first six months, the plastic box of the filter was slightly opened, so that the air could circulate by diffusion only. This blank was stored for one to four days in the room where the filter change was performed and the other filters were stored. During the second six months the plastic boxes were kept closed and were stored in the same place. The first filters were mainly used for checking the cleanliness of the room where the filters were changed and stored. The second blanks were collected for checking irregularities due to shipping over a long distance. 292 field blank filters were then used for WP7 (elemental analysis), where they were analysed by ED-XRF. For control purposes we reweighed 4 laboratory blank filters more than 500 times over 20 months. Later in a separate experiment we investigated the impact of storing time, temperature, and whether filters were stored inside the plastic box or openly (see table 10a).

Ultimately, no blank correction was applied to the measurements.

Three times during the 12 measuring periods two NO<sub>2</sub> tubes were exposed (duplicate) and a third tube was not exposed (blank). These additional tubes were used for checking the reliability of the fieldwork procedures, and the method.

## Re-weighing of PM<sub>2.5</sub> filters

As part of the quality control of the weighing procedure and the storage of the filters in the weighing laboratory, 5 % of the exposed filters (including filters from centres which started earlier) were re-weighed in March 2001, i.e. after being kept at 4°C for periods of eleven days to six months. Filters were selected non-randomly to include those with unexpected values e.g. unusually large day-to-day differences. Before and after each weighing session a standard weight and an unexposed laboratory blank were weighed.

#### Quality visits and questionnaire

We visited the three centres Verona, Antwerp and Ipswich in begin of 2001. We filled in a quality check questionnaire (see annex) together with the fieldworker. The other centres received afterwards the same questionnaire. The WP6 responsible persons in the centres were asked to fill in the questionnaire together with their fieldworkers.

## 3.7 Missing Data

#### **PM**<sub>2.5</sub>

For some months in some centres  $PM_{2.5}$  filter data were missing as described above. For two centres missing data were thus estimated:

## Antwerp City and South:

In Antwerp, where two monitors were only 11.5 km apart missing data from one monitor were estimated from data collected from the one nearby. As expected, the figures 4 a) and b) show that the correlation of parallel measurements were very high for the two sites (r = 0.94 for all matched daily concentrations based on exactly the same sampling hours)). Since the ratio Antwerp South / Antwerp City is not always the same during the year (see Fig. 5), but does not depend on the absolute concentrations (see Fig. 6), one correction factor for each period, i.e. month is used (see Fig. 5). Therefore, from linear regressions, we imputed 12.8% (Antwerp City) and 8.3% (Antwerp South), respectively of all planned sampling hours (for more details, see annex 11.1.3).

#### Verona

In Verona we could not measure during February - May 2001 and missing values remained frequent during the rest of the year. Since it was not possible to check the running time of the pump for the available filters according to our quality check (with September 2000 as an exception), data cleaning was done using internal data in addition.

Verona is in the same air shed as Pavia and Turin, i.e. in the plain of the Po Valley and the weather situation is very similar in these three cities (Bendix 2001). As a consequence, the daily variability of the PM<sub>2.5</sub> concentrations in these three cities show a similar pattern with high spatial correlations (Predicatori 2002); also see annex). In fact, the Pearson correlation between the concentrations of Pavia and Turin is high with  $R^2 = 0.75$  (all available concentrations, N = 68, see Fig. 7; winter only  $R^2 = 0.53$ , N = 23). Since the correlation between the concentrations in Pavia and the few available concentrations in Verona ( $R^2 = 0.49$ , N=27, see Fig. 8 a)) and Turin and Verona ( $R^2 = 0.59$ , N=25, see Fig. 8 b)) is also fair, we could calculate the annual mean concentrations by this correlation. The same procedure was done for the winter mean concentration ( $R^2 = 0.42$ , N=11 (Pavia) see Fig. 9 a),  $R^2 = 0.68$ , N=10 (Turin), see Fig. 9 b)). The calculated two annual mean concentrations are 38.8 and 34.6 µg/m<sup>3</sup> (derived from correlation Pavia and Verona and Turin and Verona, respectively) and the two winter concentrations are 67.0 and 62.3  $\mu$ g/m<sup>3</sup>. In the summary tables and figures we show the mean of both calculated means (mean of Verona-Pavia mean and Verona -Turin mean). It was not possible to estimate a summer mean concentration because there were not enough data available. Details are given in annex 11.1.4.

Nevertheless, we point out, that the Verona data have to be handled with care.

## $NO_2$

Only 4.2% of NO<sub>2</sub> values were missing across 20 centres. However NO<sub>2</sub> data from Verona were sparse and could only be included in the winter analyses.

## 4 Results

## 4.1 PM<sub>2.5</sub> Mass Concentrations

Tab. 4 shows the monthly mean PM<sub>2.5</sub> mass concentrations for all 21 centres, the corresponding completeness of the data are indicated in Tab. 1. The resulting annual, winter and summer mean concentrations and the corresponding completeness of data are given in Tab. 3, sorted alphabetically. The corresponding inter-quartile ranges (IQRs) can be seen in the boxplots of Fig. 10 a)-c). The boxplots in Fig. 10 a) include all daily PM<sub>2.5</sub> mass concentrations for the whole annual measuring period and the ranking order is according to the annual mean PM<sub>2.5</sub> mass concentrations. The boxplots in Fig. 10 b) and c) include only the daily PM<sub>2.5</sub> mass concentrations of the winter period (November - January) and of the summer period (May - August), respectively. Figs. 11 and 12 show the time pattern of the monthly and Fig. 13 the annual, winter and summer mean PM<sub>2.5</sub> mass concentrations for all centres, sorted by annual mean. Fig. 14 a) shows the correlation between the summer and winter mean PM<sub>2.5</sub> mass concentrations (N=20). The resulting cross-city Spearman correlation coefficient across season is r = 0.66 as indicated in Tab. 5. The individual ratios between the winter and summer mean concentrations for each centre are given in Tab. 3. The Fig. 15 shows the correlation diagram for the weekend mean versus weekday mean PM<sub>2.5</sub> mass concentrations for all centres. They are highly correlated with a Spearman correlation coefficient of  $r_s = 0.89$ (Verona excluded).

Fig. 16 shows the daily  $PM_{2.5}$  mass concentrations for January 2001 from five centres. The concentrations for the 20<sup>th</sup> and 21<sup>st</sup> of January are the same, as these two days fall on a weekend and concentrations have been calculated from only one filter exposed on both Saturday and Sunday. The centres differ substantially in their daily concentrations. Reykjavik had several low daily concentrations around 3 µg/m<sup>3</sup>, whereas Antwerp had daily concentrations of up to 160 µg/m<sup>3</sup>. While day-to-day variability in Reykjavik was small, it was large in Antwerp. The day-to-day variations of the neighbouring centres in Antwerp show very similar patterns, with the more rural Antwerp South having lower concentrations. The Italian centres in Turin and Verona show similar patterns in their daily variability, but the variation is less marked than in the centres in Antwerp.

Fig. 17 shows daily concentrations for January and February for four centres in central Europe and two northern Italian centres. In January, high PM<sub>2.5</sub> mass concentrations were recorded in all these centres but in February, this only occurred in the Italian centres.

## 4.2 NO<sub>2</sub> Concentrations

Tab. 6 shows the monthly, annual, and winter and summer mean NO<sub>2</sub> concentrations for all 21 centres, sorted alphabetically. Fig. 18 and Fig. 12 show the time pattern of the monthly, and Fig. 19 the annual, winter and summer mean NO<sub>2</sub> concentrations for all centres, sorted by the annual mean concentrations. In Fig. 14 b) the correlation between the summer and winter mean NO<sub>2</sub> mass concentrations (N=20) can be seen. The resulting cross-city Spearman correlation coefficient across season is r = 0.94 as indicated in Tab. 5. The individual ratios between the winter and summer mean concentrations for each centre are given in Tab. 6. The NO<sub>2</sub> concentrations are by a factor 1-2 higher during winter for all centres except Galdakao (ratio = 1).

## 4.3 Correlation between PM<sub>2.5</sub> and NO<sub>2</sub> Concentrations

The Spearman correlation coefficients across all means (Tab. 5) show that  $NO_2$  means were well correlated with  $PM_{2.5}$  mean, both in summer and winter.

Fig. 20 a) - c) shows the correlation diagram between the  $PM_{2.5}$  mass and  $NO_2$  annual, winter and summer mean concentrations of all 20 centres (Verona excluded). The corresponding Spearman correlation coefficient is r = 0.75. Tab. 7 presents the Spearman correlation coefficients between monthly mean concentrations of  $PM_{2.5}$  mass and  $NO_2$  for each centre, ranging from 0.18 (Gothenburg) up to 0.93 (Pavia). The corresponding correlation diagrams are shown in Fig. 21.

## 4.4 Quality Control

#### Missing Data

The quality control indicated that the SOPs were followed in all centres. Five centres had serious technical problems with the sampling pump on at least one occasion (e.g. mass flow sensor was out of order, details see Tab. 8). In three of these cases pumps were fixed within a month, but with loss of data for that month. We noted that during cold weather filters were more likely to tear when the filter holder was opened after sampling. Other problems were power failure, loose connections, errors in the sampler software, and fieldworker absence. These irregularities occurred only occasionally and seldom resulted in frequent missing data or deviations from the measurement schedule. The highest rate of missing data occurred in Verona. In Antwerp, there was only one pump available in November and December and therefore, in the winter months parallel sampling was only possible in January and February. In Albacete, sampling started in December 2000.

During the four months of the winter period the total number of sampled hours was 92.2% (median 96.3%) of all those planned, during the summer period 97.1% (median 100%) and during the whole year 94.1% (median 97.1%), though Verona is not included.

## Storage Time

In four centres less than 50% of the filters were weighed within the prescribed time after exposure due to inadequate refrigeration during local storage (Galdakao and Gothenburg) or delayed transportation of the filters (Antwerp City (50%) and Antwerp South (42%)). Thus, the period without refrigeration was longer than filters from other centres which were kept at ambient temperatures only during transport (mailing). These filters were not excluded, as we expect the loss of material to be negligible. For the remaining 17 centres, on average, more than 94% (min 87%, median 96%) of the filters could be weighed within the prescribed 30 days after exposure and storage of maximum 10 days without refrigeration. The centres had mean storage times between 15 and 22 days (median 18 days), i.e. an avarage of ~8 days due to storing

at the centres (unavoidable, since the 7 filters were sent together after the measuring period) and ~7 to ~14 days between sending and weighing.

## 5% Re-Weighing

The re-weighing of 5% of the exposed filters indicated accurate weighing procedures and no storage irregularities in the laboratory. The mean loss of the 33 re-weighed filters with sampled masses between 0.1 and 2.7 mg (mean 0.58 mg, median 0.36 mg, IQR = 0.20 - 0.79 mg, blank filters excluded) was 3.6% (standard deviation 4.0%, median 3.1%, maximum 10%). No correlation was found between the relative loss and the absolute sampled mass. The storage times between sampling and postweighing for these filters were 6 - 32 days (mean 17 days, median 16 days).

#### Blanks

The concentrations of the blank filters (both types) decreased during the study period (see Fig. 22 and Tab. 9). We have some information suggesting that the load of a blank filter increased with time stored and with storage temperature. Four laboratory blank filters showed no mass increase during the 20 months of running the laboratory, but blank filters stored in the plastic box in the weighing laboratory showed an increase in mass after as little as 7 weeks (see Tab. 10 a) and b)).

Elemental analysis (energy dispersive X-ray fluorescence (Mathys et al. 2001)) has shown that low loaded filters with the same or lower mass as the blank filters, e.g. from Reykjavik, contain the whole range of elements whereas from 292 blank filters from other centres only two (0.7%) contained elements in concentrations above the detection limit.

## 5 Discussion

## 5.1 PM<sub>2.5</sub> and NO<sub>2</sub> Concentrations

## PM<sub>2.5</sub>

Compared to other constituents of ambient air pollution (e.g. TSP, NO<sub>2</sub>, CO), PM<sub>2.5</sub> tends to be homogeneously distributed within regions. This has meant that ambient mean concentrations have been successfully used in epidemiological studies to describe aspects of air pollution shared among large populations, even with only one single monitor available (per community). We have described PM<sub>2.5</sub> concentrations across Europe, using a highly standardised measurements procedure. These data are expected to serve as a major indicator of ambient air quality in the ECRHS II health effects assessment. The power of the cross-city comparison approach in epidemiology is partly determined by the range of concentrations. We have shown that annual mean PM<sub>2.5</sub> mass concentrations vary substantially, with three centres above 35 µg/m<sup>3</sup> and two centres with values around 5 µg/m<sup>3</sup>. This variation of annual means is not as great as observed for winter means (4.8 - 69.2 µg/m<sup>3</sup>), but it is larger than that observed for all summer mean concentrations (range  $3.3 - 23.1 \,\mu g/m^3$ ). Particulate matter levels are influenced by differences in local emission sources (e.g. traffic density, domestic heating), long-range transport, population density, topography or meteorological conditions. Particulate levels are higher in the winter period in regions with temperature inversions during winter. This is well illustrated by Fig. 11, where the North Italian centres show prominent high peak concentrations during January and February. Antwerp City and South, Barcelona, Grenoble, Erfurt, Ipswich, Paris, and Basel show important high mean PM<sub>2.5</sub> mass concentration during January. Since the monthly mean concentrations of Norwich and Ipswich are well correlated (r = 0.79), Norwich may have had a high PM<sub>2.5</sub> pollution period in January, too, but January concentration is missing in Norwich. Even the pattern of the daily variability was very similar across Pavia, Turin, and Verona (Northern Italy), which are recognised to have inversions during the cold season (Bendix 2001). Although they are about 300 kilometres apart, these regions share the same largescale weather conditions such as inversions during winter. Comparison of the January concentrations across Central European cities (e.g. Antwerp, Basel,

Grenoble and Erfurt) (Figs. 16 and 17) which show similar patterns of variation, indicate that macro weather conditions may have influence over a larger distance, and on both sides of the Alps. The opposite case, i.e. higher concentrations during summer, is represented by some Spanish cities like Galdakao and to a certain extent also Oviedo and Huelva. For Huelva, this could be explained by the fact that high particulate levels are met frequently during summer because of African air mass transport events (Saharan dust) (Rodriguez et al. 2001; Rodriguez et al. 2002).

The box-plots show that the daily variability is smaller in summer than in winter for most of the centres, mainly in the high polluted centres. As Tab. 3 and Fig. 14a) illustrate, only one centre (Galdakao) has a higher mean PM<sub>2.5</sub> mass concentration during summer (than in winter) and five centres (Reykjavik, Umea, Gothenburg, Oviedo, Huelva) have no large absolute differences between their summer and winter mean PM<sub>2.5</sub> mass concentrations.

The large seasonal differences of  $PM_{2.5}$  are important as composition and particulate toxicity may also be different. Thus, it might be considered to associate health outcomes to seasonal means separately. Our data show that absolute and relative seasonal differences are very different across centres and the ranking order across the centres changes importantly from winter to summer.

#### $NO_2$

Like  $PM_{2.5}$ , the annual mean  $NO_2$  concentrations also show a large range with two centres with values above 70 µg/m<sup>3</sup> and one centre with a value of 4.4 µg/m<sup>3</sup>. The values for the rest of the centres cover the range between 14 and 60 µg/m<sup>3</sup>. In contrast to the  $PM_{2.5}$  concentrations, summer and winter ranges are not much influenced by season. In addition centres that have the highest levels in winter also tend to show the highest levels in summer. As for  $PM_{2.5}$ , the  $NO_2$  concentrations are, in most of the centres higher during winter than in summer (see Tab. 6 and Fig. 14b)). However the absolute difference between summer and winter means is more constant for  $NO_2$  than for  $PM_{2.5}$ . The lack of relevant seasonal difference in the ranking order of the centres for  $NO_2$  has useful consequences in the epidemiological application of the exposure data. It suggests that even a limited (seasonal) data set may provide some information on the likely annual mean. Since  $PM_{2.5}$  in many

centres is affected much by secondary pollutants and/or wind blown dust, NO<sub>2</sub> is a better indicator for local or urban traffic than  $PM_{2.5}$ . This fact could explain the constant ranking between centres in each seasonal period.

For Huelva the NO<sub>2</sub> concentrations might be slightly underestimated since each measurement period included an additional weekend (with usually lower concentration). But according to Hoek et al. (1997) we expected this bias not to be larger than 3%.

## 5.2 Correlation between PM<sub>2.5</sub> and NO<sub>2</sub> Concentrations

Health effects are unlikely to be related to one single pollutant and epidemiological research therefore uses single pollutants as surrogates for complex mixtures of pollutants from certain sources. We have shown that the across-city correlation between the mean concentrations of PM<sub>2.5</sub> mass and NO<sub>2</sub> is fair and does not depend on the season. In other words, in absence of PM<sub>2.5</sub> data, knowledge of annual mean NO<sub>2</sub> may be a rather useful surrogate. In the past, air monitoring has not been standardised across the ECRHS communities and PM<sub>2.5</sub> is not routinely monitored. NO2 data, however, are often available and may be measured with less effort (passive samplers) than PM<sub>2.5</sub>. These data may be very informative for ECRHS and other studies conducted in regions with limited  $PM_{2.5}$  data. As shown, however, the cross-city correlation between NO<sub>2</sub> and PM<sub>2.5</sub> is influenced by the most extreme high and low level communities. Therefore, among the remaining communities, NO<sub>2</sub> is not an optimal surrogate for fine particulate matter pollution. Health analyses across these cities may contribute to clarifying the independent contribution of these two indicators of pollution to specific health effects. We emphasise that the NO<sub>2</sub>/PM<sub>2.5</sub> surrogate assumption only holds for the long-term mean, across communities. On the short term local level, these two pollutants may be poorly associated. As Fig. 1 well illustrates, some centres show strong seasonal patterns for both, PM<sub>2.5</sub> mass and NO<sub>2</sub> or just for one pollutant. Other centres, however, do not show any seasonal patterns at all. Monthly Spearman correlations between PM<sub>2.5</sub> and NO<sub>2</sub> differ strongly between centres highlighting the dependence of this correlation on local factors. Given the absence of PM<sub>2.5</sub> data in the past, it is useful to know that NO<sub>2</sub> annual mean concentrations very closely predict PM<sub>2.5</sub> in the cities of Pavia, Tartu, and

Antwerp South, whereas in the Spanish centres, the two pollutants indicate very different aspects of pollution.

