PERSONAL EXPOSURE ABD HEALTH IMPACTS SESSIONS

THE EFFECT OF SHORT-TERM CHANGES IN AIR POLLUTION ON RESPIRATORY AND CARDIOVASCULAR MORBIDITY IN NICOSIA, CYPRUS
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THE EFFECT OF SHORT-TERM CHANGES IN AIR POLLUTION ON RESPIRATORY AND CARDIOVASCULAR MORBIDITY IN NICOSIA, CYPRUS

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ABSTRACT

This study investigates the effect of daily changes in levels of PM_{10} on the daily volume of respiratory and cardiovascular admissions in Nicosia, Cyprus during 1995-2004. After controlling for long- (year and month) and short-term (day of the week) patterns as well as the effect of weather in Generalized Additive Poisson models, some positive associations were observed with all-cause and cause-specific admissions. Risk of hospitalization increased stepwise across quartiles of days with increasing levels of PM_{10} by 1.3% (-0.3, 2.8), 4.9% (3.3, 6.6), 5.6% (3.9, 7.3) as compared to days with the lowest concentrations. For every 10μg/m^3 increase in daily average PM_{10} concentration, there was a 1.2% (-0.1%, 2.4%) increase in cardiovascular admissions. With respects to respiratory admissions, an effect was observed only in the warm season with a 1.8% (-0.22, 3.85) increase in admissions per 10μg/m^3 increase in PM_{10}. The effect on respiratory admissions seemed to be much stronger in women and, surprisingly, restricted to people of adult age.

1. INTRODUCTION

To date, a substantial amount of research has shown adverse health effects – both increased hospitalization and excess mortality – of elevated ambient levels of air pollutants, and in particular particulate matter (PM). Typically, evidence on the effect of short-term changes comes from studies with a time-series design i.e. investigating the effect of day-to-day changes in concentrations of PM on daily counts of health events. More recently, evidence has been accumulating with the use of large multi-city studies or meta-analyses of several single-city time-series studies that combine and thus produce more robust estimates of the observed effects across several places. With the exception of a major European study (i.e. the APHEA project across 15-29 European cites (Aga et al, 2003)), the majority of studies have focused on US cities e.g. the National Morbidity, Mortality and Air Pollution Study (NMMAPS) (Dominici et al, 2005). Associations have not been investigated in small/medium size cities in the Eastern Mediterranean where climatic conditions (for example, sand storms from the Sahara desert) as well as socio-economic factors (including driving patterns and access to the health care) can vary considerably. This study aims to investigate associations between hospital admissions for all, respiratory and cardiovascular diseases and levels of PM_{10} in the city of Nicosia, Cyprus during the 10-year period 1995-2004.

2. METHODOLOGY

All cardiovascular (ICD codes I00-I52) and respiratory (ICD codes J00-J99) admissions with information on gender, age, ICD code and whether a Nicosia resident were obtained from the two public hospitals in Nicosia for the period 1 Jan 1995-30 Dec 2004. In addition, the daily volume of all-cause admissions in the same period was obtained from the Cyprus Statistical Services, aggregated in 8 age/sex strata. The Air Quality Section (Ministry of Labour) provided hourly measurements of PM_{10} from two stations: (a) located centrally on the roof of the Nicosia General Hospital and (b) in the rural location of Ayia Marina Xyliatou, 40km from Nicosia. While the latter is more representative of background pollution, measurements were only available from 1999 onwards. Daily averages of temperature (air and dew point), relative humidity, wind speed and barometric pressure (measured at 08:00 and 13:00) were provided by the Meteorological Services. These were based on hourly measurements of Thermohygrographs (i.e. instantaneous values) taken at the Athalassa Meteorological station (just south east from the centre of Nicosia).

Poisson regression was used to calculate percentage increase in daily admissions both across quartiles of increasing daily average levels of PM_{10} (to assess non-linearity) as well as per 10μg/m^3 linear increase. Due to the small number of daily cause-specific admissions, only sex-specific (i.e. all ages combined) or age-specific (younger and olden than 15 years of age) associations were examined. Generalized Additive Models (GAM) with natural splines were used to remove the long-term trend (df=40) as well as penalized splines to control for possible non-linear effects of the meteorological variables on the outcome. The final model controlled for long-term trend, temperature on the same day as well as the two previous days (lag 1 and 2) and relative humidity on the same day. In order to estimate the short-term patterns, day of the week was included in the models as an indicator variable. Only days with at least 12 hourly measurements were considered when calculating daily average concentrations of PM_{10}. Extreme values of PM_{10} are thought to be the result of sand storms from the Sahara dessert affecting Cyprus to a certain extent a few days every year. Analyses were thus repeated to (a) exclude days with average concentrations >150μg/m^3 to avoid outliers (n=25) influencing the estimation of linear effects and (b) model the separate effect of sand storms – defined as days with at least one hourly measurement greater than 300μg/m^3, or days in-between (total of 192 observations or 45 days). To investigate whether exposure to air pollutants can have effects over several days, same-day, lagged exposure (up to 2 days) as well as moving averages of 2- and 3-days exposure were considered. Data manipulation was performed in STATA 9.0 and non-parametric smoothing models were fitted in R 2.2.0.

3. RESULTS AND DISCUSSION

In the period under investigation, admissions in the two Nicosia hospitals nearly doubled. There has been a 3-fold increase in cardiovascular admissions with daily admissions rising from an average of 1 in the early years to 4 towards the end of the study period. For respiratory causes, admissions have increased in adults (aged>15) and decreased slightly in children (aged <15), remaining at around 4 daily admissions on average in much of the 10 years. *Table 1* shows summary statistics of the daily number of hospital admissions before and after restricting numbers to Nicosia residents. Combining all age/sex groups ensured that there would be at least 1 cardiovascular or respiratory admission in at least 85% of days. The low number of daily events meant that it was not uncommon for as many as 75% of days with no admissions if age- and sex-groups were considered separately.

Table 1: Summary statistics of (a) daily number of admissions, (b) PM_{10} concentrations in Nicosia Central and (c) meteorological factors between 1 Jan 1995-30 Dec 2004 (N=3652 days).