Inversion and domestic heating do not hold as the only explanation for these differences in correlations between PM<sub>2.5</sub> and NO<sub>2</sub>, as both are expected to be affected similarly by weather conditions (i.e. inversion) and domestic heating contributes to both. NO<sub>2</sub> and PM<sub>2.5</sub> also share traffic, industrial emissions, and combustion in general as common sources. In urban settings, however,  $NO_2$  is mainly attributed to traffic emissions, whereas PM<sub>2.5</sub>, on the other hand, can be significantly impacted by other sources, such as wind blown dust, or long range pollution, such as secondary particles from combustion processes. However, as described above, "alternative" sources, such as Saharan dust in Spain (Rodriguez et al., 2001), probably cause some of the observed patterns. The wide range of correlations between PM<sub>2.5</sub> and NO<sub>2</sub> evokes the hypothesis that monthly PM<sub>2.5</sub> mass concentrations in some centres may be driven by traffic emissions (where correlation with NO<sub>2</sub> is high), whereas in other centres particles from other sources may be of further relevance. However, correlations may in part also be due to weather patterns, such as inversion layers, increasing concentrations of pollutants from all sources. For a confirmation of such hypotheses more detailed analyses are needed. The current (2000/2001) exposure assessment in ECRHS II includes additional indicators of air pollution, namely a variety of characteristics of PM<sub>2.5</sub> (chemical elements, reflectance (black smoke), and oxidative properties) and NO<sub>2</sub> measurements in additional 50 -200 locations per city (in 15 cities only). Such a multi-pollutant approach is aimed at measuring different aspects of air pollution, under the hypothesis that they are of different relevance for the health effects, reflecting different sources.

Interpretation and application to health analyses of these data requires an understanding of the limitations of the exposure assessment including the influence of sampler locations, the sampling schedule, and measurement quality issues.

The proximity of the monitoring location to local sources such as traffic may affect the results and the associations between pollutants. This will be investigated further when we have identified source specific  $PM_{2.5}$  constituents.  $PM_{2.5}$  and – to a lesser degree -  $NO_2$  have been shown to be less affected by proximity to traffic than primary pollutants (Janssen et al. 1997; Monn et al. 1997; Roorda-Knape et al. 1998; Hitchins et al. 2000; Röösli et al. 2000; Tiitta et al. 2002; Wu et al. 2002; Zhu et al. 2002; Zhu

et al. 2002a). We estimated that the applied sampling schedule provides annual means within approximately a 10 % deviation from a 'true' mean based on daily levels (Cyrys et al. 2003; Hazenkamp-von Arx 2003). We have not controlled for weather in this paper as the purpose is to describe the true ambient conditions. As meteorology can be influential from year to year, we will collect meteorological data to assess how representative our measuring period was compared to the conditions prevailing in other years. In addition, we will investigate meteorological determinants of the observed PM<sub>2.5</sub> concentrations. These additional activities are not part of the ECRHS II grant deliverables.

## 5.3 Method and Quality of Data

For the first, time 21 centres across Europe have used a standardised protocol, to measure PM<sub>2.5</sub>. They used the same equipment and exposed filters were weighed by one technician in a centralised laboratory to ensure comparability across the centres. However, this required longer transportation times and the time between sampling and weighing were occasionally close to the limit recommended by the US EPA (US EPA 1998). This could result in an underestimation of the true values due to lost material. According to our quality control procedures, the error is unlikely to be more than 10%. According to the manufacturer (BGI Incorporated 1998), the mass flow of the samplers does not differ more than 5% from each other and this was confirmed in a comparison study of the two BGI samplers from Gothenburg and Umea (Pfeifer - Nilsson and al. 2002).

## Sampling Locations

The location of the monitoring sites is a critical factor for the interpretation of the measured pollution levels. The data on the sampling site characteristics (Table 2) show that proximity to traffic varies between cities. A number of studies measuring  $PM_{2.5}$  mass concentrations at different distances from the road have found 10% to 30% lower  $PM_{2.5}$  levels at a distance of 150 to 300 meters away from the road, with most of the decline happening within the first 15 meters (Janssen et al. 1997; Monn et al. 1997; Roorda-Knape et al. 1998; Hitchins et al. 2000; Tiitta et al. 2002; Wu et

al. 2002; Zhu et al. 2002). These studies have usually investigated effects from very busy streets (approx. 800 -13'900 vehicles/hour) during the day. Even in the presence of very heavy traffic on a Los Angeles highway, PM<sub>2.5</sub> showed very limited spatial variability, whereas other pollutants such as ultrafine particles or CO decreased as much as 80% within the first 100 m (Zhu et al. 2002; Zhu et al. 2002a). In the ECRHS II centres, the closest roads have much smaller traffic volumes and most of the stations were more than 15 meters away. Moreover, the sampling includes the night during which the influence of traffic is much less pronounced. In fact, for the monitoring site in Basel (distance to road 6m), used as a background station by local authorities for many years, Röösli et al. (2000) showed that the  $PM_{2.5}$ levels are highly representative for the entire city. More indirect evidence for the limited influence of distance to traffic comes from our two samplers in Antwerp, one located close to a busy street (12 m) and one away from a street (40 m). The two stations not only had similar PM<sub>2.5</sub> mass concentrations, but the correlation between the daily values was very high also. This suggests that regional factors (background pollution and weather) dominate over the influence of nearby traffic in this city.

Centres in which winter mean values might be overestimated are the Northern Italian centres, where nearby traffic may have some impact. In particular, the concentrations in Turin ought to be interpreted with caution given that the sampler was both close to traffic and located in street characterised as a canyon, which would increase the contribution of nearby traffic. We have no data to directly measure the size of this effect but, based on the findings in the various studies cited above, we do not expect this to exceed 20%. The other locations are less likely to be significantly influenced by nearby traffic and the influence of the sampling heights in our study is expected to be less important than the horizontal distance.

We are currently collecting further information on monitor location, nearby sources, wind patterns, and other pollutants (elemental composition, reflectance (black smoke) of the particles). This will provide information on a potential impact of proximity to traffic. In addition, in 16 cities  $NO_2$  measurements were conducted at 50 - 200 homes. These will be used to describe the local spatial variability of  $NO_2$ .

Our main goal is to derive a reliable estimate of annual mean values of  $PM_{2.5}$ , which reflect the average values in each centre. Our approach is comparable to that used in several epidemiological studies on long-term effects of air pollution (Dockery et al.

1992; Ackermann-Liebrich et al. 1997; Peters et al. 1999; Pope et al. 2002). None of these studies have corrected the mean values of their monitor data for potential effects from local sources.

#### Missing data

There were some days and months for which data is not available which may influence our measurements particularly among centres with large temporal variability. This can be demonstrated using the Antwerp data, comparing two stations located 11.5 km apart. As expected, the two centres show the same pattern in the daily and monthly variability (Röösli et al. 2000) with some very high concentrations. This suggests that it is possible in some cases to estimate missing data in one station from measurements taken in the other location. The winter mean concentration of Antwerp City (37.0 µg/m<sup>3</sup>) is higher than that of Antwerp South (24.4 µg/m<sup>3</sup>) but there are several daily concentrations missing on non-matching days in Antwerp City. Because these centres have a high daily variability (Figs. 16 and 17) a missing day or month can affect the results. This is demonstrated by the fact that if we only used days with available (parallel) data in both locations, the 'winter mean' of Antwerp South would be 28.5 µg/m<sup>3</sup>, as compared to 32.7 µg/m<sup>3</sup> in Antwerp City. The 'City' value is still higher than that of 'South' but the difference is now smaller. In these centres the completion of the data by correlating their available data could improve the validity of the results. In contrast, in centres with low daily or monthly variability, i.e. Reykjavik, substituting missing data will not be expected to make a relevant difference.

Results from Verona need to be interpreted with caution. There were frequent technical irregularities and missing data and no systematic quality control could be done. However, the similarity of the data from Pavia and Verona – 130 kilometres away from each other, but within the same air shed, namely the plain area of Northern Italy – suggest that despite missing values the results from Verona may give an appropriate estimate of the prevailing levels. Mean concentrations from Verona were estimated by correlating data between these related centres.

#### Measurement schedule

Our monthly and winter mean concentrations were based on 23% of all possible measurement days. Therefore our annual mean will lack some precision. Daily variability differed considerably across centres and the validity and precision of our annual mean will not be the same in each city. Our mean values may be very reliable in the 'low PM<sub>2.5</sub> mass concentration' centres but the error may be larger in the most polluted areas. We had a predetermined measurement schedule and much of this error will be random in nature and hence of limited concern in the use of the data for epidemiological analyses. To assess the potential error due to our sampling scheme, we borrowed daily PM<sub>2.5</sub> mass concentrations of the two Swiss SAPALDIA (Zemp et al. 1999) centres Basel and Lugano (Jan 1999 to Jan 2000). We compared the annual means derived from a full daily schedule to the means derived when we applied all possible ECRHS schedules (7 days/month, 84 days/year). For Lugano, the mean deviation of the true annual mean concentration of 24.8 µg/m<sup>3</sup> was 1.3  $\mu$ g/m<sup>3</sup> (relative mean deviation 5.2%) with a maximum of 3.4  $\mu$ g/m<sup>3</sup>. For Basel, the mean deviation from the true annual mean of 18.4 µg/m<sup>3</sup> was 1.6 µg/m<sup>3</sup> (relative mean deviation 8.7%; max 3.2 µg/m<sup>3</sup>). As expected, the bias in the winter means may be larger (Lugano: 2.9µg/m<sup>3</sup>; max 6.5µg/m<sup>3</sup>, corresponding to 10.0% of the true winter mean of 29.2 µg/m<sup>3</sup>): Basel: 2.0µg/m<sup>3</sup>, max 5.3µg/m<sup>3</sup>, or 8.5% of true winter mean (23.5 µg/m<sup>3</sup>). Lugano mostly shares Northern Italian air sheds (Bendix 2001), and these results may give some indication of the degree of error to be expected in the mean values from the Northern Italian ECRHS centres, Turin, Pavia, and Verona. The errors in our mean values are most likely a non-systematic lack of precision. A more detailed quantification of the precision was carried out using the full annual data and daily measures of Erfurt (Cyrys et al. 2003). Our study design is comparable with PM monitoring as conducted by many authorities with measurements covering less than 20% of the time (every 6<sup>th</sup> day). To date, this level of precision has been sufficient to address important health risk aspects (Pope et al. 2002). The protocol was specifically designed to have all centres measuring on the same days. Given the influence of weather conditions on daily PM<sub>2.5</sub> and the similar weather systems across large parts of Europe, this sampling schedule reduced systematic across-city errors in our mean estimates. The schedule is also appropriate for making comparisons between stations in the case of missing values (see above). Therefore, if there are insufficient resources for continuous monitoring, our approach can be

recommended. For logistical reasons, it was not always possible for each centre to follow the schedule exactly but deviations from the planned sampling schedule were small. It should be noted that the implementation of even a very restricted air monitoring program for health research such as ours requires substantial investment (at least 10'000 USD material costs plus several months of person time per centre) in the absence of a European monitoring network. We strongly support the implementation of a standardised air quality monitoring network across Europe.

#### Representative year

We did not control for weather in these data as the purpose is to describe the true ambient conditions. Meteorology, however, may be influential from year to year, thus, we do not know how representative the annual mean is for the mean concentration over a longer period. The same considerations are true with regard to time trends of changes in air pollution.

#### Blanks

A reason for the observed decrease in net weight of blank filters could be that with time the fieldworkers became more experienced in handling the filters leading to less contamination (see Fig. 22).

Since our method for elemental analysis cannot identify light elements (Z<10) contamination, we assume that observed mass increase on filters prior to exposure might mainly be due to organic material and/or water. However, once exposed, these compounds would be sucked off by the pump during the sampling. The observation that the four laboratory blank filters showed no increase during the 20 months of running the laboratory, but blank filters stored in the plastic box in the weighing laboratory showed an increase in mass after as little as 7 weeks (see Tab. 10 a) and b)) supports the possibility that organic compounds from the plastic box may play a role. However, the storage time between the sampling and the reweighing would be marginal.

Furthermore, the inaccuracy of the pump and the mass loss during the storing time before post-weighing have a higher impact on the accuracy of the  $PM_{2.5}$  mass than a blank correction would have had.

# 6 Conclusions

- We showed that PM<sub>2.5</sub> mass concentrations largely differ across the 21 European ECRHS cities. Annual mean PM<sub>2.5</sub> mass concentrations show a wide range of values and an appropriate distribution of values between the two extremes. Our data highlight the potential for improvements in air quality in some areas of Europe. Given the lack of common PM<sub>2.5</sub> monitoring across Europe, the ECRHS II data may also be an important contribution to the European strategies for pollution abatement (http://europa.eu.int/comm/environment/air/cafe.htm)
- Our data suggest that analyses examining the association of health effects with both PM<sub>2.5</sub> winter and summer means will be useful. The ranking orders across centres show a seasonal difference; one may hypothesize that toxicity of PM<sub>2.5</sub> to be different across season due to differences in source profiles. The NO<sub>2</sub> concentrations show also a wide range but since the ranking order does not change importantly between summer and winter, seasonal means of this indicator of mostly traffic related exposure will add less information to annual means, when addressing health effects.
- The diversity in the observed patterns of PM<sub>2.5</sub> and NO<sub>2</sub> across the cities suggests that local factors, i.e. meteorological conditions, specific sources of pollution, or even the location of the air pollution monitor will have to be considered in the interpretation of the exposure assessment results and their application to health analyses.
- We successfully implemented a standardised protocol for the assessment of an annual mean PM<sub>2.5</sub> in European cities. The protocol was found to be practical, permitting appropriate quality control, and could easily be followed by diverse research groups working in nine different languages.

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# 9 Tables

| Table 1:     | Predetermined sampling schedule of the study. Start and end day of measuring period. Shaped months: Sampled |
|--------------|---|
| hours in % c | f planned hours.  |

|               | June    | July              | Aug    | Sep     | Oct    | Nov                  | Dec    | Jan                | Feb                  | Mar               | April              | May                | June              | July               | Aug                  | Sep                | Oct                  | Nov     | Dec     |
|---------------|---------|-------------------|--------|---------|--------|----------------------|--------|--------------------|----------------------|-------------------|--------------------|--------------------|-------------------|--------------------|----------------------|--------------------|----------------------|---------|---------|
|               | 12 - 25 | 10 - 23           | 7 - 20 | 11 - 24 | 9 - 22 | 6 - 19               | 4 - 17 | 8 - 21             | 5 - 18               | 5 - 18            | 16 - 29            | 7 - 20             | 11 - 24           | 9 - 22             | 13 - 26              | 10 - 23            | 15 - 28              | 12 - 25 | 10 - 23 |
| Albacete      |         |                   |        |         |        |                      | 63.1%  | 100%               | 100%                 | 100%              | 100%               | 100%               | 100%              | 100% <sup>c</sup>  | 100%                 | 100%               | 100%                 | 100%    |         |
| Antwerp City  |         |                   |        |         | b      | 57.1% <sup>b,e</sup> | no,e   | 81.9%              | 100% <sup>a</sup>    | 100% <sup>a</sup> | 100%               | 100%               | 100%              | 97.8% <sup>e</sup> | 100% <sup>a</sup>    | 96.5% <sup>e</sup> | 95.3% <sup>c,e</sup> |         |         |
| Antwerp South |         |                   |        |         |        | 100%                 | 100%   | 59.2% <sup>†</sup> | 85.7% <sup>a,t</sup> | 100% <sup>a</sup> | 57.1% <sup>†</sup> | 91.8% <sup>†</sup> | 100%              | 100%               | 88.2% <sup>a,t</sup> | 100%               | 100% <sup>c</sup>    |         |         |
| Barcelona     |         |                   | 85.7%  | 100%    | 100%   | 97.0% <sup>a</sup>   | 92.6%  | 96.8%              | 100%                 | 100%              | 100%               | 100%               | 100%              | 100% <sup>c</sup>  |                      |                    |                      |         |         |
| Basel         |         |                   |        |         | 100%   | 100%                 | 100%   | 85.7%              | 100%                 | 71.4%             | 100%               | 100%               | 71.4%             | 85.7%              | 100%                 | 100%               |                      |         |         |
| Erfurt        |         |                   |        | 100%    | 100%   | 99.4%                | 100%   | 85.7%              | 100%                 | 100%              | 100%               | 100%               | 100%              | 100%               | 100%c                |                    |                      |         |         |
| Galdakao      |         |                   |        |         | 71.4%  | 100%                 | 71.4%  | 92.3%              | no                   | 100%              | 85.7%              | 100%               | 100%              | 100%               | 100%                 | 100%               |                      |         |         |
| Gothenburg    | 100%    | 85.6%             | 71.4%  | 100%    | 85.7%  | 85.7%                | 100%   | 100%               | 100%                 | 100%              | 85.7%              | 71.4%              |                   |                    |                      |                    |                      |         |         |
| Grenoble      |         |                   |        |         |        | 100% <sup>a</sup>    | 100%   | 100%               | 100%                 | 100%              | 100%               | 85.7%              | 95.9%             | 96.2%              | 91.3% <sup>a</sup>   | 100%               | 100%                 |         |         |
| Huelva        |         |                   |        |         | 100%   | 100%                 | 100%   | 100%               | 100%                 | 100%              | 100%               | 85.7%              | 100%              | 85.7%              | 100%                 | 100%               |                      |         |         |
| lpswich       |         |                   |        |         | 100%   | 100%                 | 85.7%  | 100%               | 85.7%                | no                | 71.4%              | 100%               | 100%              | 100%               | 100%                 | 100%               |                      |         |         |
| Norwich       |         |                   |        |         | 85.7%  | 100%                 | 85.7%  | no                 | 71.4%                | 85.7%             | 100%               | 100%               | 100%              | 95.6%              | 100%                 | 100%               |                      |         |         |
| Oviedo        |         |                   |        |         | 82.3%  | 100%                 | 85.7%  | 100%               | 100%                 | 100%              | 100%               | 100%               | 100%              | 100%               | 100%                 | 100%               |                      |         |         |
| Paris         |         |                   |        |         |        | 100% <sup>a</sup>    | 100%   | 100%               | 100%                 | 100%              | 100%               | 100%               | 100%              | 100%               | 100%                 | 100%               | 100%                 |         |         |
| Pavia         |         |                   |        |         | 100%   | 100%                 | 100%   | 90.6%              | 82.9%                | 100%              | 100%               | 100%               | 100%              | 100%               | 100%                 | 100%               |                      |         |         |
| Reykjavik     |         |                   |        |         | d      | 100%                 | 100%   | 100%               | 85.7%                | 100%              | 100%               | 85.7%              | 100%              | 85.7%              | 100%                 | 85.7%              | d                    | d       | d       |
| Tartu         |         |                   |        |         | 100%   | 100%                 | 100%   | 100%               | 100%                 | 100%              | 100%               | 100%               | 100%              | 85.7%              | 100%                 | 100%               |                      |         |         |
| Turin         |         |                   |        | 100%    | 85.7%  | 100%                 | 97.4%  | 100%               | 100%                 | 100%              | 57.1%              | 100%               | 100%              | 100%               | 100%                 |                    |                      |         |         |
| Umea          | С       | 100% <sup>c</sup> | 100%   | 100%    | no     | 85.7%                | 100%   | 57.1%              | 85.7%                | 100%              | 100%               | 100%               | 100% <sup>c</sup> |                    |                      | [                  |                      |         |         |
| Uppsala       | 100%    | 100%              | 100%   | 100%    | 85.7%  | 100%                 | 71.4%  | 100%               | 70.8% <sup>b</sup>   | 95.9%             | 100%               | 100%               |                   |                    |                      |                    |                      |         |         |
| Verona        |         |                   |        | 100%    | 85.7%  | 43%                  | 29%    | 100%               | no                   | no                | no                 | no                 | 100% <sup>g</sup> | 71%                | no                   |                    |                      |         |         |

<sup>a</sup>: sampling start one week later, <sup>b</sup>: sampling start two weeks later, <sup>c</sup>: sampling start one week earlier, <sup>d</sup>: other location, <sup>no</sup>: no data available,

e: For PM<sub>2.5</sub> concentrations presented in Tab. 2 and 3 (after correction): sampled hours equals to those of Antwerp South

<sup>f</sup>: For PM<sub>2.5</sub> concentrations presented in Tab. 2 and 3 (after correction): sampled hours equals to those of Antwerp City

<sup>g</sup>: All filters available, but exact dates and sampling times not clear. Remark: Small changes in the sampling schedule not mentioned

## Table 2. Description of the measurement location characteristics.

| Study centre<br>Name of the station  | Altitude (m a. s. l.) | Sampling height [meters above<br>ground] | Site = official measurements<br>station or other parallel | Other PM measurements in city     | Type of Zone <sup>A</sup> | Characterisation of Zone <sup>B</sup> | Emission source within 500m <sup>c</sup> | Distance to nearest street [m] | Type of nearest street [m] <sup>D</sup> | Traffic volume on nearest street <sup>E</sup> | Wide/Canyon (nearest street) <sup>F</sup> | Frequency of heavy traffic <sup>G</sup> | Street type within 100m <sup>D</sup> | Highest traffic vol. within 100 m <sup>E</sup> | Relevant sites within 100m <sup>H</sup> | Objective traffic info available |
|--|-----------------------|--|---|-----------------------------------|---------------------------|---------------------------------------|--|--------------------------------|---|---|---|---|--------------------------------------|--|---|----------------------------------|
| Antwerp City, VMM plantin & Moretuslei, 2018 A.  | 0                     | 2  | У   | PM <sub>10</sub>                  | u                         | С                                     | t;c                                      | <15                            | m                                       | h   | w   | С                                       | m                                    | h  | b;t;c; bs;g                             | у                                |
| Antwerp South, Hogezandvelden 10, Reet   | 0                     | 2  | n   | n                                 | r                         | r                                     | no                                       | 40                             | S                                       | Ι   | w   | s                                       | S                                    | I  | n                                       | n                                |
| Albacete, Hospital General C/ Hermanos Falco<br>s/n  | 704                   | 12                                       | n   | У                                 | u                         | r                                     | t;c                                      | 30                             | m                                       | m   |   | с                                       | s                                    | m  | b;t;bs                                  | n                                |
| Barcelona, La Sagrera  | 24                    | 3  | У   | PM <sub>10</sub>                  | u                         | r                                     | t;c                                      | 20                             | s                                       | m   | с   | s                                       | m; s                                 | h  | b;t;c                                   | y                                |
| Basel, St. Johannsplatz  | 260                   | 4  | y   | PM <sub>10</sub>                  | u                         | r                                     | t  | <15                            | m                                       | m   | w   | s                                       | m;s                                  | m  | b;c;bs                                  | y                                |
| Erfurt, GSF Environmental Monitoring Station   | 220                   | 2  | у   | PM <sub>10</sub> , 2.5            | u                         | r                                     | t  | 30                             | m                                       | h   | w   | f                                       | m;s                                  | h  | b;t;c;g                                 | у                                |
| Galdakao, Hospital de Galdakao   | 60                    | 14                                       | n   | PM <sub>10</sub>                  | r                         | r                                     | no                                       | 50                             | s                                       | Ι   | w   | s                                       | s                                    | Ι  | b;c                                     | n                                |
| Grenoble, CHW Grenoble, Pneumologie RCH  | 220                   | 6  | n   | PM <sub>10</sub>                  | s                         | r                                     | t;c                                      | 50                             | s                                       |   | w   | n                                       | S                                    | Ι  | С                                       | у                                |
| Gothenburg, Femman, Nils Ericssonsg. 5   | 30                    | 25                                       | у   | PM <sub>10</sub> , <sub>2.5</sub> | u                         | С                                     | t  | 30                             | m                                       | h   | w   | с                                       | m;s                                  | h  | b;t;c;r                                 | У                                |
| Huelva, Manuel Lois, Via Paisajista s/n  | 50                    | 4  | у   | PM <sub>10</sub> , 2.5            | u                         | c; r                                  | t;c                                      | <15                            | s                                       | I   | w   | s                                       | S                                    | m  | b;t;c;g                                 | n                                |
| Ipswich, Environmental Agency, Cobham Road   | 50                    | 8  | n   | n                                 | s                         | r                                     | t;c                                      | 100                            | s                                       | Ι   | w   | s                                       | s                                    | Ι  | r                                       | n                                |
| Norwich, Guildhall Hill  | 50                    | 10                                       | у   | PM <sub>10</sub> , 2.5            | u                         | с                                     | t  | <15                            | s                                       | I   | w   | s                                       | m                                    | m  | b;t;c                                   | у                                |
| Oviedo, Consejería/Lab. de Medio Ambiente  | 276                   | 2  | у   | n                                 | u                         | r                                     | p;t;c                                    | 15                             | S                                       | m   | w   | s                                       | S                                    | m  | b;c;t                                   | у                                |
| Pavia, Pavia1, Via Folperti  | 70                    | 2  | у   | PM <sub>10</sub>                  | u                         | r                                     | t;c                                      | <15                            | m                                       | m   | w   | f                                       | m;s                                  | m  | b;c                                     | у                                |
| Paris, Lab. d'Hygiene de la ville de Paris   | 75                    | 13                                       | У   | PM <sub>10</sub> , <sub>2.5</sub> | u                         | r;c                                   | t;c                                      | 25                             | S                                       | m   | С   | s                                       | S                                    | m  | b;c                                     | у                                |
| Reykjavik, Vifilsstadir  | 53                    | 5  | n   | n                                 | S                         | r                                     | t  | 35                             | S                                       | I   | w   | S                                       | S                                    | I  | b                                       | у                                |
| Tartu, Dep. Pub. Health, Uni. Tartu  | 84                    | 17                                       | n   | n                                 | u                         | r                                     | p;t                                      | 50                             | m                                       | m   | w   | f                                       | m;s                                  | m  | b                                       | у                                |
| Turin, V. M. Cristina  | 239                   | 2  | У   | PM <sub>10</sub>                  | u                         | r;c                                   | t;c                                      | <15                            | m                                       | m   | С   | t                                       | m                                    | m  | b;t;c;bs                                | n                                |
| Umea, Roof station Biblioteket (IDUN1)   | 10                    | 15                                       | У   | PM <sub>10</sub> , 2.5            | u                         | r;c                                   | t  | 15                             | S                                       |   | С   | S                                       | m;s                                  | m  | b;c;bs                                  | У                                |
| Uppsala, 50 m from station Kungsgatan  | 8                     | 15                                       | n   | PIM <sub>10</sub> , 2.5           | u                         | r;c                                   | t  | 50                             | m                                       | n   | w   | T                                       | m                                    | n  | b;t;c;r                                 | У                                |
| verona, Corso Milano   | 60                    | 4  | У   | PM <sub>10</sub>                  | u                         | r                                     | t;C                                      | <15                            | m                                       | m   | w   | С                                       | m;s                                  | m;I  | b;c;g                                   | у                                |
| <ul> <li><sup>A</sup>: u: Urban (Station is located within the city)</li> <li>s: Suburban (Station is located in the outskirts (fringe) of a city, or in small residential areas outside the main city)</li> <li>r: Rural (Station is located outside the city)</li> <li><sup>B</sup>: The major activity in the representative area. More than one is possible.</li> <li>r: Residential</li> <li>c: Commercial</li> <li>i: Industrial</li> <li><sup>C</sup>: Major emission source in station environment within 500 meters:</li> <li>p: Public power, co-generation and district heating</li> <li>t: Traffic</li> <li>c: Commercial, institutional and residential combustion</li> <li>i: Industrial activities</li> <li>no: No emission source within 500 meters</li> <li><sup>D</sup>: m: Main street</li> <li>s: Side street</li> <li>h: Highway</li> </ul> |                       |  |   |                                   |                           |                                       |  |                                |   |   |   |   |                                      |  |   |                                  |

| Table 3: | Ar     | nual, sum   | mer and wir | nter mean  | concentra | ations of         | PM <sub>2.5</sub> r | nass and |
|----------|--------|-------------|-------------|------------|-----------|-------------------|---------------------|----------|
| correspo | onding | ratios of w | vinter/summ | ner mean ( | concentra | tions in <b>j</b> | ug/m³.              |          |

| Centre             | Annual PM <sub>2.5</sub> | Ranking                 | Winter PM <sub>2.5</sub> | Summer PM <sub>25</sub> | Ratio w/s        |
|--------------------|--------------------------|-------------------------|--------------------------|-------------------------|------------------|
| Contro             | % hours                  | Annual PM <sub>25</sub> | % hours                  | % hours                 | PM <sub>25</sub> |
|                    | 13.1                     | 17                      | 15.4                     | 11.5                    | 1.34             |
| Albacete (AL)      | 96.9%                    |                         | 90.8%                    | 100%                    |                  |
|                    | 23.3*                    | 4                       | 31.6*                    | 17.5*                   | 1.81             |
| Antwerp City (AC)  | 98.5%                    |                         | 95.5%                    | 100%                    |                  |
|                    | 21.2*                    | 6                       | 26.6*                    | 17.0*                   | 1.56             |
| Antwerp South (AS) | 98.5%                    | _                       | 95.5%                    | 100%                    |                  |
|                    | 22.2                     | 5                       | 30.2                     | 20.0                    | 1.51             |
| Barcelona (BA)     | 97.7%                    |                         | 96.6%                    | 96.4%                   |                  |
|                    | 17.4                     | 9                       | 23.7                     | 13.7                    | 1.73             |
| Basel (BS)         | 92.9%                    |                         | 96.4%                    | 89.3%                   |                  |
| , <i>i</i>         | 16.3                     | 13                      | 19.9                     | 11.0                    | 1.81             |
| Erfurt (ER)        | 98.8%                    |                         | 96.3%                    | 100%                    |                  |
|                    | 16.3                     | 12                      | 10.8                     | 20.9                    | 0.52             |
| Galdakao (GA)      | 85.1%                    |                         | 65.9%                    | 100%                    |                  |
|                    | 19.0                     | 7                       | 28.0                     | 12.9                    | 2.17             |
| Grenoble (GN)      | 97.4%                    |                         | 100%                     | 92.3%                   |                  |
|                    | 12.7                     | 18                      | 12.5                     | 11.4                    | 1.10             |
| Gothenburg (GO)    | 90.5%                    |                         | 96.4%                    | 82.1%                   |                  |
|                    | 17.3                     | 10                      | 17.2                     | 16.9                    | 1.02             |
| Huelva (HU)        | 97.6%                    |                         | 100%                     | 92.9%                   |                  |
|                    | 16.5                     | 11                      | 21.3                     | 15.0                    | 1.42             |
| Ipswich (IP)       | 86.9%                    |                         | 92.9%                    | 100%                    |                  |
|                    | 16.2                     | 14                      | 17.7                     | 14.6                    | 1.21             |
| Norwich (NO)       | 85.3%                    |                         | 64.3%                    | 98.9%                   |                  |
|                    | 15.9                     | 15                      | 17.5                     | 16.7                    | 1.05             |
| Oviedo (OV)        | 97.3%                    |                         | 96.4%                    | 100%                    |                  |
|                    | 35.3                     | 3                       | 55.3                     | 19.9                    | 2.78             |
| Pavia (PA)         | 97.8%                    |                         | 93.4%                    | 100%                    |                  |
|                    | 17.8                     | 8                       | 21.0                     | 15.9                    | 1.32             |
| Paris (PS)         | 100%                     |                         | 100%                     | 100%                    |                  |
|                    | 3.7                      | 21                      | 4.8                      | 3.3                     | 1.45             |
| Reykjavik (RE)     | 86.9%                    |                         | 96.4%                    | 92.9%                   |                  |
|                    | 14.8                     | 16                      | 15.6                     | 10.2                    | 1.53             |
| Tartu (TA)         | 98.8%                    |                         | 100%                     | 96.4%                   |                  |
|                    | 44.9                     | 1                       | 69.2                     | 23.1                    | 3.00             |
| Turin (TU)         | 95.0%                    |                         | 99.4%                    | 100%                    |                  |
|                    | 5.6                      | 20                      | 5.8                      | 4.9                     | 1.18             |
| Umea (UM)          | 85.7%                    |                         | 82.1%                    | 100%                    |                  |
|                    | 10.4                     | 19                      | 11.6                     | 7.2                     | 1.61             |
| Uppsala (UP)       | 93.7%                    |                         | 85.6%                    | 100%                    |                  |
|                    | 35.7***                  | 2                       | 64.7***                  |                         |                  |
| Verona (VE)        | 45.2%                    |                         | 42.9%                    |                         |                  |

\* Corrected PM<sub>2.5</sub> values, see section methods, uncorrected values: Annual: 24.0 (AC), 20.8 (AS); Summer: 17.5 (AC), 17.3 (AS); Winter: 37.0 (AC), 24.4 (AS)

\*\* The NO<sub>2</sub> November concentrations do not correspond to the same time period

\*\*\* Estimated values, see section methods
| Table 4: | Monthly | / mean PM <sub>2.5</sub> mass | concentrations in µ | ւg/m³ |
|----------|---------|-------------------------------|---------------------|-------|
|----------|---------|-------------------------------|---------------------|-------|