A. Hospital admissions (in parentheses, when restricting numbers to Nicosia residents only)								
	Total number (% Nicosia residents)	Mean	SD	Min	25%	Median	75%	Max
All-causes	178091	48.8	20.1	4	31	50	63	111
Cardiovascular	10896 (75%)	3.0 (2.2)	2.4 (1.9)	0 (0)	1 (1)	3 (2)	4 (3)	22 (11)
Respiratory	14827 (86%)	4.1 (3.5)	3.7 (3.1)	0 (0)	1 (1)	3 (2)	6 (5)	20 (18)
B. PM_{10} in Nicosia Central (μg/m^3)								
	Observations/ Days (%)	Mean	SD	Min	5%	Median	95%	Max
Hourly values	77788 (88.8%)	55.37	69.29	3.00	16.10	45.70	119.9	4965.95
Daily averages	3217 (88.1%)	55.42	42.69	5.00	25.85	50.57	90.81	1370.61
C. Meteorological factors								
Temperature	3652 (100%)	19.41	7.45	1.88	8.7	19.18	30.39	35.49
Rel. Humidity	3591 (98.3%)	64.99	13.95	16.58	38.46	66.21	85.79	96.50

Table 1 also shows the distribution of daily average concentrations of PM_{10} and the meteorological factors considered in the models. PM measurements were not available for as many as 354 days (10%). Restricted to days with at least 12 measurements, daily mean levels of PM_{10} ranged from 5.00 to 1370.61μg/m^3 (interquartile range: 40.04-64.11), levels comparable to southern European cities, and exceeding the European standard of 75μg/m^3 between 11-57 days a year. Concentrations of PM_{10} peaked between the hours of 7:00-9:00 and were lower during weekends, reflecting patterns of traffic in the centre of Nicosia – see *Figure 1*. Concentrations were higher during colder months. This, however, was not true in the case of the rural station where concentrations appeared lower during cold months, exhibiting a higher degree of agreement with levels in the centre during the warm season. Correlations between PM_{10} and the meteorological factors only ranged between -0.30 and 0.17, most strongly correlating with wind speed during the cold months.

As expected, respiratory admissions showed a strong cyclic pattern with higher admissions during the colder months. With the exception of possibly lower cardiovascular admissions during the summer months and weekends, there was not a strong seasonal or weekly pattern in cardiovascular admissions. Most striking was the large drop in respiratory admissions on Tuesdays on either side of high volume on Mondays and Wednesdays – see *Figure 1*. With the exception of the elderly (aged 65+), similar patterns were observed in all other age/sex groups. It was not clear whether this was a function reduced bed availability following a high volume of demand.

It might be indicative that people admitted on a Monday were more likely to be discharged by Wednesday than any other day while those admitted on a Wednesday were more likely to be discharged by the following Monday.

Figure 1: Weekly patterns of daily number of admissions and average levels of PM_{10} concentrations.

Cardiovascular admissions | Respiratory admissions | Daily average levels of PM_{10}

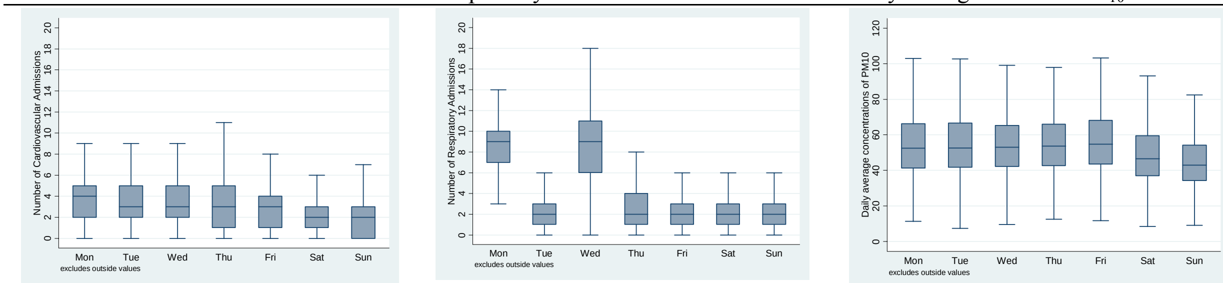

Table 2 shows the percentage increase in all- and cause-specific admissions per 10μg/m^3 increase in PM_{10} in Nicosia Central. Positive associations were observed with the risk of hospitalization. The effect appeared more pronounced in people of adult age (aged 15+ rather than children) and amongst men. Generally, associations were similar, if not smaller, when levels oh PM_{10} as measured in the rural station were used instead. While not statistically significant (due to the small number of events), similar, if not slightly stronger, effects were observed with admissions for cardiovascular causes. For every 10μg/m^3 increase in daily average PM_{10} concentrations, for instance, there was a 1.2% 95%CI (-0.1%, 2.4%) increase in cardiovascular admissions. Surprisingly, no overall effect was observed with total volume of respiratory admissions. However, the effect seemed to strengthen when analyses where repeated to exclude the 14% of all non-Nicosia residents among those admitted. This was not true in the case of cardiovascular admissions. Furthermore, the effect on respiratory admissions, if any, seemed to be restricted to people of adult age and, unlike cardiovascular admissions, was more pronounced in women. Finally, in contrast to the positive associations with hospital admissions observed with levels of PM_{10} on the same day, no positive effects were observed with levels of PM_{10} the 2 previous days. While lagged effects have commonly been observed with mortality in previous studies, it might not be as surprising that in the case of hospital admissions the strongest associations are observed with same day levels of pollution (Dominici et al, 2006). Perhaps, even more so in the case of Cypriot cities where easy access to health care may differ considerably from those in larger European or US cities.

Table 2: Percentage increase (and 95% CI) in all, cardiovascular and respiratory admissions per 10μg/m^3 increase in PM_{10} (restricted to days with daily average <150μg/m^3) after adjusting for long- (i.e. season) and short-term (i.e. day of the week) patterns as well as the effect of weather.