|                |            |              |             | 2.5 11144  |      |             | anon       | μg          |             | 1            | 1           |             |
|----------------|------------|--------------|-------------|------------|------|-------------|------------|-------------|-------------|--------------|-------------|-------------|
|                | Jan        | Feb          | Mar         | Apr        | Мау  | Jun         | Jul        | Aug         | Sep         | Oct          | Nov         | Dec         |
| Albacete       | 12.2       | 15.8         | 81          | 10.2       | 93   | 11.3        | 114        | 13.9        | 14.9        | 167          | 14.8        | 18.9        |
| sd             | 3.5        | 6.9          | 2.0         | 1.3        | 3.3  | 2.3         | 5.5        | 3.1         | 3.8         | 5.3          | 6.7         | 3.5         |
| median         | 11.8       | 15.3         | 7.2         | 9.6        | 8.1  | 11.7        | 12.6       | 13.0        | 13.1        | 17.6         | 17.8        | 20.2        |
| Antwerp City*  | 64.4       | 28.3         | 30.8        | 14.3       | 17.7 | 15.6        | 13.5       | 23.0        | 16.1        | 21.8         | 15.4        | 18.2        |
| sd             | 51.6       | 13.1         | 10.9        | 8.2        | 5.9  | 5.2         | 3.0        | 14.1        | 4.5         | 11.8         | 3.7         | 8.3         |
| median         | 35.6       | 25.6         | 32.4        | 12.3       | 18.1 | 15.2        | 14.5       | 14.2        | 18.5        | 21.4         | 14.1        | 15.0        |
| Antwerp South* | 52.8       | 25.9         | 30.1        | 11.5       | 16.2 | 15.9        | 11.2       | 24.6        | 19.8        | 18.5         | 12.7        | 15.0        |
| sd             | 42.3       | 10.2         | 13.4        | 6.9        | 6.1  | 3.6         | 2.8        | 14.9        | 6.5         | 9.8          | 3.0         | 6.9         |
| median         | 29.0       | 27.0         | 29.1        | 9.5        | 17.1 | 16.1        | 12.1       | 14.9        | 22.5        | 19.9         | 11.6        | 12.4        |
| Barcelona      | 35.0       | 32.3         | 21.8        | 16.9       | 21.0 | 24.8        | 19.7       | 14.5        | 13.0        | 13.8         | 22.1        | 31.5        |
| Sd             | 13.5       | 16.2<br>25.9 | 12.2        | 3.9        | 9.4  | 12.4        | 4.3        | 1.7         | 3.7         | 8.2          | 9.1         | 13.6        |
| Bacol          | 42.6       | 22.4         | 14.0        | 0.0        | 15.5 | 12.2        | 79         | 19.5        | 15.4        | 10.5         | 12.0        | 1/ 0        |
| Dasei          | 42.0       | 13.1         | 14.9        | 29         | 32   | 29          | 1.0        | 46          | 10.4        | 13.0         | 81          | 79          |
| median         | 38.8       | 24.8         | 8.2         | 10.6       | 14.3 | 14.9        | 8.0        | 20.6        | 10.6        | 11.5         | 13.4        | 12.3        |
| Erfurt         | 41.9       | 15.3         | 21.5        | 10.0       | 13.3 | 11.3        | 7.2        | 12.1        | 24.6        | 15.3         | 10.6        | 11.9        |
| sd             | 24.2       | 6.9          | 12.7        | 4.9        | 6.4  | 5.2         | 1.9        | 6.7         | 4.2         | 8.7          | 3.4         | 7.0         |
| median         | 46.7       | 13.6         | 20.4        | 8.0        | 10.9 | 12.6        | 7.8        | 16.0        | 25.3        | 13.0         | 9.8         | 11.9        |
| Galdakao       | 10.2       |              | 12.8        | 11.7       | 15.8 | 27.0        | 17.0       | 23.8        | 17.3        | 21.0         | 14.6        | 7.5         |
| sd             | 3.7        |              | 5.2         | 2.6        | 6.9  | 10.2        | 5.7        | 10.8        | 7.4         | 8.3          | 6.9         | 4.6         |
| median         | 12.9       |              | 13.6        | 11.1       | 16.6 | 29.6        | 16.0       | 21.5        | 14.6        | 19.3         | 13.3        | 5.8         |
| Grenoble       | 38.5       | 23.3         | 15.2        | 13.4       | 14.0 | 13.4        | 11.6       | 12.5        | 12.9        | 23.3         | 16.6        | 33.4        |
| Sd             | 15.2       | 12.3         | 9.5         | 2.4        | 3.7  | 5.6<br>15.9 | 2.3        | 6.7         | 6.1<br>11 0 | 5.5<br>25.9  | 6.0<br>19.1 | 18.7        |
| Gothonburg     | 1/ 0       | 29.7         | 19.7        | 12.0       | 14.7 | 12.0        | 0.0        | 10.2        | 69          | 20.0<br>10 / | 10.1        | 20.0        |
| Somenburg      | 4.5        | 42           | 13.0        | 8.0        | 49   | 82          | 27         | 44          | 1.2         | 74           | 23          | 42          |
| median         | 15.2       | 8.7          | 14.6        | 12.3       | 10.8 | 11.5        | 8.1        | 8.0         | 6.8         | 19.3         | 12.9        | 8.7         |
| Huelva         | 19.0       | 15.7         | 11.0        | 13.8       | 14.9 | 19.0        | 12.6       | 21.2        | 25.5        | 20.7         | 11.4        | 22.7        |
| sd             | 7.3        | 3.9          | 4.2         | 6.8        | 5.4  | 11.6        | 5.0        | 10.6        | 13.5        | 12.4         | 2.6         | 9.9         |
| median         | 18.3       | 16.0         | 8.8         | 10.2       | 16.3 | 15.5        | 12.4       | 23.4        | 28.6        | 16.4         | 11.7        | 18.0        |
| lpswich        | 31.3       | 17.6         |             | 7.5        | 15.0 | 12.1        | 8.9        | 24.2        | 13.2        | 15.2         | 18.1        | 18.1        |
| sd             | 20.6       | 10.9         |             | 1.0        | 7.1  | 3.7         | 4.8        | 14.2        | 3.6         | 7.8          | 18.2        | 12.9        |
| median         | 37.4       | 14.9         | 00.0        | 1.4        | 13.1 | 11.3        | 7.4        | 21.6        | 14.1        | 12.1         | 8.9         | 11.3        |
| Norwich        |            | 22.4         | 22.Z        | 13.3       | 12.0 | 13.5        | 8.0        | 24.8        | 13.0        | 18.1         | 13.9        | 10.9        |
| median         |            | 14.5         | 19.1        | 10.3       | 10.2 | 10.4        | 2.9<br>7.8 | 19.1        | 14.0        | 9.0<br>17.6  | 12.1        | 11.7        |
| Oviedo         | 19.6       | 18.1         | 10.2        | 12.4       | 15.0 | 16.6        | 12.4       | 22.9        | 16.6        | 14.6         | 13.4        | 18.8        |
| sd             | 5.0        | 6.3          | 5.4         | 2.4        | 7.6  | 6.9         | 3.3        | 7.6         | 5.5         | 5.2          | 4.7         | 10.8        |
| median         | 18.5       | 16.2         | 9.4         | 13.0       | 11.0 | 16.0        | 13.1       | 24.8        | 19.0        | 15.0         | 14.3        | 16.1        |
| Pavia          | 61.4       | 83.9         | 36.4        | 21.9       | 23.2 | 19.5        | 14.2       | 22.8        | 25.8        | 38.1         | 32.3        | 43.8        |
| sd             | 18.5       | 36.0         | 19.5        | 8.2        | 14.5 | 10.1        | 5.1        | 7.4         | 15.9        | 8.0          | 9.3         | 15.0        |
| median         | 51.8       | 67.9         | 32.6        | 20.5       | 16.0 | 24.2        | 14.1       | 24.6        | 23.3        | 37.4         | 27.6        | 39.7        |
| Paris          | 36.0       | 21.9         | 18.2        | 10.8       | 18.7 | 15.5        | 10.9       | 18.6        | 14.4        | 22.7         | 11.8        | 14.1        |
| S0<br>median   | 10.1       | 12.0         | 18.8        | 4.6        | 8.0  | 4.1<br>16.4 | 1.9        | 8.4<br>16.0 | 4.9         | 7.9          | 2.8<br>11.8 | 3.4<br>13.8 |
| Rovkiavik      | 41.2<br>ΛΛ | 75           | 27          | 3.6        | 5.6  | 27          | 27         | 2.0         | 26          | 20.1         | 17          | 27          |
| sd             | 3.6        | 3.2          | 1.6         | 1.1        | 1.1  | 0.7         | 0.9        | 0.9         | 1.0         |              | 1.0         | 1.6         |
| median         | 3.2        | 7.0          | 2.4         | 3.6        | 5.1  | 2.2         | 2.9        | 2.0         | 2.6         |              | 4.6         | 1.7         |
| Tartu          | 13.8       | 14.5         | 15.1        | 15.3       | 8.1  | 9.2         | 12.1       | 11.6        | 16.9        | 26.7         | 21.2        | 12.8        |
| sd             | 5.5        | 11.1         | 5.6         | 6.7        | 4.1  | 4.1         | 2.0        | 6.6         | 9.0         | 10.4         | 7.3         | 4.7         |
| median         | 14.6       | 7.4          | 14.4        | 14.8       | 7.9  | 7.7         | 11.7       | 10.0        | 13.1        | 27.0         | 21.0        | 15.0        |
| Turin          | 73.4       | 87.3         | 43.2        | 24.2       | 26.2 | 21.1        | 20.4       | 24.5        | 45.3        | 56.6         | 49.3        | 66.9        |
| sd             | 19.5       | 26.4         | 25.2        | 6.5        | 11.4 | 8.7         | 8.2        | 7.6         | 15.8        | 19.9         | 21.8        | 29.4        |
| median         | /0.6       | 84.1         | 32.7        | 23.4       | 25.0 | 26.3        | 21.0       | 25.0        | 46.8        | 47.1         | 37.8        | /6.6        |
| umea           | 0.3        | 5.3          | 8.4         | 5.3        | 4.2  | 4.3         | 0.4        | 4.6         | 5.5         |              | 5.6         | b.1         |
| S0<br>median   | 67         | 3.9          | 4.7<br>6.4  | 2.2<br>4 7 | 1.2  | 3.U<br>3.6  | 2.0<br>6.8 | 4.0         | 2.4<br>5 0  |              | 1.7         | 2.2<br>7.0  |
| Unnsala        | 10.2       | 13.5         | <u>0.</u> 4 | 10.6       | 4.2  | 9.0<br>Q () | 76         | 70          | 72          | 22.3         | 11 /        | 10.6        |
| shhang         | 4.6        | 5.4          | 5.9         | 2.1        | 1.7  | 4.5         | 3.3        | 2.7         | 2.9         | 8.6          | 3.3         | 3.1         |
| median         | 12.6       | 14.5         | 9.0         | 10.7       | 3.8  | 7.8         | 7.7        | 8.4         | 8.0         | 22.3         | 11.8        | 11.7        |
| Verona         | 61.4       | 1            |             |            |      | **          | 16.0       |             | 38.6        | 29.5         | 30.8        | 60.9        |
| sd             | 21.4       |              |             |            |      |             | 6.3        |             | 19.3        | 2.7          | 17.6        | 0.3         |
| median         | 61.6       |              |             |            |      |             | 14.2       |             | 28.3        | 28.6         | 21.2        | 60.9        |

\* Corrected values, see section methods

\*\*Filters available, but exact dates and sampling time not clear

|                               | PM <sub>2.5</sub> | PM <sub>2.5</sub> | PM <sub>2.5</sub> | NO <sub>2</sub> | NO <sub>2</sub> |
|-------------------------------|-------------------|-------------------|-------------------|-----------------|-----------------|
|                               | Annual            | Winter            | Summer            | Annual          | Winter          |
| $PM_{2.5}$ Winter $\rho$      | 0.946             |                   |                   |                 |                 |
| N                             | 21                |                   |                   |                 |                 |
| $PM_{2.5}$ Summer $\rho$      | 0.815             | 0.656             |                   |                 |                 |
| N                             | 20                | 20                |                   |                 |                 |
| NO <sub>2</sub> Annual ρ      | 0.754             | 0.777             | 0.725             |                 |                 |
| N                             | 20                | 20                | 20                |                 |                 |
| NO <sub>2</sub> Winter $\rho$ | 0.821             | 0.858             | 0.723             | 0.976           |                 |
| N                             | 21                | 21                | 20                | 20              |                 |
| NO <sub>2</sub> Summer $\rho$ | 0.686             | 0.686             | 0.729             | 0.982           | 0.935           |
| N                             | 20                | 20                | 20                | 20              | 20              |

Tab. 5: Spearman correlation coefficient  $\rho$  of the annual, winter and summer mean concentrations of PM<sub>2.5</sub> and NO<sub>2</sub>.

N is the number of centres involved, all p's were statistically significant with p < 0.002.

| WP6 | ECRHS II |
|-----|----------|
|-----|----------|

### FINAL REPORT

| Table 6:       Annual, winter, summer and monthly mean NO₂ concentrations in µg/m³ |      |      |      |      |      |      |      |      |      |      |       |      |                           |                           |                           |                              |
|--|------|------|------|------|------|------|------|------|------|------|-------|------|---------------------------|---------------------------|---------------------------|------------------------------|
| Centre   | Jan  | Feb  | Mar  | Apr  | Мау  | Jun  | Jul  | Aug  | Sep  | Oct  | Νον   | Dec  | Annual<br>NO <sub>2</sub> | Winter<br>NO <sub>2</sub> | Summer<br>NO <sub>2</sub> | Ratio w/s<br>NO <sub>2</sub> |
| Albacete   | 29.5 | 23.3 | 22.2 | 17.3 | 16.4 | 15.8 | 15.7 | 16.9 | 15.4 |      |       | 32.7 | 20.5                      | 28.5                      | 16.2                      | 1.76                         |
| Antwerp City   | 63.7 | 62.9 | 91.0 | 8.2  |      | 32.2 | 22.9 | 72.2 | 74.8 | 76.5 | 85.7* |      | 59.0                      | 70.8                      | 42.4                      | 1.67                         |
| Antwerp South  | 48.9 | 38.1 | 43.4 | 7.6  | 18.9 | 13.6 | 8.2  | 32.4 | 28.7 | 31.1 | 26.2* | 29.8 | 27.2                      | 35.8                      | 18.3                      | 1.96                         |
| Barcelona  | 87.6 | 70.3 | 91.0 | 87.1 | 85.4 | 75.0 | 45.6 | 60.6 | 66.3 | 63.4 | 58.9  | 61.8 | 71.1                      | 69.7                      | 66.7                      | 1.04                         |
| Basel  | 45.9 | 50.4 | 39.0 | 24.6 | 42.5 | 25.6 | 25.0 |      | 32.6 | 41.8 | 41.0  | 44.0 | 37.5                      | 45.3                      | 31.0                      | 1.46                         |
| Erfurt   | 40.8 | 29.7 | 31.0 | 22.7 | 24.2 | 20.4 | 14.9 | 20.0 | 29.5 | 31.9 | 29.2  | 26.3 | 26.7                      | 31.5                      | 19.9                      | 1.58                         |
| Galdakao   | 33.7 |      | 28.6 | 31.1 | 31.4 | 32.4 | 27.7 | 29.3 | 30.1 | 39.1 | 33.9  | 21.8 | 30.8                      | 29.8                      | 30.2                      | 0.99                         |
| Grenoble   | 47.3 | 40.2 | 48.7 | 33.4 | 30.1 | 27.7 | 24.9 | 26.5 | 30.6 | 41.2 | 39.7  | 47.8 | 36.5                      | 43.8                      | 27.3                      | 1.60                         |
| Gothenburg   | 43.8 | 44.9 | 34.3 | 37.2 | 37.2 | 24.4 | 24.2 | 27.5 | 29.0 | 27.3 | 30.5  | 30.5 | 32.6                      | 37.4                      | 28.3                      | 1.32                         |
| Huelva   | 28.6 | 29.9 | 21.4 | 10.2 | 18.7 | 14.9 | 10.7 | 12.8 | 18.6 | 27.6 | 26.6  | 37.1 | 21.4                      | 30.6                      | 14.3                      | 2.14                         |
| lpswich  | 43.8 | 40.5 |      | 30.2 | 31.2 | 21.0 | 17.6 | 28.2 | 22.6 | 36.2 | 42.2  | 35.1 | 31.7                      | 40.4                      | 24.5                      | 1.65                         |
| Norwich  | 51.5 | 46.4 | 42.6 | 33.2 | 34.7 | 32.7 | 27.6 | 33.5 | 33.0 | 42.9 | 49.9  | 45.7 | 39.5                      | 48.4                      | 32.1                      | 1.51                         |
| Oviedo   | 49.4 | 54.6 | 39.0 | 36.6 | 42.1 | 36.2 | 33.3 | 40.8 | 41.1 | 41.7 | 47.3  | 49.2 | 42.6                      | 50.1                      | 38.1                      | 1.31                         |
| Pavia  | 57.3 | 72.1 | 54.8 | 39.7 | 42.9 | 36.9 | 31.9 | 25.8 | 43.3 | 49.1 | 52.0  | 59.0 | 47.1                      | 60.1                      | 34.4                      | 1.75                         |
| Paris  | 55.1 | 56.6 | 52.7 | 47.5 | 49.2 | 49.0 | 39.7 | 49.6 | 45.9 | 55.5 | 57.0  | 52.3 | 50.8                      | 55.3                      | 46.9                      | 1.18                         |
| Reykjavik  |      | 4.7  | 6.2  | 2.9  | 4.3  |      | 3.7  | 4.0  | 3.7  |      | 8.0   | 6.2  | 4.4                       | 4.9                       | 4.0                       | 1.23                         |
| Tartu  | 12.9 | 12.5 | 16.5 | 15.4 | 10.4 | 10.5 | 11.4 | 11.0 | 13.3 | 17.3 | 16.6  | 14.8 | 13.5                      | 14.2                      | 10.8                      | 1.31                         |
| Turin  | 76.0 | 86.1 | 78.1 | 68.1 | 74.9 | 72.1 | 64.8 | 46.3 | 84.8 | 65.6 | 72.2  | 75.7 | 72.1                      | 77.5                      | 64.5                      | 1.20                         |
| Umea   | 32.7 | 27.2 | 21.6 | 17.1 | 15.5 | 12.7 | 12.0 | 13.3 | 18.4 | 16.3 | 24.5  | 23.6 | 19.6                      | 27.0                      | 13.4                      | 2.01                         |
| Uppsala  | 28.5 | 24.0 | 23.9 | 22.0 | 19.7 | 17.3 | 15.6 | 18.3 | 28.2 | 25.2 | 24.7  | 20.2 | 22.3                      | 24.3                      | 17.7                      | 1.37                         |
| Verona   | 40.9 |      |      |      |      |      |      |      | 77.6 |      | 91.7  | 39.8 |                           | 57.5                      |                           |                              |

\* Do not correspond to the same period

| Tab. 7:                | Spearman                | correlatio | n coefficie  | nt r <sub>s</sub> betweer | monthly mean   | n concentrations of |
|------------------------|-------------------------|------------|--------------|---------------------------|----------------|---------------------|
| PM <sub>2.5</sub> mass | and NO <sub>2</sub> . p | is the sig | nificance ar | <u>nd N th</u> e num      | ber of monthly | y values included.  |

| Centre        | r <sub>s</sub> | р       | N  |
|---------------|----------------|---------|----|
| Albacete      | 0.261          | 0.467   | 10 |
| Antwerp City  | 0.617          | 0.077   | 9* |
| Antwerp South | 0.902          | 0.001   | 12 |
| Barcelona     | 0.266          | 0.404   | 12 |
| Basel         | 0.811          | 0.0025  | 11 |
| Erfurt        | 0.799          | 0.0018  | 12 |
| Galdakao      | 0.236          | 0.484   | 11 |
| Gothenburg    | 0.179          | 0.577   | 12 |
| Grenoble      | 0.842          | 0.0006  | 12 |
| Huelva        | 0.210          | 0.512   | 12 |
| lpswich       | 0.688          | 0.019   | 11 |
| Norwich       | 0.573          | 0.066   | 11 |
| Oviedo        | 0.526          | 0.079   | 12 |
| Paris         | 0.511          | 0.090   | 12 |
| Pavia         | 0.930          | <0.0001 | 12 |
| Reykjavik     | 0.291          | 0.448   | 9  |
| Tartu         | 0.909          | <0.0001 | 12 |
| Turin         | 0.636          | 0.026   | 12 |
| Umea          | 0.342          | 0.304   | 11 |
| Uppsala       | 0.522          | 0.082   | 12 |

\* November NO<sub>2</sub> concentration excluded, since the time period does not correspond to that of PM<sub>2.5</sub>.

## Table 8: Problems in the centre

| Centre             | Problems  |
|--------------------|---|
| Albacete (AL)      | Power failure in December 2000  |
| Antwerp City (AC)  | From October - December 2000, the pump of AS was used, some filters were overloaded, pump stopped   |
| Antwerp South (AS) | Some filters were overloaded, pump stopped  |
| Barcelona (BA)     | Mass flow sensor defect in October 2001   |
| Basel (BS)         | Broken filters during cold periods  |
| Erfurt (ER)        |   |
| Galdakao (GA)      | Mass flow sensor defect in February 2001  |
| Grenoble (GN)      |   |
| Gothenburg (GO)    | Broken filters during cold periods  |
| Huelva (HU)        | Thread was worn out at the beginning of study   |
| Ipswich (IP)       | Mass flow sensor defect in March 2001   |
| Norwich (NO)       | Mass flow sensor defect January 2001  |
| Oviedo (OV)        |   |
| Pavia (PA)         | Some filters were overloaded, pump stopped  |
| Paris (PS)         |   |
| Reykjavik (RE)     | During October 2000, the pump was located next to a chimney, from October - December 2001 next to street  |
| Tartu (TA)         |   |
| Turin (TU)         | Some filters were overloaded, pump stopped  |
| Umea (UM)          | Thread was worn out in October 2000, broken filters during cold periods   |
| Uppsala (UP)       | Broken filters during cold periods  |
| Verona (VE)        | Several technical and organisational problems. No pump from February<br>- Mai 2001. Filters from August 2001 lost. No downloaded pump<br>information available. |

Tab. 9:Mass of blanks filters

|        | in μg/m³   |      |         |       |       |        |          |           |          |        |        |           |       |          |       |       |         |         |        |               |              |      |
|--------|------------|------|---------|-------|-------|--------|----------|-----------|----------|--------|--------|-----------|-------|----------|-------|-------|---------|---------|--------|---------------|--------------|------|
| Centre | Gothenburg | Umea | Uppsala | Turin | Pavia | Verona | Albacete | Barcelona | Galdakao | Huelva | Oviedo | Reykjavik | Basel | Grenoble | Paris | Tartu | Norwich | Ipswich | Erfurt | Antwerp South | Antwerp City | mean |
| 1      | 34         | 20   | 15      | 285   | 40    | 76     | 30       | 56        | 16       | 45     | 44     | 82        | 16    | 34       | 45    | 18    | 58      | 40      | 33     | 25            | 43           |      |
| 2      | 47         | 34   | 43      | 107   | 48    | 15     | 86       | 77        | 35       | 30     | 53     | 24        | 33    | 35       | 22    | 54    | 77      | 37      | 86     | 36            | 17           | 47.4 |
| 3      | 101        | 12   | 40      | 94    | 10    | 11     | 154      | 69        | 23       | 9      | 20     | 37        | 14    | 31       | 10    | 20    | 57      | 19      | 17     | 15            | 26           | 37.6 |
| 4      | 11         | 20   | 11      | 22    | 25    | 8      | 31       | 25        | 53       | 36     | 52     | 9         | 28    | 14       | 16    | 45    | 11      | 21      | 13     | 23            | 30           | 24.0 |
| 5      | -4         | 32   | 52      | 9     | 14    | 13     | 8        | 13        | 14       | 52     | 16     | 25        | 16    | 16       | 11    | 60    | 10      | 42      | 46     | 16            | 55           | 24.6 |
| 6      | 6          | 25   | 18      | 6     | 12    | 14     | 17       | 14        | 16       | 11     | 11     | 29        | 18    | 27       | 28    | 8     | 35      | 7       | 24     | 43            | 42           | 19.6 |
| 7      | 27         | 6    | 16      | 30    | 14    | 58     | 10       | 5         | 29       | 4      | 18     | 3         | 18    | 8        | 2     | 12    | 21      | 18      | 10     | 30            | 35           | 17.8 |
| 8      | 21         | 21   | 17      | 14    | 16    |        | 2        | 14        | 2        | 50     | 24     | 22        |       | 18       | 16    | 13    | 13      | 15      | -14    | 15            | 21           | 15.8 |
| 9      | 41         | 12   | 17      | 33    | 1     |        | 30       | 31        | 23       | 44     | 7      | 8         |       | 19       | 17    | 11    | 42      | 19      | 21     | 8             | 21           | 21.3 |
| 10     | 27         | 13   | 15      | 5     | 18    |        | 12       | 36        | 2        | 17     | 4      | 4         |       | 23       | 29    | 19    | 5       | 15      | 4      | 9             | 10           | 14.1 |
| 11     | 1          | 7    | 14      | 36    | 16    |        | 8        | 23        | 19       | 34     | 15     | 11        |       | 15       | -3    | 45    | 32      | 35      |        | 13            | 26           | 19.3 |
| 12     |            | 17   | 18      | 22    | 13    |        | 19       | 8         | 13       | 42     | 3      | 6         |       | 9        | 10    | 12    | -5      | 7       |        | 30            |              | 14.0 |
| 13     |            |      |         |       | 5     |        | -7       | 20        | 18       | 6      | 12     | 11        |       |          |       |       | 24      |         |        |               |              | 11.1 |
| 14     |            |      |         |       |       |        |          | 28        | 13       | 32     | -9     | 7         |       |          |       |       | 26      |         |        |               |              | 16.2 |
| 15     |            |      |         |       |       |        |          | 25        | 17       | 14     | 1      |           |       |          |       |       | 5       |         |        |               |              | 12.4 |
| 16     |            |      |         |       |       |        |          |           |          | 13     |        |           |       |          |       |       | 7       |         |        |               |              | 10.0 |

|            |            |         | · · · · · · · · · · · · · · · · · · · |      |
|------------|------------|---------|---------------------------------------|------|
| Date       | Date       | Storing | Filter No.                            | Net  |
| 1.Weighing | 2.Weighing | time    |                                       | [µg] |
|            |            |         |                                       |      |
| 09.04.2002 | 24.04.2002 | 15 days | A                                     | 0    |
| 09.04.2002 | 24.04.2002 | 15 days | В                                     | 3    |
| 09.04.2002 | 24.04.2002 | 15 days | С                                     | 1    |
| 09.04.2002 | 24.04.2002 | 15 days | D                                     | 12   |
| 02.05.2002 | 26.06.2002 | 55 days | E                                     | 1    |
| 02.05.2002 | 26.06.2002 | 55 days | F                                     | 23   |
| 02.05.2002 | 26.06.2002 | 55 days | G                                     | 21   |
| 02.05.2002 | 26.06.2002 | 55 days | Н                                     | 30   |

#### Tab 10 a): Experiment with Laboratory blank filters

- A, E = in the weighing laboratory at 22°C, 50%
- B, F = in the weighing laboratory at 22°C, 50% in plastic box
- C, G = in a stove at  $40^{\circ}$ C
- D, H = in a stove at  $40^{\circ}$ C in plastic box

b) Difference between the weighed laboratory blank mass during the study and the beginning of the study. Statistics of the 4 laboratory filters which were weighed 558 times during 20 months. Mass in  $\mu$ g.

| mean | median | minimum | maximum | std |
|------|--------|---------|---------|-----|
| 1.4  | 2      | -7      | 8       | 2.2 |

# 10 Figures



#### Fig. 1: Map of Europe with the 21 ECRHS centres.

### Fig. 2: Basel PM<sub>2.5</sub> sampler and other equipment



## Fig. 3:Predetermined measurement schedule

| Daily C                                    | Concent                               | ration | S:      |          |         | 24    | hour          | s (we                               | ekda  | y) an<br>(00:0 | d 48 l<br>0 h - 2 | 1 <b>0urs</b><br>4:00 h | (wee     | ekend | I)            |               |              |                         |
|--|---------------------------------------|--------|---------|----------|---------|-------|---------------|-------------------------------------|-------|----------------|-------------------|-------------------------|----------|-------|---------------|---------------|--------------|-------------------------|
| Month                                      | ly Mean                               | Conc   | entra   | ation:   |         | 5 v   | veeko         | days a                              | and 1 | wee            | kend              |                         |          |       |               | $\rightarrow$ | 7 days       | (168 hours)             |
|  |                                       |        |         |          |         |       |               |                                     |       | dist           | ribute            | ed ov                   | er 14    | days  |               |               |              |                         |
| Winter                                     | Mean C                                | once   | ntrati  | on:      |         | 4 N   | <i>l</i> onth | nly Me                              | ean C | once<br>Nov    | ntrati<br>00 - Fo | ons<br>eb 01)           |          |       |               | $\rightarrow$ | 28 days      | (672 hours)             |
| Summ                                       | er Mean                               | Cond   | centra  | ation:   |         | 4 N   | <b>/</b> onth | nly Me                              | ean C | once<br>(Mav   | ntrati            | ons                     |          |       | $\rightarrow$ | 28 days       | (672 hours)  |                         |
| Annua                                      | l Mean (                              | Conce  | entrat  | ion:     |         | 12    | Mon           | thly N                              | lean  | Conc           | entra             | tions                   |          |       | $\rightarrow$ | 84 days       | (2016 hours) |                         |
| Daily r                                    | neasure                               | ment   | sche    | me o     | f the   | mont  | hs No         | ovem                                | ber 2 | 000 -          | Febr              | uary                    | 2001     |       |               |               |              |                         |
|  | Nov                                   | 6      | 7       | 8        | 9       | 10    | 11            | 12                                  | 13    | 14             | 15                | 16                      | 17       | 18    | 19            | _             |              |                         |
|  | Dec                                   | 4      | 5       | 6        | 7       | 8     | 9             | 10                                  | 11    | 12             | 13                | 14                      | 15       | 16    | 17            | _             |              |                         |
|  | Jan                                   | 8      | 9       | 10       | 11      | 12    | 13            | 14                                  | 15    | 16             | 17                | 18                      | 19       | 20    | 21            | _             |              |                         |
|  | Feb                                   | 5      | 6       | 7        | 8       | 9     | 10            | 11                                  | 12    | 13             | 14                | 15                      | 16       | 17    | 18            | _             |              |                         |
|  |                                       | Мо     | Tu      | We       | Th      | Fr    | Sa            | Su                                  | Мо    | Tu             | We                | Th                      | Fr       | Sa    | Su            |               |              |                         |
|  |                                       |        |         |          |         |       |               |                                     |       |                |                   |                         |          |       |               | sha           | ded: date of | measurement             |
| Daily n                                    | neasure                               | ment   | sche    | me o     | f the   | whole | e stu         | dy Ju                               | ne 20 | 00 - I         | Nover             | nber                    | 2001     |       |               |               |              |                         |
| June 20                                    | 00                                    | 1      | 2, 14,  | 16, 20,  | 22, 24  | 4/25  | Ja            | nuary 2                             | 2001  |                | 8, 10             | ), 12, 1                | 6, 18, 2 | 20/21 |               | July 2001     | 9,           | 11, 13, 17, 19, 21/22   |
| July 200                                   | 0                                     | 1      | 0, 12,  | 14, 18,  | 20, 22  | 2/23  | Fe            | bruary                              | 2001  |                | 5, 7,             | 9, 13,                  | 15, 17,  | /18   |               | August 2001   | 13           | , 15, 17, 21, 23, 25/26 |
| August 2                                   | 2000                                  | 7      | , 9, 11 | , 15, 17 | 7, 19/2 | 0     | Ma            | arch 20                             | 01    |                | 5, 7,             | 9, 13,                  | 15, 17,  | /18   |               | September 20  | 01 10        | , 12, 14, 18, 20, 22/23 |
| September 2000 11, 13, 15, 19, 21, 23/24 A |                                       |        |         |          |         |       | Ap            | oril 200                            | 1     |                | 16, 1             | 8, 20,                  | 24, 26   | 28/29 | )             | October 2001  | 15           | , 17, 19, 23, 25, 27/28 |
| October                                    | October 2000 9, 11, 13, 17, 19, 21/22 |        |         |          |         |       |               |                                     | 1     |                | 7, 9,             | 11, 15                  | , 17, 19 | 9/20  |               | November 200  | 01 12        | , 14, 16, 20, 22, 24/25 |
| Novemb                                     | November 2000 6, 8, 10, 14, 16, 18/19 |        |         |          |         |       |               | June 2001 11, 13, 15, 19, 21, 23/24 |       |                |                   |                         |          |       |               |               |              |                         |
| Decemb                                     | December 2000 4, 6, 8, 12, 14, 16/17  |        |         |          |         |       |               |                                     |       |                |                   |                         |          |       |               |               |              |                         |



### Fig. 4: Correlation diagrams of daily concentrations AC versus AS



#### Fig. 5: Concentration ratios AS/AC



Fig. 6: Correlation between AS/AC and a) AS and b) AC, respectively (matched days only).



#### Fig. 7: Correlation between PM<sub>2.5</sub> in Turin and Pavia for a) all concentrations and b) winter concentrations only.



#### Fig. 8: Correlation between PM<sub>2.5</sub> in Pavia and Verona and Turin and Verona, respectively.

Fig. 9:Correlation between PM<sub>2.5</sub> in Pavia and Verona and Turin and Verona, respectively. WINTER (Nov 00 - Jan 01, Feb in Verona is missing)



Fig. 10 a)-c):Boxplots of daily PM2.5 mass concentrations measured at 21 ECRHS centres a) during a 12 month period in 2000 and 2001 b) during the four winter months, and c) during the four summer months. The box contains 50% of all measurements, the black line represents the median value. The included sampled hours are given in Tab. 1 as % of planned hours (100% = 168 hours per month). Sorted by annual mean.



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#### Fig. 10 b)





Fig. 11: Monthly PM<sub>2.5</sub> mass concentrations, sorted by annual mean.





Month

#### Fig. 12: continued...



Month



#### Fig. 13: Annual, winter and summer mean PM<sub>2.5</sub> mass



Fig. 14: Winter versus summer mean concentrations for a) PM<sub>2.5</sub> and b) for NO<sub>2</sub>

Fig. 15: Weekday (wd) mean versus Weekend (mean) mean  $PM_{2.5}$  mass concentrations in  $\mu$ g/m<sup>3</sup> and Spearman correlation coefficient.



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Fig. 16: Example of daily PM<sub>2.5</sub> mass concentrations in January 2001: 5 different ECRHS centers. Running time was 24 hours unless indicated in parenthesis (48 hours on weekends).



Fig. 17: Daily PM<sub>2.5</sub> mass concentrations in 6 different ECRHS centers showing different patterns in January and February 2001. Running time was 24 hours unless indicated in parenthesis (48 hours on weekends).







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## Fig. 21 a)-c): Correlation between monthly concentrations of NO<sub>2</sub> and PM<sub>2.5</sub> for each centre. Concentrations in µg/m<sup>3</sup>.



 $NO_2$ 

 $NO_2$ 





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# 11 Annexes

# 11.1 Important Instructions and Questionnaires

## 11.1.1 PM<sub>2.5</sub> Station Information Questionnaire

| 1.Country:                   |             |                          |                        |                         |                            | 2.City:                             |          |            |       |  |    |  |
|------------------------------|-------------|--------------------------|------------------------|-------------------------|----------------------------|-------------------------------------|----------|------------|-------|--|----|--|
| 3. PM2.5 St                  | ation nan   | ne / addres              | s:                     |                         |                            |                                     |          |            |       |  |    |  |
| 4.Altitude (m) a.s.l.:       |             |                          |                        |                         |                            | 5.Sampling height (m) above ground: |          |            |       |  |    |  |
| 6.Is the PM                  | 2.5 sampl   | er placed a              | nt an air o            | uality me               | asurement                  | station?                            |          | yes        | no    |  |    |  |
| 0010 010 1 11                | SuP         |                          |                        |                         |                            | 5                                   |          |            |       |  |    |  |
| 7.If yes, wh                 | ich other   | pollutants               | are meası              | ired there              | ?                          |                                     |          |            |       |  |    |  |
| SO2                          | CO          | NO2                      | NO                     | TSP                     | PM10                       | PM2.5                               | BS       | 03         |       |  |    |  |
|                              |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| 8.Please giv                 | e us the na | ame and the              | address o              | f the statio            | n and the r                | esponsible j                        | person:  |            |       |  |    |  |
| Name of the                  | station: _  |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| Address of t                 | he station: | :                        |                        |                         |                            |                                     |          |            |       |  |    |  |
| Name of the                  | responsit   | ole person:_             |                        |                         |                            |                                     |          |            |       |  |    |  |
| Phone:                       |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| Fax:                         |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| E-mail:                      |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| 9.Are there<br>10.if yes, pl | other PM    | I10 or PM2<br>us the nam | 2.5 measu<br>e and the | rement sit<br>address o | es in your<br>f the statio | city?<br>ons and the                | responsi | ble person | yes □ |  | no |  |
| Name of the                  | station: _  |                          |                        |                         |                            |                                     | -        | •          |       |  |    |  |
| Address of t                 | he station: | ·                        |                        |                         |                            |                                     |          |            |       |  |    |  |
| Name of the                  | responsib   | le person:_              |                        |                         |                            |                                     |          |            |       |  |    |  |
| Phone:                       | _           | _                        |                        |                         |                            |                                     |          |            |       |  |    |  |
| Fax:                         |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| E-mail:                      |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| Name of the                  | station: _  |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| Address:                     |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| Name of the                  | e responsib | le person:_              |                        |                         |                            |                                     |          |            |       |  |    |  |
| Phone:                       |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| Fax:                         |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| E-mail:                      |             |                          |                        |                         |                            |                                     |          |            |       |  |    |  |
| 11.Please                    | give us     | the name                 | and ad                 | dress of                | the near                   | est meteo                           | rologica | al station | :     |  |    |  |
| Name of t                    | he statio   | n:                       |                        |                         |                            |                                     |          |            |       |  |    |  |
| WP6 ECRHS II  |                         | FINAL R                | EPORT             |
|---|-------------------------|------------------------|-------------------|
| Address of the station:   |                         |                        |                   |
| Name of the responsible person                                      | n:                      |                        |                   |
| Phone:  |                         |                        |                   |
| Fax:  |                         |                        |                   |
| E-mail:   |                         |                        |                   |
| 12.Is the PM2.5 device expos  | ed to direct sunlight   | ? yes $\Box$ no $\Box$ |                   |
| if yes, estimate the average how                                    | urs per day of direct s | unlight:               |                   |
| Summer: hours per da  | ay Winter:              | hours per day          |                   |
| 13.PM2.5 Station classifi   | cation:                 |                        |                   |
| 13a)Station type:   | traffic                 | industrial $\Box$      | background $\Box$ |
| 13b)Type of zone:   | urban 🗆                 | suburban 🗆             | rural 🗆           |
| <b>13c)Caracterization of zone:</b> other major activity (specify): | residential             |                        | industrial □      |

| Explanations:       |  |
|---------------------|--|
| Station type:       |  |
| Traffic:            | Station used for monitoring traffic induced air pollution (right next to a street)                 |
| Industrial:         | Station used for monitoring industrial air pollution (on a industrial area)                        |
| Background:         | Station used for monitoring background air pollution levels. These stations can be located         |
|                     | inside (urban/background) as well outside (rural/background) cities                                |
| Type of zone:       |  |
| Urban:              | Station is located within the city   |
| Suburban:           | Station is located in the outskirts (fringe) of a city, or in small residential areas outside the  |
|                     | main city  |
| Rural:              | Station is located outside the city  |
|                     |  |
| Characterisatio     | n of zone:   |
| Please give us th   | e major activity in the representative area of the station (residential, commercial, industrial or |
| other). If there is | more than one major activity in the area, please mark each of them with a cross or a plus.         |

# 14.Major emission sources in PM2.5 station environment within 500 meters:

| Public power, co-generation and district heating     | yes 🗆 | no 🗆 |
|--|-------|------|
| Traffic  | yes 🗆 | no 🗆 |
| Commercial, institutional and residential combustion | yes 🗆 | no 🗆 |
| Industrial activities                                | yes 🗆 | no 🗆 |

#### If industrial activities, what kind of?

Other major emission sources (specify):

#### 15.Remarkable changes in PM2.5 station environment (type and date) during the

#### measurement period (for example: road work, bulding sites, changes in traffic volume,

| Туре: | Date: |
|-------|-------|
| Type: | Date: |

No remarkable changes  $\Box$ 

etc):

# About traffic arround the PM2.5 station:

| 16.Distance to the near | rest street? | meters |
|-------------------------|--------------|--------|
|-------------------------|--------------|--------|

17.Description of this street:

| Highway      |        | High (>10'000 vehicles/day) □               | Wide (D/H*>1.5) □   |
|--------------|--------|---|---------------------|
| Main street  |        | Medium (2'000 – 10'000 vehicles/day) $\Box$ | Canyon (D/H*<1.5) □ |
| Side street  |        | Low (< 2'000 vehicles/day) $\Box$           |                     |
| Other type o | f stre | et (specify):                               |                     |

| * | D = Distance between axis of the street and the buildings |
|---|---|
|   | H = Heigh of the buildings at the roadside                |

| 18. How often do heav | y vehicles (e.g. | trucks/buses) | pass this street? |
|-----------------------|------------------|---------------|-------------------|
|-----------------------|------------------|---------------|-------------------|

| constantly $\Box$ | frequently | seldom 🗆 | never |
|-------------------|------------|----------|-------|
|                   |            |          |       |

#### 19.Street type within 100 meters radius

| (more than one | is possible) | (street with highest traffic volume within 100 m) |  |
|----------------|--------------|---|--|
| Highway        |              | High (>10'000 vehicles/day) □                     |  |
| Main street    |              | Medium (2'000 – 10'000 vehicles/day) $\Box$       |  |
| Side street    |              | Low (< 2'000 vehicles/day) $\Box$                 |  |
| No street      |              |   |  |

Other type of street (specify): \_\_\_\_\_

#### **20.Is there within 100 meter radius**

| Busstop       |                       |
|---------------|-----------------------|
| Traffic light |                       |
| Crossing      |                       |
| Railway       |                       |
| Road works/   |                       |
| Building site | Type and time period: |
|               |                       |

Other important things for PM2.5 measurement (specify):

21.Do you have any objective information about the traffic density for the above mentioned streets and/or for your city?