	All admissions[3]	Cardiovascular[4]	Respiratory	Cardiovascular + Respiratory
All age/sex groups	0.85 (0.55,1.15)	1.18 (-0.10,2.37)	0.10 (-0.91,1.11)	0.56 (-0.21,1.34)
Nicosia residents[1]		0.64 (-0.69,2.00)	0.25 (-0.84,1.36)	0.38 (-0.47,1.23)
Males	0.96 (0.54,1.39)	1.27 (-0.15,2.72)	0.10 (-1.32,1.53)[2]	0.63 (-0.34,1.62)
Females	0.74 (0.31,1.18)	0.99 (-1.11,3.14)	0.58 (-1.13,2.32)[2]	0.59 (-0.68,1.87)
Aged <15	0.47 (-0.13,1.08)		-0.46 (-1.99,1.10)[2]	
Aged >15	0.98 (0.63,1.33)		1.00 (-0.56,2.59)[2]	

Notes: [1 2] After restricting numbers to Nicosia residents, [3] Only aggregate numbers of all-cause admissions were available, thus it was not possible to restrict numbers to Nicosia residents, [4] Only includes people of adult age (15+) due to the rarity of cardiovascular events in those aged less than 15.

Stepwise increases in the risk of hospitalization were observed across quartiles of days with increasing levels of PM_{10} for either all-cause or cardiovascular admissions, with 5.6% (3.9, 7.3) and 9.2% (2.4, 16.5) increased admissions respectively in the quartile of days with the highest levels of PM_{10}. This was not the case among respiratory admissions, where the risk of adult admissions rose by 4.9% (-2.8, 13.3), 3.1% (-4.8, 11.6) and 6.1% (-2.1, 14.9) compared to the quartile of days with the lowest levels of PM_{10}. The extent of this non-linearity was further explored in models where the effect of an increase in PM_{10} on respiratory admissions was stratified by cold and warm months – see *Table 3*. Interestingly, there appeared to be some pronounced differential effects by season. While an increase in PM_{10} concentrations did not seem to have much of an effect on respiratory admissions during the colder months, some previously undetected associations were observed during the warm season, with effects as strong as 1.8% (-0.22,3.85) increase in respiratory admissions per 10μg/m^3 increase in PM_{10}. More adverse effects of PM_{10} on warm than cold days have previously been reported (Ren & Tong, 2006). Once again, the effect on respiratory admissions seemed to be restricted to people of adult age and much stronger in women. Finally, with respect to the separate effect of sand storms, admissions were 4.7% (0.1, 9.5), 7.6% (-10.9, 29.9), and 5.6% (-10.3, 24.2) higher on sand storm days for all, cardiovascular and respiratory admissions respectively. Furthermore, some smaller lingering effects were observed the day after a sand storm, particularly with respiratory admissions. Any inference from these associations is, however, limited due to the small number of sand storm days (n=45) in the 10-year period.

Table 3: Differential effects of 10μg/m^3 increase in PM_{10} on admissions for respiratory admissions during cold and warm months separately, after adjusting for long- and short-term patterns as well as the effect of weather.

	Percentage increase (and 95% CI) per 10 μg/m^3 increase in PM_{10}	
	Cold months[1]	**Warm months[2]**
All admitted	-0.33 (-1.47,0.82)	1.42 (-0.42,3.31)
Nicosia Residents	-0.22 (-1.45,1.02)	1.80 (-0.22,3.85)
Males	-0.16 (-1.76,1.46)	1.10 (-1.47,3.74)
Females	-0.26 (-2.18,1.70)	3.27 (-0.00,6.65)
Aged <15	-0.31 (-2.02,1.42)	-0.59 (-3.53,2.45)
Aged 15+	0.02 (-1.76,1.83)	3.89 (1.05,6.80)

Notes: [1 2] Cold months include Jan, Feb, Mar Apr, Nov and Dec, while warm months include all the rest.

4. CONCLUSIONS

Short-term exposure to PM_{10} increases the risk of hospitalization for all-cause, cardiovascular and respiratory admissions. At around 1% increase in daily admissions per 10μg/m^3 increase in PM_{10}, estimates in the city of Nicosia seem consistent with the size of effects seen across several European cities. (Le Tertre et al, 2002; Katsouyanni, 2003) An effect on respiratory admissions was observed only during the warm season and, surprisingly, only in adults. Unfortunately, the small number of events do not permit a finer cause-specific analysis so that the effect on specific respiratory events in children, e.g. the risk of asthma, can be investigated. Such effects can be further explored in a case-crossover design, an adaptation of the case-control design where each case serves as their own control and where the effect of experiencing an outcome on a certain day – and thus, concentration levels on that day – is compared to several other days that serve as the controls.

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AIR POLLUTION AND HOSPITAL ADMISSIONS BEFORE AND AFTER INDUSTRY CLOSURE IN THE LOWER HUNTER REGION, AUSTRALIA

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ABSTRACT

Adverse health effects of air pollution have been found in many studies in various parts of the world. This study investigated the impacts of closing a major source of pollutants, a steel works, in the Lower Hunter region, Australia. PM_{10}, NEPH, NO_2 and SO_2; and number of hospital admissions for all respiratory disease, CVD, COPD, asthma, IHD and total diseases were considered in the study. Mixed Model and GAM were employed in the statistical analysis. Inconsistent changes of pollutants and counts of hospital admissions in the whole study area challenged the publically-viewed significant role of BHP closure on health. However, specific results showed different effects in the closest and most distant Local Government Areas to the industry. In general, incidence of diseases decreased significantly in Newcastle, where the steel works was operating. Also, after closing the steel works, the most significant links between pollutants and health outcomes disappeared in this LGA.

Keywords: air pollution, hospital admission, industry impacts, mixed model, Poisson regression.