(if yes, please send us the material, or write down the address of the contact person)

#### 22. Maps and Pictures:

Please send us (complete original maps, no copies if possible):

- a) A map of your entire city (the area where the ECRHS population is supposed to live), showing the location of the PM2.5 monitoring station (marked with a red cross)
  Attention: Please mark the streets (within 5 km radius) with high traffic (more than 10`000 vehicles per day) with a yellow high lighter and industrial areas with a green high lighter.
- b) A map, showing the close environment (about 1 km radius) of the PM2.5 monitoring station (streets, industries, buildings)
  Attention: Please mark all streets with high traffic (more than 10`000 vehicles per day) with a yellow high lighter and industrial areas with a green high lighter.
- c) A few pictures of the PM2.5 measurement station (some overviews and some detailed pictures) to give us an idea about the very close environment of the measurement site (buldings, streets, etc.)
- d) A few pictures of the place where you change the filters

Comments: \_\_\_\_\_

#### This questionnaire was filled in by:

| Name:    |  |
|----------|--|
| Address: |  |
| Phone:   |  |
| Fax:     |  |
| e-mail:  |  |
| and      |  |
| Name:    |  |
| Address: |  |
| Phone:   |  |
| Fax:     |  |
| e-mail:  |  |
|          |  |

# 11.1.2 WP6: Quality Check at the Local Centre Centre: Date:

| Ouality inspector:               |  |
|----------------------------------|--|
| <b>C</b> ,                       |  |
| Present persons from the centre: |  |

| 1. How does the place look where the PM2.5-sampler stands?:                     |  |
|---|--|
| $\pi$ Air Monitoring Station $\pi$ tidy $\pi$ messy                             |  |
| $\pi$ Sampler stands stable $\pi$ not well fixed                                |  |
| $\pi$ protected against vandalism   |  |
| Comments:   |  |
|   |  |
| 2. Are there abnormal sources which produce dust?                               |  |
| $\pi$ no $\pi$ yes source:  |  |
| Comments:   |  |
|   |  |
| 3. What is the height of the NO <sub>2</sub> -Box with respect to the ground    | Should be 2 - 3 m above the ground and |
| level?  | higher than a balustrade.              |
| $\pi < 1.5 \text{ m}$ $\pi 1.5 \text{ m} - 2 \text{ m}$ $\pi > 2 \text{ m}$     |  |
| If the location is a balcony: Is the NO <sub>2</sub> -Box fixed higher than the |  |
| balustrade? $\pi$ yes $\pi$ no  |  |
|   |  |
| Comments:   |  |
|   |  |
| 4. How does the lab look where the filter and oil change is                     |  |
| performed?:   |  |
| $\pi$ Lab of the Air Monitoring Station   |  |
| $\pi$ tidy $\pi$ messy $\pi$ clean $\pi$ dusty/dirty                            |  |
| Comments:   |  |
|   |  |
| 5. Where are the unexposed Filters kept:  | The place should be clean, safe and at |
| $\pi$ Lab $\pi$ padded envelope $\pi$ closed box                                | room temperature.                      |
| $\pi$ elsewhere:  | Important: Not in the fridge!          |
|   | (may lead to condensation when         |
| Comments:   | exposed)                               |
|   |  |

| 6. Where are the exposed Filters kept:                            | Must be in a fridge at about 4°C.            |
|---|--|
| $\pi$ clean fridge $\pi$ dirty fridge $\pi$ not in a fridge       | Important: not below 0°C. If the fridge is   |
| $\pi$ T = ca. 4°C $\pi$ T = - 20°C $\pi$ T =                      | dirty and has chemicals inside, the filters  |
| $\pi$ closed box $\pi$ padded envelope                            | should be in an airtight closed box.         |
| Comments:   |  |
|   |  |
| 7. Where are the unexposed NO <sub>2</sub> -tubes kept:           | Should be in a fridge at about 4°C.          |
| $\pi$ clean fridge $\pi$ dirty fridge $\pi$ not in a fridge       | Important: not below 0°C.                    |
| $\pi$ T = ca. 4°C $\pi$ T = - 20°C $\pi$ T =                      | If the fridge is dirty and has chemicals     |
| $\pi$ closed box  | inside, the filters should be in an airtight |
| Comments:   | closed box.                                  |
|   |  |
| 8. Where are the exposed NO <sub>2</sub> -tubes kept:             | Should be in a fridge at about 4°C.          |
| $\pi$ clean fridge $\pi$ dirty fridge $\pi$ not in a fridge       | Important: not below 0°C.                    |
| $\pi$ T = ca. 4°C $\pi$ T = - 20°C $\pi$ T =                      | If the fridge is dirty and has chemicals     |
| Comments:   | inside, the filters should be in an airtight |
|   | closed box.                                  |
| 9. How do they cover the pump when it is running?                 |  |
| $\pi$ Plastic bag $\pi$ else                                      |  |
|   |  |
| Comments:   |  |
|   |  |
| <b>10.</b> (a) Why do they have to cover the pump? (b) Why should | (a) It is possible that the pump is not      |
| they not cover the pump airtight?                                 | waterproof.                                  |
| $\pi$ (a) OK $\pi$ (b) OK   |  |
| Comments:   | (b) The air has to leave the pump via the    |
|   | exhaust port.                                |
| <b>11.</b> How do they change the filter when it is raining?      | They should describe. Important: No          |
| $\pi$ umbrella $\pi$ a second person                              | water has to fall on the filter.             |
| $\pi$ else  |  |
| Comments:   |  |
|   |  |
|   |  |
|   |  |
|   |  |
|   |  |
|   |  |
|   |  |

| 12. PM10-Inlet:                 |           |          |   |
|---------------------------------|-----------|----------|---|
| Outside, clean and intact:      | π yes     | π no     |   |
| Screen, clean and intact:       | π yes     | π no     |   |
| Water jar, clean and intact     | π yes     | π no     |   |
| Water jar, well closed          | π yes     | π no     | Take away the water jar and after       |
| O-rings, clean and intact       | π yes     | π no     | inspection fix it well.                 |
| Stick-O-rings, greased          | π yes     | π no     | Stick: Should be greased                |
| Comments:                       |           |          |   |
| 13. Impactor:                   |           |          |   |
| Outside, clean and intact:      | π yes     | π no     |   |
| Inside, clean and intact:       | π yes     | πno      |   |
| O-rings, clean and intact:      | π yes     | πno      | Stick: Should be greased                |
| Stick-O-rings, greased          | π yes     | πno      | Screw: Should be not greased            |
| Screw-O-rings, greased          | π yes     | πno      |   |
| Thread intact:                  | π yes     | πno      |   |
| Comments:                       |           |          |   |
| 14. Impactor Cup Assembly       |           |          |   |
| Cup assembly, clean and intact  | π ves     | πno      |   |
| O-ring, clean and intact        | πves      | πno      |   |
| Stick-O-ring, greased           | π yes     | π no     | Stick: Should be greased                |
| Fibergl. Filter, in Oil         | π yes     | π no     |   |
| Filter covered totally with Oil | π yes     | π no     | Should covered totally                  |
| Bubbles in the Oil              | π yes     | π no     | There should be no bubbles, they should |
| Pipette clean                   | π yes     | π no     | take them away with a clean pipette     |
| Tweezers clean                  | π yes     | π no     |   |
| Comments:                       |           |          |   |
| 15. Filter Holder               |           |          |   |
| Outside, clean and intact:      | $\pi$ yes | π no     |   |
| Inside, clean and intact:       | π yes     | π no     |   |
| O-rings, clean and intact:      | $\pi$ yes | π no     |   |
| Screw-O-rings, greased          | $\pi$ yes | $\pi$ no | Screw: Should be not greased            |
| Thread between sections intact: | $\pi$ yes | π no     |   |
| Thread, to stub intact:         | $\pi$ yes | π no     |   |
| Comments:                       |           |          |   |
|                                 |           |          |   |

| 16. Mounting plate                  |           |      |   |
|-------------------------------------|-----------|------|---|
| Thread of Stub intact               | π yes     | π no |   |
| Screw-O-ring Stub, clean and intact | π yes     | π no | Screw: Should be not greased              |
| Stub inside clean                   | π yes     | π no |   |
| Valve can be opened by fingers      | $\pi$ yes | π no | Press with a finger and show this         |
| Rubber hose, clean outside          | π yes     | π no |   |
| Rubber hose, clean inside           | π yes     | π no |   |
| Rubber hose connected to stub       | π yes     | π no |   |
| Rubber hose connected to pump       | π yes     | π no |   |
| Comments:                           |           |      |   |
|                                     |           |      |   |
| 17. Pump                            |           |      |   |
| Hose adapter intact and clean       | $\pi$ yes | π no | Remove the hose adapter                   |
| Thread of Hose adapter intact       | $\pi$ yes | π no |   |
| Screw-O-ring of hose adapter        | π yes     | π no | Screw: Should be not greased              |
| Exhaust port is fixed               | π yes     | π no | Should be not possible to turn, otherwise |
| Comments:                           |           |      | it is not waterproof                      |
|                                     |           |      |   |
|                                     |           |      |   |

| 18. Leak Test by Fieldworker                      |           |          | The fieldworker demonstrates you at      |
|---|-----------|----------|--|
| Takes the Test Filter                             | $\pi$ yes | π no     | least the beginning of a leak test.      |
| Opens the Plastic Box carefully                   | $\pi$ yes | π no     |  |
| Checks if the Filter is intact, clean             | $\pi$ yes | π no     |  |
| Puts it in the F. Holder carefully                | $\pi$ yes | π no     |  |
| Screws the F. Holder on the stub <i>carefully</i> | $\pi$ yes | π no     |  |
| Calibrator stands rigidly                         | $\pi$ yes | $\pi$ no |  |
| Calibrator stands horizontally                    | $\pi$ yes | $\pi$ no | Charld have an bigh                      |
| Rubber hose has a kink                            | $\pi$ yes | $\pi$ no | Important to have single values          |
| Presses after each bubble ON                      | $\pi$ yes | $\pi$ no | There is too much soan in it gives wrong |
| Several bubbles by one press                      | $\pi$ yes | $\pi$ no | results                                  |
| Shows the Manometer                               | $\pi$ yes | $\pi$ no |  |
| Shows the Thermometer                             | $\pi$ yes | π no     |  |
| Fieldworker uses the manual                       | $\pi$ yes | $\pi$ no |  |
| Fieldworker understands what she/he does          | $\pi$ yes | π no     |  |
| Comments:   |           |          |  |
| 19. Test Filter                                   |           |          |  |
| Test Filter is intact                             | $\pi$ yes | π no     |  |
| Test Filter is clean                              | $\pi$ yes | π no     |  |
| Test Filter is grey/exposed                       | $\pi$ yes | π no     |  |
| Comments:   |           |          |  |

**Between the measuring periods:** The pump, the whole assembly (inlet, impactor, filter holder) should be on a clean and safe place in the lab. The stub and the rubber hose should be protected waterproof by a plastic bag.

## 11.1.3 PM<sub>2.5</sub> in Antwerp South and City

(Email sent to the working group in December 2002)

#### **Problems:**

- Not all data are measured at the same days (and hours) in Antwerp City (AC) and South (AS). Admittedly, the annual and winter mean concentrations can be determined, but not precise enough for comparing them with each other.

- No parallel data available until Jan 01 (only one sampler was available before)
- Dec. 00 is missing in AC
- In Jan, the variability of the concentrations was very large, and just at the days with very high concentrations, the pump stopped after different hours in AC and AS (due to overload), and didn't run in AS.

#### Action:

- As written in paper 1, the correlation between AC and AS is very good (see also Fig. 4 above in this report, first picture), missing values can be filled in, when concentration is available in one centre only. Also values based on less than the preset running hours can be replaced.

- As Fig. 5 (see report above) illustrates the ratios AS/AC is not always the same during the year. Fig. B shows also that these ratios do not depend on the absolute concentrations. Thus, a correction factor should not be a) one single factor for the whole year, and b) not dependent on the absolute concentration. The suggestions is, to take one correction factor for each period.

- In Fig. 6 (see report above), the correction factors for each period are given. The correction factor for November and December is the mean of the correction factors of October 01 and January 01. The correction factors of February, March and June will not be used since during these periods, no values are missing and all concentrations are based on 100% of the preset hours. (For details, see Table C)

Table A shows the monthly, winter and annual mean concentrations for the three casesa) corrected as described above,

b) involved all concentrations as they were measured in each centre without corrections as described above and

c) matched days only (same date and same running time only).

The deviations between the different calculations are small for the annual mean concentrations, but larger for the winter concentrations. But as already mentioned in the winter paper, the centres in Antwerp are extreme examples for such deviations. I do not expect such deviations in other centres.

Table B shows that the ratio AS/AC in the annual mean is 0.91. Since the pump flow can differ by 5% according to BGI, a part of the approx. 10% difference between the two centres is real. If this difference is due to different background levels or due to the fact that the pump in Antwerp City is too much influenced by the nearby traffic can not be explained.
(My opinion is that there is in fact a difference of 5-10% in the background levels, and probably more during winter (time of inversion) than during summer)

#### Suggestion:

All details of the problem will be explained in the EC-report. In the paper: we use the corrected values, with a short explanation in the paper with reference to the report.

# Table A:Monthly, winter and annual means of AS and AC, calculated fordifferent cases

| a)        | b)  | C)  | a)  | b)  | C)   |
|-----------|---|---|---|---|--|
| AS corr   | AS all  | AS  | AC corr   | AC all  | AC   |
| suggested |   | matched   | suggested   |   | matched  |
| for 2nd   |   | only  | for 2nd   |   | only   |
| paper     |   |   | paper   |   |  |
| 14.1      |   |   | 16.8  | 16.8  |  |
| 12.7      | 12.7  |   | 15.4  | 18.4  |  |
| 15.0      | 15.0  |   | 18.1  |   |  |
| 52.8      | 42.6  | 29.9  | 64.4  | 64.4  | 35.6   |
| 25.9      | 27.2  | 27.2  | 28.3  | 28.3  | 29.7   |
| 30.1      | 30.1  | 30.1  | 30.8  | 30.8  | 30.8   |
| 11.5      | 14.3  | 14.3  | 14.3  | 14.3  | 17.8   |
| 16.2      | 16.3  | 14.5  | 17.7  | 17.7  | 15.8   |
| 15.9      | 15.9  | 15.9  | 15.6  | 15.6  | 15.6   |
| 11.2      | 11.2  | 11.6  | 13.5  | 13.6  | 14.0   |
| 24.6      | 25.8  | 23.2  | 23.0  | 23.0  | 21.8   |
| 19.8      | 19.8  | 19.3  | 16.1  | 16.1  | 15.7   |
| 18.5      | 18.5  | 19.7  | 21.8  | 22.4  | 23.2   |
|           |   |   |   |   |  |
| ) 21.18   | 20.8  | 20.6  | 23.25   | 24.0  | 22.0   |
|           | 1.9   | 2.9   |   | -3.4  | 5.4  |
|           |   |   |   |   |  |
| 26.61     | 24.4  | 28.5  | 31.55   | 37.0  | 32.7   |
|           | 8.4   | -7.2  |   | -17.4   | -3.6   |
|           |   |   |   |   |  |
|           | a)<br>AS corr<br>suggested<br>for 2nd<br>paper<br>14.1<br>12.7<br>15.0<br>52.8<br>25.9<br>30.1<br>11.5<br>16.2<br>15.9<br>11.2<br>24.6<br>19.8<br>18.5<br>) 21.18 | a)       b)         AS corr       AS all         suggested       AS all         for 2nd       AS all         paper       AS all         14.1       AS all         12.7       12.7         15.0       15.0         52.8       42.6         25.9       27.2         30.1       30.1         16.2       16.3         15.9       15.9         16.2       16.3         16.2       16.3         16.2       16.3         17.9       15.9         18.5       18.5         19.8       19.8         19.8       19.8         221.18       20.8         226.61       24.4         8.4       8.4 | a)       b)       c)         AS corr       AS all       AS         suggested       matched       only         paper       only       paper         14.1       12.7       12.7         15.0       15.0       15.0         52.8       42.6       29.9         25.9       27.2       27.2         30.1       30.1       30.1         16.2       16.3       14.3         16.2       16.3       14.5         16.2       16.3       14.5         16.2       16.3       14.5         16.2       16.3       14.5         17.9       15.9       15.9         18.5       18.5       19.7         24.6       25.8       23.2         19.8       19.8       19.3         18.5       18.5       19.7         26.61       24.4       28.5         26.61       24.4       28.5         26.61       24.4       28.5 | a)         b)         c)         a)           AS corr         AS all         AS         AC corr           suggested         matched         suggested           for 2nd         only         for 2nd           paper         i         and           14.1         i         and           114.1         i         and           115.2         i         and           111.5         i         and           111.5         i         and           111.5         i         and           111.2         i         and           111.2         i         and           111.2         i         and | a)       b)       c)       a)       b)         AS corr       AS all       AS       AS       AC corr       AC all         suggested       matched       suggested       for 2nd       paper       paper         14.1       matched       for 2nd       paper       16.8       16.8         14.1       matched       suggested       16.8       16.8         14.1       matched       augested       16.8       16.8         15.0       15.0       15.0       18.1       18.4         15.0       15.0       27.2       27.2       28.3       28.3         30.1       30.1       30.1       30.8       30.8       30.8         11.5       14.3       14.3       14.3       14.3         16.2       16.3       14.5       17.7       17.7         11.2       11.2       11.6       13.5       13.6         14.3       14.3       14.3       14.3 |