1. INTRODUCTION

Until the late past century, the Hunter Region, which is situated on the southeast coast of Australia in New South Wales, was known for air pollution caused by the local major chemical, steel making industries and mining. The Lower Hunter which is the sixth most populated region in Australia includes five LGAs (Local Government Area) with total population of about 470000. The climate of region is best described as mainly under sub-tropical influence which is generally warm in summer, mild in autumn and spring and cool in winter. The levels of air pollution are relatively low, so that, almost all the time, the pollutant concentrations have been reported within air-quality standards. The area has fully integrated industries, steel works, mining and rail and harbour activities. In October 1999, economic considerations led to the closure of the biggest local steel works industry, BHP Rod and Bar, located in Newcastle LGA near the coast. This industry was the principal source of SO_2, NO_2 and particulate matter (Bridgman *et al.* 1992). It was thought to endanger public health by emitting pollutants in the air, although, there was no clear evidence.

This study investigated the role of BHP closure on air quality and public health on a local scale. The specific targets were investigating the differences in air quality, the incidence of the diseases associated with air pollution and the link between air pollution and health variables before and after the closure of BHP in the region. The study period included 7 years, 3.5 years before and 3.5 years after BHP closure (1/1/1996 to 30/06/1999 and 1/1/2001 to 30/06/2004). The time between 30/06/1999 and 1/1/2001 was considered as phase out of the industry. The pollutants considered were PM_{10}, fine particles (NEPH), NO_2 and SO_2. The statistical analysis involved Mixed Model investigating the change of air pollution and health variables, and Generalized Additive Model (GAM) examining the relationship between levels of air pollution and daily hospital admissions.

2. METHODOLOGY

2-1. DATA COLLECTION AND MANIPULATION

Data on daily hospital admission for respiratory and heart diseases for Lower Hunter residents were obtained from the Centre for Clinical Epidemiology and Biostatistics (CCEB) at the University of Newcastle. We chose four age groups to examine, children (0-14 years), adults (15-64 years), the elderly (65+ years), and all ages. For total diseases and all respiratory and cardiovascular diseases (CVD) only all age group was considered. For COPD and IHD only 65+ years group was examined, and the analysis was restricted to 0-14 years and all age groups for asthma admissions. The count of admission for influenza and pneumonia was also included for modeling purposes, as a confounder.

Hourly average air pollution data were obtained from three air quality monitoring stations operating by the New South Wales Department of Environment and Conservation (NSWDEC). The pollutants considered in the study

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included PM_{10}, fine particles (NEPH), Nitrogen Dioxide (NO_2) and Sulphur Dioxide (SO_2). Hourly data on temperature and relative humidity were also collected from the same stations.

2-2.STATISTICAL ANALYSIS

Regarding the impacts of BHP closure, the analysis was applied for whole study area, Lower Hunter, the nearest LGA to industry, Newcastle; and the most distant LGA to BHP, Port Stephens. Corresponding to the study objectives, the study employed two models. The statistical approach used to examine the difference of air pollution variables and health outcomes before and after closing BHP utilized Mixed Model. The mixed model is a generalization of the standard linear model that examines statistical inferences along the data set. The model compute estimates of fixed effects and valid standard errors (SAS Institute Inc, 2004; Littlle, *et al.*, 2000). In this study, the model compares the mean of each variable, pollutants and health outcomes, over the same proportion of each pair of study period. In the model, each LGA was considered as an object which experienced two different situations in two different periods of time, before and after BHP closure. BHP closure, as main factor, was hypothetically associated with these differences. The potential confounding factors for counts of hospital admissions were seasonal variation, day of week and public holidays, population, and viral epidemics. For examining the pollutants, only seasonal variation was included in the model as a confounding factor. To adjust for population, incidence of each disease was considered in the analysis. The type of test is t-test for pair wise comparison adjustments with the P value = 0.05 and 95% confidence interval.

To investigate the associations between air pollutants and health outcomes before and after BHP closure, generalized additive model (GAM) was applied (Schwartz, 1993; Hagen, *et al.*, 2000). GAM extends Poisson regression to model the nonlinear effects of the covariates; using local nonparametric loess smoother to control for seasonal pattern and long-term trends. The study used a stepwise modeling strategy. Initially, for each health outcome, the potential confounders including temperature, humidity, day of week, public holidays and influenza epidemics were included in the model. When only the significant confounders remained, each of the pollutants was then added as a linear term to a Poisson regression model. Each age and location category of hospital admission data was analyzed in relation to each pollutant. All analyses were conducted using SAS statistical software (SAS Institute Inc, 2004).

3. RESULTS AND DISCUSION

3-1. DIFFERENCE OF POLLUTANTS

The averaged PM10 levels increased, and the other pollutants showed decrease after BHP closure. Table 1 shows the results of mixed model applied for the change of pollutants before and after BHP closure. The highest decrease was found for SO_2 concentrations, and NO_2 showed the least difference. With the exception of NO_2, the changes of the rest of pollutants were significant before and after BHP closure. Statistical analysis also found that, except for PM_{10}, the difference of pollutants became less significant toward the end of each study period.

Table 1: Difference of pollutants estimation in Lower Hunter, before and after BHP closure

	1996-99	**2001-04**	**Difference**	**P Value**
PM_{10}	18.206	20.608	+13.2%	0.0210
NEPH	0.274	0.246	-10.2%	<0.0001
NO_2	0.923	0.892	-3.4%	NS
SO_2	0.299	0. 178	-40.5%	<0.0001

3-2. DIFFERENCE OF DISEASES

Changes in disease admissions in the Lower Hunter Region: In Lower Hunter, apart from COPD, incidence of total and all other diagnosis dropped after BHP Closure. Although the estimations of incidence of total disease as well as CVD and IHD 65+ were lower in the period 2001-04, the results of mixed model revealed that the changes of these diagnosis groups were not significant. Statistical analysis also found non-significant change for all respiratory diseases despite an 11.7% decrease after closing BHP. The estimations of COPD 65+ were higher almost all the time in period 2001-04. It increased on the whole by 21.9% in period 2001-04; therefore, the mixed model found the change significant. Due to considerable decrease of all-age asthma as well as childhood asthma in the period after closing BHP, the mixed model found the differences significant.

Table 2 summarized the difference of diseases incidence estimations in the whole study location before and after the closure of BHP. As shown, the highest difference was observed for all-age asthma, and the lowest dissimilarity was found for all-age CVD admissions.

Table 2: Difference of disease estimation in Lower Hunter, before and after BHP closure

Location	Disease	1996-99	2001-04	Difference	P Value
LOWER HUNTER	Total Diseases	10.169	9.352	-8.0%	NS
	All Respiratory	4.252	3.753	-11.7%	NS
	COPD 65+	2.961	3.608	*+21.9%*	0.001
	Asthma	0.723	0.458	-36.7%	<0.0001
	Asthma 0-14	1.795	1.252	-30.3%	0.004
	CVD	5.916	5.598	-5.4%	NS
	IHD 65+	10.381	9.574	-7.8%	NS

Changes in diseases admissions in Newcastle and Port Stephens: Except for COPD, the incidence of total and all diagnoses dropped in both locations. The estimations of total diseases as well as CVD in Newcastle were apparently higher before BHP closure. But in Port Stephens, except for the first year of study period, there was no considerable change for incidences of these diagnoses. The statistical analysis, therefore, found the differences of total diseases and CVD significant in Newcastle and non-significant in Port Stephens. In spite the fact that the incidence of all respiratory disease and elderly IHD were lower after BHP closure, the mixed model resulted in non-significant changes in both LGAs. However, the decrease of both categories was noticeably higher in Newcastle than in Port Stephens. Increase of COPD 65+ and decreases of all-age asthma and childhood asthma were found statistically significant at both LGAs and more noticeable in Newcastle.

Table 3 shows the summary of diagnoses estimations in both LGAs before and after BHP closure. Of the diseases considered in the analysis, only COPD 65+ increased after BHP closure while the rest of disease categories decreased. Comparing before and after the closure of BHP, all age asthma in the Newcastle LGA showed the highest difference, and the lowest change was observed for all-age CVD in Port Stephens. In general, the change incidence of diseases was more significant in Newcastle than in Port Stephens.

Table 3: Difference of disease estimation in Newcastle and Port Stephens, before and after BHP closure

Location	Disease	1996-99	2001-04	Difference	P Value
NEWCASTLE	Total Diseases	10.287	8.722	-15.2%	0.0256
	All Respiratory	4.170	3.585	-14.0%	NS
	COPD 65+	2.671	3.656	*+36.9%*	<0.0001
	Asthma	0.714	0.429	-39.9%	0.0002
	Asthma 0-14	2.199	1.450	-34.1%	0.0031
	CVD	6.120	5.140	-16.0%	0.0122
	IHD 65+	9.489	7.948	-16.2%	NS
PORT STEPHENS	Total Diseases	9.984	9.120	-8.7%	NS
	All Respiratory	4.109	3.682	-10.4%	NS
	COPD 65+	3.243	4.264	*+31.5%*	0.0003
	Asthma	0.750	0.488	-34.9%	0.0002
	Asthma 0-14	1.652	1.048	-36.6%	0.0008
	CVD	5.870	5.432	-7.5%	NS
	IHD 65+	10.974	9.480	-13.6%	NS

Figure 1 compares the overall estimations of diseases in the whole study area (Lower Hunter), Newcastle and Port Stephens. As shown, total diseases and CVD incidences in Newcastle were higher than the average in Lower Hunter as well as in Port Stephens in 1996-99 period. After BHP closure, they changed dramatically and shifted to the lowest levels. All respiratory disease, childhood asthma and IHD 65+ decreased roughly in the same way in all locations. The decrease of all-age asthma and increase of COPD were more apparent in Newcastle. In comparison, except for childhood asthma, the differences of disease incidences in before and after BHP closure were more significant in Newcastle than in Port Stephens and Lower Hunter. Of seven disease categories, only the differences of all respiratory disease and IHD 65+ were not significant in Newcastle. In Port Stephens, only the changes of COPD 65+ and asthma categories were significant. The highest decrease of disease incidences was found for all age asthma in Newcastle, and the least difference was observed for CVD in Lower Hunter.

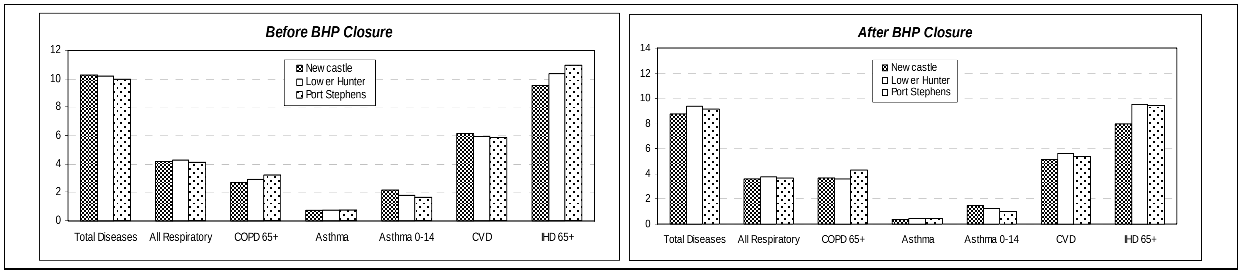

Figure 1: Incidence estimations of disease in Newcastle and Port Stephens, before and after BHP closure

3-3. POLLUTANTS EFFECTS

Before closing BHP, significant associations were found between total admissions and PM_{10} and fine particles in Newcastle. All respiratory in Lower Hunter, and CVD in Newcastle were statistically related to only NEPH. Elderly COPD was significantly associated with NEPH in all locations. There were significant links between childhood asthma and NO_2 in Lower Hunter; and NO_2 and PM_{10} in Newcastle. Elderly IHD was associated with PM_{10} and SO_2. The other links between health outcomes and pollutants appeared to be non-significant in the analysis. Except for COPD, the rest of significant associations disappeared after BHP closure. In period 2001-04, COPD was found to be related to almost all pollutants in Lower Hunter and Newcastle. In general COPD 65+ and all-age asthma showed the most and the least significant associations respectively.