# Table B:

|                 | AS/AC           | AS/AC                |
|-----------------|-----------------|----------------------|
|                 | ratio in winter | ratio in annual mean |
| a) corr         | 0.843           | 0.911                |
| b) all          | 0.658           | 0.864                |
| c) matched only | 0.873           | 0.935                |

# Table C: Details of the PM<sub>2.5</sub> concentrations in AS and AC

|            | Conc.     | conc. corr. | For paper  |         | Conc.                    | conc. corr. | For paper  |         |       |
|------------|-----------|-------------|------------|---------|--------------------------|-------------|------------|---------|-------|
|            | AS(µg/m³) | per month   | with corr. |         | AC ( µg/m <sup>3</sup> ) | per month   | with corr. |         |       |
| Expdate    |           | AS          | AS         | Exptime |                          | AC          | AC         | Exptime | ratio |
| 21.10.2000 |           | 21.7        | 21.7       |         | 25.9                     |             | 25.9       | 48      | 0.841 |
| 24.10.2000 |           | 12.0        | 12.0       |         | 14.3                     |             | 14.3       | 24      | 0.841 |
| 26.10.2000 |           | 11.7        | 11.7       |         | 13.9                     |             | 13.9       | 24      | 0.841 |
| 30.10.2000 |           | 9.4         | 9.4        |         | 11.1                     |             | 11.1       | 24      | 0.841 |
| 01.11.2000 |           | 8.4         | 8.4        |         | 10.0                     |             | 10.0       | 24      | 0.841 |
| 06.11.2000 | 17.2      |             | 17.2       | 24      |                          | 20.8        | 20.8       |         | 0.827 |
| 08.11.2000 | 12.5      |             | 12.5       | 24      |                          | 15.1        | 15.1       |         | 0.827 |
| 10.11.2000 | 11.6      |             | 11.6       | 24      |                          | 14.1        | 14.1       |         | 0.827 |
| 14.11.2000 | 16.5      |             | 16.5       | 24      |                          | 20.0        | 20.0       |         | 0.827 |
| 16.11.2000 | 11.5      |             | 11.5       | 24      |                          | 13.9        | 13.9       |         | 0.827 |
| 18.11.2000 | 9.8       |             | 9.8        | 48      |                          | 11.9        | 11.9       |         | 0.827 |
| 21.11.2000 |           | 11.6        | 11.6       |         | 14.1                     |             | 14.1       | 24      | 0.827 |
| 23.11.2000 |           | 15.0        | 15.0       |         | 18.1                     |             | 18.1       | 24      | 0.827 |
| 27.11.2000 |           | 16.5        | 16.5       |         | 20.0                     |             | 20.0       | 24      | 0.827 |
| 29.11.2000 |           | 17.7        | 17.7       |         | 21.4                     |             | 21.4       | 24      | 0.827 |
| 04.12.2000 | 12.4      |             | 12.4       | 24      |                          | 15.0        | 15.0       |         | 0.827 |
| 06.12.2000 | 16.0      |             | 16.0       | 24      |                          | 19.4        | 19.4       |         | 0.827 |
| 08.12.2000 | 10.0      |             | 10.0       | 24      |                          | 12.0        | 12.0       |         | 0.827 |
| 12.12.2000 | 7.8       |             | 7.8        | 24      |                          | 9.4         | 9.4        |         | 0.827 |
| 14.12.2000 | 10.5      |             | 10.5       | 24      |                          | 12.7        | 12.7       |         | 0.827 |
| 16.12.2000 | 24.2      |             | 24.2       | 48      |                          | 29.3        | 29.3       |         | 0.827 |
| 08.01.2001 | 19.9      |             | 19.9       | 24      | 29.6                     |             | 29.6       | 24      |       |
| 10.01.2001 | 29.3      | 29.0        | 29.0       | 15.9    | 35.6                     |             | 35.6       | 24      | 0.813 |
| 12.01.2001 | 20.0      |             | 20.0       | 24      | 24.2                     |             | 24.2       | 24      |       |
| 16.01.2001 | 49.7      |             | 49.7       | 24      | 53.0                     |             | 53.0       | 24      |       |
| 18.01.2001 | 141.3     | 129.6       | 129.6      | 11.5    | 159.5                    |             | 159.5      | 19.93   | 0.813 |
| 20.01.2001 |           | 84.8        | 84.8       |         | 104.4                    |             | 104.4      | 21.72   | 0.813 |
| 05.02.2001 | 14.3      |             | 14.3       | 24      | 14.0                     |             | 14.0       | 24      |       |
| 07.02.2001 |           | 14.4        | 14.4       |         | 15.3                     |             | 15.3       | 24      | 0.940 |
| 09.02.2001 |           | 12.3        | 12.3       |         | 13.1                     |             | 13.1       | 24      | 0.940 |
| 12.02.2001 | 13.9      |             | 13.9       | 24      | 19.0                     |             | 19.0       | 24      |       |
| 14.02.2001 | 32.3      |             | 32.3       | 24      | 30.7                     |             | 30.7       | 24      |       |
| 16.02.2001 | 43.8      |             | 43.8       | 24      | 56.1                     |             | 56.1       | 24      |       |
| 20.02.2001 | 19.0      |             | 19.0       | 24      | 21.5                     |             | 21.5       | 24      |       |

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| 22.02.2001 |      | 18.4 | 18.4 |      | 19.6 |      | 19.6 | 24    | 0.940 |
|------------|------|------|------|------|------|------|------|-------|-------|
| 24.02.2001 | 27.0 |      | 27.0 | 48   | 25.6 |      | 25.6 | 48    |       |
| 12.03.2001 | 11.3 |      | 11.3 | 24   | 14.0 |      | 14.0 | 24    |       |
| 16.03.2001 | 51.7 |      | 51.7 | 24   | 43.2 |      | 43.2 | 24    |       |
| 20.03.2001 | 17.8 |      | 17.8 | 24   | 20.4 |      | 20.4 | 24    |       |
| 22.03.2001 | 36.8 |      | 36.8 | 24   | 42.7 |      | 42.7 | 24    |       |
| 24.03.2001 | 29.1 |      | 29.1 | 48   | 32.4 |      | 32.4 | 48    |       |
| 28.03.2001 | 34.8 |      | 34.8 | 24   | 30.7 |      | 30.7 | 24    |       |
| 16.04.2001 |      | 6.2  | 6.2  |      | 7.8  |      | 7.8  | 24    | 0.799 |
| 18.04.2001 |      | 8.3  | 8.3  |      | 10.4 |      | 10.4 | 24    | 0.799 |
| 20.04.2001 | 12.0 |      | 12.0 | 24   | 14.7 |      | 14.7 | 24    |       |
| 24.04.2001 | 26.3 |      | 26.3 | 24   | 31.9 |      | 31.9 | 24    |       |
| 26.04.2001 |      | 8.5  | 8.5  |      | 10.6 |      | 10.6 | 24    | 0.799 |
| 28.04.2001 | 9.5  |      | 9.5  | 48   | 12.3 |      | 12.3 | 48    |       |
| 07.05.2001 | 8.7  |      | 8.7  | 24   | 9.1  |      | 9.1  | 24    |       |
| 09.05.2001 | 31.1 | 23.6 | 23.6 | 11.7 | 26.0 |      | 26.0 | 24    | 0.908 |
| 11.05.2001 | 18.1 | 17.1 | 17.1 | 22.6 | 18.9 |      | 18.9 | 24    | 0.908 |
| 15.05.2001 | 17.0 |      | 17.0 | 24   | 22.0 |      | 22.0 | 24    |       |
| 17.05.2001 | 7.4  |      | 7.4  | 24   | 11.5 |      | 11.5 | 24    |       |
| 19.05.2001 | 19.7 |      | 19.7 | 48   | 18.1 |      | 18.1 | 48    |       |
| 11.06.2001 | 16.0 |      | 16.0 | 24   | 11.0 |      | 11.0 | 24    |       |
| 13.06.2001 | 22.5 |      | 22.5 | 24   | 25.1 |      | 25.1 | 24    |       |
| 15.06.2001 | 16.7 |      | 16.7 | 24   | 19.4 |      | 19.4 | 24    |       |
| 19.06.2001 | 12.0 |      | 12.0 | 24   | 11.2 |      | 11.2 | 24    |       |
| 21.06.2001 | 12.0 |      | 12.0 | 24   | 11.9 |      | 11.9 | 24    |       |
| 23.06.2001 | 16.1 |      | 16.1 | 48   | 15.2 |      | 15.2 | 48    |       |
| 09.07.2001 | 13.0 |      | 13.0 | 24   | 14.5 |      | 14.5 | 24    |       |
| 11.07.2001 | 9.4  |      | 9.4  | 24   | 10.8 |      | 10.8 | 24    |       |
| 15.07.2001 | 7.7  |      | 7.7  | 24   | 10.6 |      | 10.6 | 24    |       |
| 17.07.2001 | 15.5 |      | 15.5 | 24   | 18.2 |      | 18.2 | 24    |       |
| 19.07.2001 | 8.7  |      | 8.7  | 24   | 10.5 | 10.6 | 10.6 | 20.35 | 0.827 |
| 21.07.2001 | 12.1 |      | 12.1 | 48   | 15.0 |      | 15.0 | 48    |       |
| 20.08.2001 | 34.9 | 12.8 | 12.8 | 9.6  | 11.9 |      | 11.9 | 24    | 1.077 |
| 22.08.2001 | 42.5 | 43.3 | 43.3 | 18.6 | 40.2 |      | 40.2 | 24    | 1.077 |
| 24.08.2001 | 31.3 |      | 31.3 | 24   | 31.0 |      | 31.0 | 24    |       |
| 28.08.2001 | 10.8 |      | 10.8 | 24   | 8.9  |      | 8.9  | 24    |       |
| 30.08.2001 | 44.2 |      | 44.2 | 24   | 40.8 |      | 40.8 | 24    |       |
| 01.09.2001 | 14.9 |      | 14.9 | 48   | 14.2 |      | 14.2 | 48    |       |

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| 10.09.2001 | 6.9             |          | 6.9  | 24       | 8.5  |          | 8.5  | 24       |       |
|------------|-----------------|----------|------|----------|------|----------|------|----------|-------|
| 12.09.2001 | 22.7            |          | 22.7 | 24       | 19.0 | 18.5     | 18.5 | 18.03    | 1.224 |
| 14.09.2001 | 17.7            |          | 17.7 | 24       | 14.1 |          | 14.1 | 24       |       |
| 18.09.2001 | 18.4            |          | 18.4 | 24       | 20.8 |          | 20.8 | 24       |       |
| 20.09.2001 | 22.5            |          | 22.5 | 24       | 12.8 |          | 12.8 | 24       |       |
| 22.09.2001 | 25.1            |          | 25.1 | 48       | 19.0 |          | 19.0 | 48       |       |
| 08.10.2001 | 7.0             |          | 7.0  | 24       | 9.7  |          | 9.7  | 24       |       |
| 10.10.2001 | 11.2            |          | 11.2 | 24       | 14.6 | 13.3     | 13.3 | 16.17    | 0.841 |
| 12.10.2001 | 25.1            |          | 25.1 | 24       | 28.2 |          | 28.2 | 24       |       |
| 16.10.2001 | 11.3            |          | 11.3 | 24       | 14.6 |          | 14.6 | 24       |       |
| 18.10.2001 | 35.2            |          | 35.2 | 24       | 44.2 |          | 44.2 | 24       |       |
| 20.10.2001 | 19.9            |          | 19.9 | 48       | 21.4 |          | 21.4 | 48       |       |
|            |                 |          |      |          |      |          |      |          |       |
|            |                 | annual   |      |          |      | winter   |      |          |       |
|            |                 | hours    |      | days     |      | hours    |      | days     |       |
|            |                 | included |      | included |      | included |      | included |       |
|            | corr            | 1975     |      | 82.3     |      | 642      |      | 26.7     |       |
|            | matched<br>only | 1320     |      | 55       |      | 216      |      | 9        |       |

#### 11.1.4 PM<sub>2.5</sub> in Verona

(Email sent to the working group in December 2002)

#### **Problems:**

- not enough data available according to QC to determine an annual and a winter mean concentration

- No data from Feb May 01
- Sometimes only 2 to 3 concentrations per month available

- It was not possible to check the running time of the pump for the available filters according to QC, with Sep 00 as exception.

- We have to mention in the paper that we don't have a valuable annual and winter mean concentration for Verona.

#### Action:

- Data cleaning of the available data can be done with the TSP and  $PM_{10}$  (in June and July 01 only) data from Verona (received from the local authorities), see Fig. A. (Limitations: The comparison of absolute values of TSP and  $PM_{2.5}$  (and  $PM_{10}$ ) is not possible as Figs. B and C show.)

- The correlation diagram in Fig 7 (see report above) shows that the correlation between the concentrations of Pavia, and Turin is with  $R^2 = 0.75$  (N = 68) high (winter only  $R^2 = 0.53$ , N = 23). Since all three cities are located in the plain of the river Po and the distance between Pavia and Turin (120 km) is only hardly smaller than between Pavia and Verona (150 km, distance Turin - Verona 260 km) it can be expected that the weather influence in Verona is the similar to that in Turin and even more to that in Pavia (as we already demonstrated in the first paper "methods and winter data"). Thus, the correlation between Pavia/Turin and Verona could be expected to be fair, too.

Indeed, Fig. 8 (see report above) shows that the correlation between Pavia and Verona ( $R^2 = 0.49$ ) and Turin and Verona ( $R^2 = 0.59$ ) is fair. Fig. 9 (see report above) shows the same

correlation diagrams but with the winter concentrations only (PA vs. VE:  $R^2 = 0.42$ , TU vs. VE:  $R^2 = 0.68$ ).

#### Suggestion:

From the correlation between Verona vs. Turin and Pavia, respectively, an annual and winter mean concentration can be estimated:

| Winter mean:   |   |                       |
|--|---|-----------------------|
| From <b>Pavia</b> :<br>(R <sup>2</sup> = 0.42, N = 11) | $x = (55.3 \ \mu g/m^3 - 17.4 \ \mu g/m^3) / \ 0.566 = 67.0 \ \mu g/m^3$  | Verona winter         |
| From <b>Turin</b> :<br>(R <sup>2</sup> = 0.68, N = 10) | x = ( <b>69.2 <math>\mu</math>g/m<sup>3</sup> - 14.8 <math>\mu</math>g/m<sup>3</sup>) / 0.872 = <b>62.3 <math>\mu</math>g/m<sup>3</sup></b></b> | Verona winter         |
| Mean   | <u>64.7 µ</u>   | $ng/m^{3}$ (sd = 3.3) |

```
Annual mean:From Pavia:<br/>(R^2 = 0.49, N = 27)x = (35.4 \ \mu g/m^3 - 13.0 \ \mu g/m^3) / 0.577 = 38.8 \ \mu g/m^3Verona annualFrom Turin:<br/>(R^2 = 0.59, N = 25)x = (55.3 \ \mu g/m^3 - 17.3 \ \mu g/m^3) / 0.800 = 34.6 \ \mu g/m^3Verona annualMean\underline{36.7 \ \mu g/m^3} (sd = 3.0)
```

All details of the problem will be explained in the EC-report.

In the paper: We use the above estimated mean values, with a short explanation in the paper with reference to the report. We point out that in an health analysis these values may be used with caution.

### LOCATIONS

#### Verona:

Corso Milano (CM) and San Giacomo (SG) are "traffic-urban" and Torricelle (TC) and Cason (CS) are "background- rural" stations.

 $PM_{2.5}$  was measured at CM, Sept 00 - July 01  $_{(Feb\mathchar`-May\$ 

 $PM_{10}$  was measured at CM and SG , April 01 - July 01, gravimetric TSP was measured at all four stations, beta-meter, every 2 hours



#### Pavia and Turin:

Location are comparable to CM and SG in Verona

PM<sub>2.5</sub> was measured from Sep. 00 - Aug 01 at seven days per month, gravimetric

## LIMITATIONS AND PROBLEMS IN VERONA:

#### - PM<sub>2.5</sub>: From Oct 00 - end of measuring:

No pump data were downloaded. The fieldworker wrote down what was written on the pump.

# > No objective control was possible for the exact pump running time and thus also assignment of the filters less reliable

#### - PM<sub>2.5</sub>: From Feb 01 - May 01:

The pump was defect and a replacement pump was used. The replacement pump (Zambelli pump) pumped not at a controlled flow, but only for a mean flow of 16.67  $I/m^3$ . Thus, the cut-off diameter of the particles were not the same during the pumping, sometimes larger sometimes smaller than 2.5  $\mu$ m

# > These filters cannot be used for the $\text{PM}_{2.5}$ mass, but probably for black smoke and elements

#### - PM<sub>10</sub> data:

Not enough data available for good comparison  $PM_{2.5}$  vs.  $PM_{10}$ . But possible for June & July 01. See Fig. A.

#### - TSP data:

Inset of Fig. B shows that TSP data cannot be used regarding the absolute value. But the course (time pattern) can be used since in the TSP the course is mostly similar at all four locations, as illustrated in Figs. A, (B and C).

#### DATA CLEANING:

- Fig. A (2, Oct 00) shows that the high value in Oct 00 is wrong (probably due to too long running time and not dirt. Black smoke analysis and elemental analysis support this suspicion).

- Fig. A (10, June 01) shows that the assignment of the filters to the measuring dates for  $PM_{2.5}$  is not correct. Thus, it is not clear which filter belongs to the weekend measurement and if one filter is measured for 48 h at all. All June values cannot be used.





#### FINAL REPORT



#### **FINAL REPORT**

Figure B:  $PM_{2.5}$  and TSP (TRAFFIC) data from Verona in  $\mu$ g/m<sup>3</sup>.  $PM_{2.5}$  data before data cleaning. Inset left: ratio  $PM_{2.5}$ /TSP. Inset right: correlation  $PM_{2.5}$  vs. TSP.



#### **FINAL REPORT**

Figure C:  $PM_{2.5}$  and TSP (BACKGROUND) data from Verona in  $\mu$ g/m<sup>3</sup>.  $PM_{2.5}$  data before data cleaning. Inset right: correlation  $PM_{2.5}$  vs. TSP



# 11.1.5 Galdakao: Comparison of PM<sub>10</sub> and PM<sub>2.5</sub> Concentrations

We measured in winter low  $PM_{2.5}$  concentrations in Galdakao with respect to the summer. This is in contrast to all other centres. In order to be sure, a comparison between  $PM_{10}$  measured in Bilbao (data received from N. Muniozguren).  $PM_{10}$  was measured at Bilbao which is 17 km away from Galdakao and is not included in the study area. It is located in the roof of the building where N. Muniozguren works (Public Health) in Bilbao. Height of about 10 meters. It is used as traffic station, with medium traffic. The building is surrounded by 3 medium traffic streets at a distance of 2m, 2m and 10m, respectively. The measuring principle is a gravimetric method. The table below gives the characteristics of the  $PM_{2.5}$  station in Galdakao. The other figures and tables below are not commented. But we concluded, together with the literature (Viana, 2003), that our findings are correct.

| Study centre<br>Name of the station | Altitude (m a. s. l.) | Sampling height [meters above<br>ground] | Site = official measurement station | Other PM measurements in city | Type of Zone <sup>8</sup> | Characterisation of Zone <sup>c</sup> | Emission source within 500m <sup>b</sup> | Distance to nearest street | Type of nearest street <sup>E</sup> | Traffic volume on nearest street <sup>F</sup> | Wide/Canyon (nearest street) <sup>G</sup> | Frequency of heavy traffic <sup>H</sup> | Street type within 100m <sup>E</sup> | Highest traffic vol. within 100 $m^{F}$ | Relevant sites within 100m <sup>l</sup> | Objective traffic info available |
|-------------------------------------|-----------------------|--|-------------------------------------|-------------------------------|---------------------------|---------------------------------------|--|----------------------------|-------------------------------------|---|---|---|--------------------------------------|---|---|----------------------------------|
| Galdakao, Hospital de Galdakao      | 60                    | 14*                                      | no                                  | PM <sub>10</sub> (in Bilbao)  | r                         | r                                     | no                                       | 50                         | S                                   | Ι   | w   | S                                       | S                                    | I                                       | b;c                                     | n?                               |









Pattern of monthly mean concentrations is the same. Correlation diagram of monthly mean concentrations shows a fair correlation.





| data month    | PM <sub>2.5</sub> -Galdakao* | PM <sub>10</sub> -Bilbao* | months (number) | PM <sub>2.5</sub> /PM <sub>10</sub> |  |
|---------------|------------------------------|---------------------------|-----------------|-------------------------------------|--|
| date, month   | in µg/m³                     | in µg/m³                  | months (number) |                                     |  |
|               |                              |                           |                 |                                     |  |
| June2000      | 20.7                         | 63.8                      | 6               | 0.32                                |  |
| July2000      | 17.4                         | 41.4                      | 7               | 0.42                                |  |
| August2000    | 23.9                         | 52.25                     | 8               | 0.46                                |  |
| September2000 | 30.9                         | 72.8                      | 9               | 0.42                                |  |
| October2000   | 22.7                         | 62.75                     | 10              | 0.36                                |  |
| November2000  | 14.9                         | 42                        | 11              | 0.35                                |  |
| December2000  | 7.5                          | 28                        | 12              | 0.27                                |  |
| January2001   | 10.0                         | 35.25                     | 1.01            | 0.28                                |  |
| February2001  |                              |                           | 2.01            |                                     |  |
| March2001     | 13.6                         | 42.25                     | 3.01            | 0.32                                |  |
| April2001     | 12.1                         | 31.5                      | 4.01            | 0.38                                |  |
| May2001       | 15.6                         | 46.6                      | 5.01            | 0.34                                |  |
| June2001      | 26.6                         | 69.6                      | 6.01            | 0.38                                |  |
| July2001      | 17.2                         | 60.6                      | 7.01            | 0.28                                |  |
| August2001    | 24.2                         | 48.75                     | 8.01            | 0.50                                |  |
| September2001 | 17.8                         | 59.4                      | 9.01            | 0.30                                |  |

\* Mean concentration calculated according to the method described in this report.

| date     | PM2.5-   | PM10-   | ratio      |
|----------|----------|---------|------------|
|          | Galdakao | Bilbao  | PM2.5/PM10 |
|          | (µg/m³)  | (µg/m³) |            |
| 12.06.00 | 12.5     | 47      | 0.27       |
| 14.06.00 | 15.2     | 66      | 0.23       |
| 16.06.00 | 25.7     | 78      | 0.33       |
| 20.06.00 | 40.3     | 87      | 0.46       |
| 22.06.00 | 15.9     | 41      | 0.39       |
| 24.06.00 | 14.4     |         |            |
| 10.07.00 |          | 25      |            |
| 12.07.00 | 10       | 40      | 0.25       |
| 14.07.00 | 17       | 23      | 0.74       |
| 18.07.00 | 18.6     | 51      | 0.36       |
| 20.07.00 | 22.5     | 68      | 0.33       |
| 22.07.00 | 18.7     |         |            |
| 07.08.00 | 16.4     | 44      | 0.37       |
| 09.08.00 | 29       | 66      | 0.44       |
| 11.08.00 | 41.5     | 67      | 0.62       |
| 15.08.00 | 21.1     |         |            |
| 17.08.00 | 17.3     | 32      | 0.54       |
| 19.08.00 | 18.3     |         |            |
| 11.09.00 | 44.9     | 86      | 0.52       |
| 13.09.00 | 11.0     | 103     | 0.02       |
| 15.09.00 | 50.5     | 08      | 0.52       |
| 10.00.00 | 10.0     | 30      | 0.52       |
| 19.09.00 | 13.1     | 23      | 0.57       |
| 21.09.00 | 16.2     | 54      | 0.30       |
| 23.09.00 | 12.6     |         |            |
| 09.10.00 | 23.7     | 50      | 0.47       |
| 11.10.00 |          | 26      |            |
| 13.10.00 |          |         |            |
| 17.10.00 | 19.3     | 84      | 0.23       |
| 19.10.00 | 33.5     | 91      | 0.37       |
| 21.10.00 | 14.2     |         |            |
| 06.11.00 | 5        | 16      | 0.31       |
| 08.11.00 | 7.9      | 38      | 0.21       |
| 10.11.00 | 23.6     | 76      | 0.31       |
| 14.11.00 | 18       | 63      | 0.29       |
| 16.11.00 | 21.4     | 17      | 1.26       |
| 18.11.00 | 13.3     |         |            |
| 04.12.00 | 5.5      | 16      | 0.34       |
| 06.12.00 | 6.4      |         |            |
| 08.12.00 | 4.2      |         |            |
| 12.12.00 | 5.8      | 48      | 0.12       |
| 14.12.00 | 15.7     | 20      | 0.79       |
| 16.12.00 |          |         |            |
| 08.01.01 | 14.1     |         |            |
| 10 01 01 | 5        | 10      | 0.26       |
|          | J        | 19      | 0.20       |

|          | -    |     |      |
|----------|------|-----|------|
| 12.01.01 | 8.4  | 48  | 0.18 |
| 16.01.01 | 13.5 | 47  | 0.29 |
| 18.01.01 | 6.6  | 27  | 0.24 |
| 20.01.01 | 12.6 |     |      |
| 05.03.01 | 13.6 | 27  | 0.50 |
| 07.03.01 | 8.8  |     |      |
| 09.03.01 | 22.1 | 50  | 0.44 |
| 13.03.01 | 15.7 | 55  | 0.29 |
| 15.03.01 | 13.6 | 37  | 0.37 |
| 17.03.01 | 8    |     |      |
| 16.04.01 |      |     |      |
| 18.04.01 | 11.7 | 31  | 0.38 |
| 20.04.01 | 13.8 | 20  | 0.69 |
| 24.04.01 | 8.2  | 23  | 0.36 |
| 26.04.01 | 16.3 | 52  | 0.31 |
| 28.04.01 | 10.5 |     |      |
| 07.05.01 | 14.1 | 38  | 0.37 |
| 09.05.01 | 18.2 | 53  | 0.34 |
| 11.05.01 | 26.2 | 84  | 0.31 |
| 15.05.01 | 3.1  | 26  | 0.12 |
| 17.05.01 | 15.6 | 32  | 0.49 |
| 19.05.01 | 16.6 |     |      |
| 11.06.01 | 9.4  | 46  | 0.20 |
| 13.06.01 | 34.4 | 67  | 0.51 |
| 15.06.01 | 26.2 | 44  | 0.60 |
| 19.06.01 | 19.7 | 61  | 0.32 |
| 21.06.01 | 40.4 | 130 | 0.31 |
| 23.06.01 | 29.6 |     |      |
| 09.07.01 | 11.8 | 45  | 0.26 |
| 11.07.01 | 21.8 | 52  | 0.42 |
| 13.07.01 | 27.2 | 133 | 0.20 |
| 17.07.01 | 14   | 33  | 0.42 |
| 19.07.01 | 12.1 | 40  | 0.30 |
| 21.07.01 | 16   |     |      |
| 13.08.01 | 19.6 | 59  | 0.33 |
| 15.08.01 | 39.8 |     |      |
| 17.08.01 | 10.3 | 34  | 0.30 |
| 21.08.01 | 17.3 | 59  | 0.29 |
| 23.08.01 | 36.8 | 43  | 0.86 |
| 25.08.01 | 21.5 |     |      |
| 10.09.01 | 13.1 | 49  | 0.27 |
| 12.09.01 | 21   | 72  | 0.29 |
| 14.09.01 | 11.3 | 32  | 0.35 |
| 18.09.01 | 14.3 | 55  | 0.26 |
| 20.09.01 | 32.4 | 89  | 0.36 |
| 22.09.01 | 14.6 |     |      |
| 1        | -    | 1   |      |

# 11.2 Abstracts and Posters from Meetings



The follow-up of cohorts of adults from 29 European centers of the former ECRHS 1 (1980-92) investigates long-term effects of exposure to ambient air pollution. PM2-s is not routinely monitored in Europe. We measured PM2-s concentrations in 21 participating centers following a standardized protocol. Annual mean concentrations ranged from 4 µg/m<sup>3</sup> to 44 µg/m<sup>3</sup> and winter mean concentrations ranged from 5 µg/m<sup>3</sup> to 64 µg/m<sup>3</sup>. The range of PM2-s concentrations conductantion to the participating in ECRHS II is larger than in other transact cohorts conductantions on them terms of the of air softwine. ent cohort studies on long-term effects of air pollution

Introduction

The follow-up of cohorts of adults from 29 European centers of the former ECRHS I (1989-92) investigates long-term effects of exposure to ambient air pollution on incidence, course, and prognosis of respiratory diseases, in particular asthma and change of pulmonary function. To date, Europe has no common, standardized,

publicly available, air pollution monitoring network and PM<sub>25</sub> is not routinely monitored. Therefore, ECRHS collated existing fixed site monitoring data of the past 20 years, and

developed and implemented a PM2.5 monitoring schedule across 21 participating centers



To estimate the annual mean PM2.5 concentrations, following a standardized protocol across 21 European centers



PM2.5 annual and winter means



Annual and winter mean distributed over a wide differs from that of annual

#### Discussion

- PM<sub>2.5</sub> protocol was successfully applied and guality controlled by research groups speaking 9 languages
- PM<sub>2.5</sub> shows a wide range across Europe. This could be explained by varving emission density, meteorology, and topography
- Future comparisons using further indicators (NO2, black smoke, elemental analysis) will elucidate differences in exposure across European cities
- Influence of sampling location characteristics on measured levels of PM<sub>2.5</sub> and other pollutants will be further investigated

Seasonal pattern of PM<sub>2.5</sub> monthly mean concentrations

concentrations of PM<sub>2.5</sub> are ever range. Ranking of winter me



PM<sub>2.5</sub> day-to-day variability for winter and rest of the year \*



150 125 Verona ITA
 Erfurt GER
 Grenoble FRA
 Basel SWI
 Antwerp C BEL
 Antwerp S BEL 100 ¥., 50 25 6.1.0 20.1.01 21.9 12.1.01 6.2.01

variability is higher to have

#### Conclusions

1st standardized PM<sub>2.5</sub> data in a large European setting

- PM<sub>2.5</sub> annual and winter mean concentrations show a wide range within Europe
- Observed wide exposure range increas power to detect associations between air pollution and health
- Due to lack of common routine PM<sub>2.5</sub> data, ECRHS may contribute to European strategies for air pollution abatement

#### ERS 2002, Stockholm

**ECRHS II** 

UNI

BASEL

#### NO<sub>2</sub> Assessment and Comparison with PM<sub>2.5</sub> Measurements at 21 Study Centers of ECRHS II (European Community Respiratory Health Survey II)

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#### Abstract

We measured NO<sub>2</sub> (passive sampling) and PM<sub>2.5</sub> concentrations in 21 European cities to estimate 'background' exposure. Annual mean concentrations show a wide range from 5.5  $\mu$ g/m<sup>3</sup> in Iceland to more than 70  $\mu$ g/m<sup>3</sup> in southern European cities. Pearson correlation coefficient between NO<sub>2</sub> and PM<sub>1.5</sub> annual means is 0.79. However, correlations of monthly values differ remarkably between cities. The different patterns for the pollutants in different cities can be helpful to identify predictors for the measured exposure (sources, site characteristics, etc.). The relevance of characteristics of sampling location will be further investigated.

#### Introduction

Within ECRHS several indicators of exposure to ambient air pollution will be available (PM2.5, NO2, Black Smoke, chemical elements on PM2.5, and historic air quality data of the past 20 years). The combined data of several of these indicators allow to better characterize various, potentially health relevant aspects of population exposure. However, indicators less homogeneously distributed within a city (i.e. traffic indicators) will be more influenced by characteristics of sampling sites (e.g. traffic exposure) than others (PM2.5, S). Site characteristics though are only available in a descriptive form and are often imprecise. Comparisons of measured pollutant levels can provide additional quantitative information to assess influence of site characteristics on measured exposure levels.

#### Aims

To provide annual mean concentrations of  $NO_2$  measured parallel to  $PM_{2.5}$  at the same measuring sites, and to compare these two major indicators of urban air pollution exposure across the 21 ECRHS centers.

#### Methods

• Parallel exposure measures, same sites:

- NO<sub>2</sub> (Palmes Tubes, passive sampling)
- PM<sub>2.5</sub> (Basel Sampler/BGI, see other poster)
- Historic NO<sub>2</sub> data:
  - from 50 stations in ECRHS II centers
  - (100 European stations provided historic data)
  - Station characteristics (e.g. *traffic* or background) described by local agencies
- Additional exposure measures:
  - Elemental analysis (e.g. S, Ca, Pb, Cd, Br)
  - Black Smoke (reflection of PM<sub>2.5</sub> filters)
  - NO<sub>2</sub> at home (in some cities only)
- Possible approaches for interpretation:
  - $NO_2$  can be a traffic indicator
  - PM<sub>2.5</sub> is a background indicator
  - Seasonal pattern can indicate influence of meteorology, topography, and sources
  - Correlation of pollutants can indicate common sources of pollutants



Results

NO<sub>2</sub> winter mean concentrations are generally higher than the nnual mean. In Turin and Barcelona the winter effect is very mall compared to PM<sub>2.5</sub> (see other poster).



Annual means show a general association between  $NO_2$  and  $PM_{2.5}$  (r<sub>rearum</sub> = 0.78), however, associations differ strongly from city to city.

#### Discussion

 $NO_2$  and  $PM_{2.5}$  annual means correlate well (r=0.78), but comparison of  $NO_2$  and  $PM_{2.5}$  monthly values reveals different patterns for the centers.

Possible interpretations are:

- Good correlation and seasonal pattern: Meteorological and topographic factors dominate. (Pavia, Grenoble)
- Seasonal pattern for PM<sub>2.5</sub> but not for NO<sub>2</sub>: NO<sub>2</sub> traffic dominated. PM<sub>2.5</sub> background. (Turin, Barcelona)
- Good correlation within one city, no seasonal pattern, high NO<sub>2</sub> levels in city center: Local meteorology dominates. Central measuring site traffic influenced. (Antwerp).
- High correlation on low level: Background pollution (?). (Tartu)
- Seasonal pattern for NO<sub>2</sub> but not for PM<sub>2.5</sub>: Seasonal pattern for traffic (?). (Umea)

Use of (historic) data from multiple stations within one city can be useful to assess influence of location where traffic related pollutants are measured.

#### NO2 (•) vs. PM2.5 (+) monthly means of selected cities



fonthly mean concentrations show different patterns of NO<sub>2</sub> and PM<sub>2,5</sub>. In Pavia and Grenoble both pollutants show a strong casonal pattern. In Turin and Barcelona, NO<sub>2</sub> shows no seasonal pattern but  $PM_{2,2}$  does. In Antwerp, pollution levels are orrelated between both stations, but NO<sub>2</sub> levels in Antwerp (Dity are much higher. In Tartu, NO<sub>2</sub> and PM<sub>2,5</sub> correlate well on a ather low pollution level. In Umea NO<sub>2</sub> shows a seasonal pattern, but not PM<sub>2,5</sub>.

#### NO<sub>2</sub> current (00/01) and historic





The comparison of NO<sub>2</sub> concentrations from ECRHS II stations (00/01) with levels from 1997-99 from the same cities but different stations, shows the relevance of the location of the monitoring sites for a traffic related pollutant like NO<sub>2</sub>.

#### Conclusions

- Comparisons of NO $_2$  and PM $_{2.5}$  provide useful information on exposure situation and sampling locations
- NO<sub>2</sub> and PM<sub>2.5</sub> may indicate different aspects of air pollution exposure, across seasons, and within Europe
- Health effect analysis have to take different aspects of air pollution into account

#### Outlook

 We will further investigate influence of characteristics of sampling locations on measured exposure levels

## ISEE 2002, Vancouver

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