4. CONCLUSIONS

BHP Rod & Bar was the major source of NO_2, SO_2 and particles in the area. Inconsistent changes in pollutant levels after closing BHP, particularly for PM_{10} and SO_2, implies the complications of impact of the industry on air quality in the region. Similar to other cities around the world, change in emissions source of pollutants, mainly because of increasing number of vehicles, has contributed in a different air pollution mixture (WHO, 2000). In whole study area, only COPD and asthma categories showed significant changes after closing BHP. Lack of local air quality monitoring stations did not allow the study to investigate the BHP impacts directly in each LGA; instead, the indirect effects (health outcomes) were examined. Ignoring COPD, the LGA results suggest some BHP impacts on public health. Significant decrease of total diseases, respiratory disease and CVD in Newcastle, where the industry was operating, which was higher than in Port Stephens as well as whole study location, might be related to BHP closure. In addition, the disappearance of links between pollutants and health variables after closing BHP, particularly in Newcastle, was consistent with these findings. Despite different methodologies, the results of this study were similar to the studies elsewhere.

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SOURCES INFLUENCING PM_{10} AND $PM_{2.5}$ LEVELS IN THREE EUROPEAN CITIES – IMPLICATIONS FOR POPULATION EXPOSURE.

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ABSTRACT

Although vehicle technology improvements are responsible for the reductions in primary particle emissions, both high PM_{10} and $PM_{2.5}$ levels have been observed in several European cities in recent years. High PM_{10} and $PM_{2.5}$ concentrations have persisted over periods of several days, resulting in exceedences of EU target values and affecting public health.
In this study, $PM_{10}/PM_{2.5}$ data from Athens (Greece), Madrid (Spain) and Birmingham (UK) have been analysed for relationships to other air pollutants (NO, NO_2, CO, O_3 and SO_2) and several meteorological parameters (wind velocity, temperature, relative humidity, precipitation, solar radiation and atmospheric pressure) during 2005. Principal component and regression analyses were used to quantify the contribution of both combustion and non-combustion sources to the observed $PM_{10}/PM_{2.5}$ levels. The contribution of non-combustion sources ranged substantially in the three examined cities. Positive correlations between PM_{10}, NO_x, CO were observed, while negative correlations between PM_{10}, O_3, wind speed and precipitation were found.

Keywords: Particulate matter, PM_{10}, $PM_{2.5}$, air quality; urban background, seasonal variability, non-combustion sources

1. INTRODUCTION

Particulate pollution is an issue of increasing public concern during the recent years. A large number of epidemiological studies have underlined the link between enhanced particle concentrations and daily excesses in mortality and morbidity (Dockery and Pope, 1994). In the past several authors have analysed air quality and meteorological data series to identify the factors influencing particulate pollution levels in Berlin (Lenschow et al, 2001), ten English cities (Harrison and Deacon, 1998), Helsinki (Kukkonen et al, 2005), Athens (Chaloulakou et al, 2003) etc. The objective of this work is to identify factors influencing $PM_{10}/PM_{2.5}$ levels in three major European cities, Athens (Greece), Birmingham (UK) and Madrid (Spain) with similar population size but different climate and topographic characteristics. The analysis presented includes the seasonal variability, the estimation of combustion and non combustion sources and the sources of particulate air pollution in stations having similar characteristics in the three cities.

2. METHODOLOGY

One year hourly data sets (2005) of PM_{10}, $PM_{2.5}$, NOx, NO, NO_2, SO_2, O_3 and CO from one traffic oriented monitoring site in each city among with local meteorological parameters (wind speed and direction, temperature, relative humidity, precipitation, atmospheric pressure, solar radiation) were analysed. The selected locations are: new likovrisi (ANL) in Athens, Bristol Road (BBR) in Birmingham and Paseo de Recoletos (MPR) in Madrid. All this sites are traffic oriented and could be described as areas with intense traffic conditions in the three cities. All monitoring data were subdivided in 2 periods the ‘cold’ (16 October-15 April) and ‘warm’ period (16 April-15 October), in order to disassociate the factors influencing particulate levels in different seasons (Vardoulakis and Kassomenos, 2007). Hourly data from the selected monitoring sites were averaged over 24-h when at least 70% of the hourly data were available for each day.
Initially a Pearson correlation analysis was attempted between PM_{10} and $PM_{2.5}$ concentrations paired with air quality concentrations of CO, NO_2, NO, SO_2, O_3. The same analysis was made between PM_{10} and $PM_{2.5}$ and the meteorological parameters. Principal Component Analysis (PCA) was also attempted to identify major air pollution sources at the selected monitoring locations through the aid of software package SPPS 14.0. Principal Component Analysis (PCA) with Varimax rotation was applied separately for cold and warm periods of the year in each monitoring site. Least square analysis was employed between daily PM_{10} and $PM_{2.5}$ (depended variables) and NOx and CO concentrations (independent variables) for the same seasons.

The regression analysis was revealed the relative contributions of combustion and non-combustion sources to the observed $PM_{10}/PM_{2.5}$ concentration levels in the traffic sites.

3. RESULTS AND DISCUSSION

The seasonal mean of PM_{10} concentrations is higher in Athens (exceeding the mean annual value of 40μgm^{-3}) following by Madrid and Birmingham. The similar concentrations of $PM_{2.5}$ are also higher, by far, than 10 μgm^{-3} (a possible, under discussion, limit for $PM_{2.5}$ in the European Union). The exceedances of the 50 μgm^{-3} 24h mean value for PM_{10} were 122 for ANL and 91 for MPR while the exceedances for the scheduled 25 μgm^{-3} 24h mean value for $PM_{2.5}$ were 111 for BBR, 132 for ANL and 143 for MPR (Table 1).

Table 1. Seasonal mean PM_{10}, $PM_{2.5}$ (μgm^{-3}), $PM_{2.5}/PM_{10}$

Site	PM_{10} Cold	PM_{10} Warm	$PM_{2.5}$ Cold	$PM_{2.5}$ Warm	$PM_{2.5}/PM_{10}$ Cold	$PM_{2.5}/PM_{10}$ Warm
BBR	23.42 (0)	21.78 (0)	15.13 (44)	13.89 (67)	0.65	0.63
ANL	49.61 (64)	48.07 (58)	24.35 (71)	22.01 (61)	0.49	0.48
MPR	38.12 (47)	38.22 (44)	24.02 (55)	31.07 (88)	0.63	0.81

The daily mean $PM_{10}/PM_{2.5}$ concentrations were correlated with the other gaseous pollutants during warm and cold seasons in all three sites (Table 2). Particles and CO concentrations present significant correlations in Madrid, while in Birmingham the correlations between particles and NO, NO_2 NOx are generally stronger. The relationship between particles and SO_2 are stronger in Madrid than Birmingham. The strong correlations especially in MPR during the whole year maybe due to the nature of the site and could come from both the domestic heating and the traffic. Correlations of particles in Athens with all primary pollutants were moderately high exceeding or being in the vicinity of 0.5 (with the reasonable exception of SO_2 during the warm period). Daily mean O_3 concentrations correlated negatively with $PM_{10}/PM_{2.5}$. This negative correlation may be explained by the reaction of O_3 with NO, which is a major sink of ozone. The weaker relationship obtained during the warm seasons is probably due to higher concentrations of natural and secondary particles.

Table 2. Pearson correlation coefficients between daily PM_{10}, $PM_{2.5}$ and gaseous pollutant concentrations averaged over cold and warm seasons in the three measuring sites used. $PM_{2.5}$ in brackets.

Site	CO		NO_2		NO		NOx		O_3		SO_2	
	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm
BBR	0.31 (0.34)	0.13 (0.23)	0.75 (0.72)	0.59 (0.6)	0.71 (0.74)	0.36 (0.36)	0.73 (0.75)	0.44 (0.44)	-0.32 (-0.49)	0.10[a] (-0.4[a])	0.28 (0.23)	0.12[a] (0.13)
ANL	0.66 (0.66)	0.48 (0.53)	0.55 (0.52)	0.5 (0.61)	0.68 (0.66)	0.55 (0.49)	0.69 (0.70)	0.6 (0.62)	-0.31 (-0.41)	-0.32 (0.1[a])	0.48 (0.53)	0.35 (0.39)
MPR	0.71 (0.67)	0.66 (0.70)	0.36 (0.27)	0.51 (0.41)	0.64 (0.63)	0.67 (0.72)	0.63 (0.60)	0.70 (0.71)	-0.4 (-0.32)	-0.32 (-0.19)	0.62 (0.43)	0.65 (0.67)

All correlations are statistically significant at 99% confidence interval (unless otherwise indicated)
[a]correlation non significant

A PCA analysis was used to identify 3-4 uncorrelated factors explaining around 90% of the total variance in the air quality data sets obtained in the three cities. The results varied substantially in the three cities reflecting the different traffic behaviour, traffic intensity and patterns, as well as the different climatic conditions. Specifically in Madrid a clear traffic factor (PC1) was found having high loadings for CO, NO and $PM_{2.5}$ accompanied by moderate loadings of PM_{10} during the cold season, while during the warm season this factor is accompanied by high loadings of SO_2. This factor explained 26-39% of the variance. The strong correlation of SO_2 indicates that during the warm season in the traffic stations the measured SO_2 is possibly due to traffic. PC2 indicated the photochemical activity presenting high loadings for O_3 and moderate loadings for NOx explaining 20-17% of the total variance. PC3 factor, having high loadings for PM_{10} and moderate loadings of $PM_{2.5}$, CO, indicated natural or other fugitive sources of particles. This factor explains 22-15% of the variance. A fourth factor (PC4) presented high loadings of NO_2 and moderate or high loadings of SO_2 explaining 34-16% of the total variance.

In Birmingham, PC1 presented high loadings for NO, NO_2, PM_{10}, $PM_{2.5}$ during cold and high loadings for CO and $PM_{2.5}$ during the warm season. PC2 had very high loadings for O_3 during cold and O_3 and SO_2 during the warm seasons. PC3 presented very high loadings for CO and moderate loadings for PM_{10}, $PM_{2.5}$ during cold and high loadings for PM_{10} during the warm season. Finally PC4 presents very high loadings for SO_2 during the cold and NO and NO2 during the warm season.

Table 3. Principal component loadings and variance explanation for air quality obtained in, Madrid, Athens and Birmingham.

	Cold seasons				Warm Seasons			
MPR	PC1	PC2	PC3	PC4	PC1	PC2	PC3	PC4
CO	**0.61**	0.38	0.26	0.60	**0.83**	0.28	0.32	0.22
NO	**0.61**	0.49	0.14	0.54	**0.83**	0.31	0.33	0.22
NO_2	0.09	0.23	0.05	**0.92**	0.27	0.06	0.20	**0.93**
O_3	-0.15	**-0.93**	-0.13	-0.28	-0.26	**-0.95**	-0.09	-0.06
SO_2	0.25	0.22	0.33	**0.85**	**0.86**	0.20	0.27	*0.27*
PM_{10}	0.47	0.17	**0.82**	0.25	0.30	0.21	**0.84**	0.30
$PM_{2.5}$	**0.89**	0.09	0.35	0.10	0.61	-0.07	**0.69**	0.06
Variance(%)	26.3	19.8	14.5	33.9	39.1	16.7	21.8	16.2

ANL	PC1	PC2	PC3	PC1	PC2	PC3
CO	**0.61**	**0.80**	0.17	**0.80**	0.48	0.19
NO	**0.67**	0.45	0.39	**0.73**	**0.60**	0.23
NO_2	0.22	**0.92**	0.11	**0.74**	0.48	0.37
O_3	-0.29	-0.21	-0.50	**-0.83**	-0.12	0.23
SO_2	0.20	**0.69**	**0.62**	0.25	**0.92**	0.16
PM_{10}	**0.90**	0.31	0.10	0.42	**0.70**	0.45
$PM_{2.5}$	**0.88**	0.06	0.30	0.32	0.23	**0.90**
Variance(%)	36.8	27.6	22.5	39.4	31.4	19.8

BBR	PC1	PC2	PC3	PC4	PC1	PC2	PC3	PC4
CO	0.08	0.36	**0.828**	0.09	**0.883**	0.15	0.13	0.33
NO	**0.88**	0.21	-0.25	-0.12	0.37	0.10	0.06	**0.89**
NO_2	**0.90**	0.11	0.03	0.06	0.38	0.47	0.10	**0.71**
O_3	-0.04	**-0.89**	-0.24	-0.07	0.22	**-0.85**	0.15	0.10
SO_2	0.01	0.07	0.07	**0.99**	-0.14	**0.86**	-0.16	-0.19
PM_{10}	**0.75**	0.32	0.41	0.06	0.09	0.22	**0.97**	0.08
$PM_{2.5}$	**0.80**	0.12	0.41	0.05	**0.86**	0.27	0.03	0.30
Variance(%)	39.9	15.8	16.4	14.6	26.8	26.3	14.5	22.4

For Athens, during the cold season the analysis isolates factors related with traffic related gaseous and particulate emissions (PC1), primary emissions (vehicular and fugitive industrial) related to suspension of coarse particles (PC2-zero loading on $PM_{2.5}$), and fuel oil combustion mainly for space heating (PC3-high loadings on SO_2). The first two components are of similar character also for the warm period. The association of PM_{10} with PC2 becomes more pronounced due to favouring climatic conditions, which invalidate the heating component, resulting at a third factor possibly associated with secondary fine particulate formation.

Table 4. Linear least square regression coefficients for the relationships [PM_{10}]=slopeX[NOx] + intercept and [$PM_{2.5}$]=slopeX[NOx] + intercept. $PM_{2.5}$ in brackets.

Site	Slope Cold	Slope Warm	Intercept Cold	Intercept Warm	R Cold	R Warm
BBR	0.110±0.008	0.083±0.013	14.56±0.775	16.445±0.880	0.733	0.493
	(0.153±0.015)	(0.289±0.021)	(2.002±2.517)	(0.989±0.289)	(0.603)	(0.714)
ANL	0.296±0.027	0.358±0.037	21.491±2.902	22.481±3.030	0.691	0.601
	(0.156±0.018)	(0.124±0.013)	(9.653±1.652)	(11.563±1.134)	(0.696)	(0.618)
MPR	0.184±0.017	0.225±0.017	11.662±2.837	14.854±1.999	0.628	0.697
	(0.087±0.006)	(0.062±0.009)	(7.976±0.58)	(9.856±0.659)	(0.751)	(0.442)

Linear regression analysis was carried out to estimate the non-combustion related PM_{10} in the three sites. For this reason PM_{10} and $PM_{2.5}$ daily mean concentrations were regressed with NOx daily mean concentrations (Table 4). The results varied substantially between sites. Specifically the PM_{10} non-combustion contribution in MPR was 30-39% depending of the season while in Birmingham an opposite behaviour was detected, where the non-combustion contribution of PM_{10} was between 61-75%. The higher non-combustion values were found during cold periods. In Athens, the combustion-related component accounted for 57% and 53% of PM_{10}, measured at ANL, during cold and warm seasons respectively, contributions highly comparable to those reported for other traffic-impacted measurement locations on the area (Grivas et al., 2004). In what concerns $PM_{2.5}$ the non combustion contribution was 3-8% in MPR and 39-47% in BBR, with the higher vales detected during the cold season of the year. On the contrary in Athens, the impact of fuel combustion on fine particles was more intense during the cold period, reflecting the use of fuel oil for space heating and higher total traffic volumes. The season specific contributions were estimated at 60% and 56%, values approaching past estimates for a canyon-type, kerbside monitoring location in downtown Athens (Chaloulakou et al., 2005).

Table 5. Pearson correlation coefficients between daily $PM_{10}/PM_{2.5}$ and meteorological parameters. $PM_{2.5}$ in italics.

Site	Wind speed		Temperature		Relative Humidity		Precipitation		Atmospheric Pressure		Radiation	
	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm	Cold	Warm
BBR	-0.44 *-0.52*	-0.1[a] *-0.19*[b]	-0.07[a] *0.08*[a]	0.23 *0.36*	0.13[a] *0.4*	-0.11[a] *0.12*[a]	0.18[b] *-0.1*[a]	-0.25 *-0.13*[a]	0.32 *0.24*	0.09[a] *0.02*[a]	0.19[b] *0.08*[a]	0.07[a] - *0.06*[a]
ANL	-0.43 *-0.44*	-0.20[b] *0.19*[b]	0.26[b] *-0.15*[a]	-0.02[a] *0.44*	-0.05[a] *0.08*[a]	-0.05[a] *-0.19*[b]	-0.13[a] *-0.12*[a]	-0.05[a] *0.05*[a]	0.08[a] *0.08*[a]	-0.11[a] *-0.05*[a]	0.16[a] *-0.03*[a]	-0.06[a] *0.10*[a]
MPR	-0.34 *-0.38*	0.03[a] *-0.27*	0.31 *0.18*	0.56 *0.45*	0.03[a] *0.03*[a]	-0.44 *-0.38*	-0.23 *-0.2*	-0.1 *-0.06*	0.33 *0.27*	0.03 *0.22*	0.03 *0.08*	-0.09 *0.09*

All correlations are statistically significant at 99% confidence interval (unless otherwise indicated)
[a] Correlation not significant
[b] Correlation significant at 95% c.l

The $PM_{10}/PM_{2.5}$ were correlated negatively with wind speed at all sites especially during the cold season. The weaker correlations during the warm season could be attributed to soil re-suspension and road dust that is higher during summer due to the dry environment. A significant negative correlation between precipitation and $PM_{10}/PM_{2.5}$ was observed especially in Madrid and Birmingham during cold periods. Relative humidity presented significant anti-correlation only in Madrid during the warm season, while significant positive correlations were found for temperature and atmospheric pressure in Madrid probably due to increased contribution of secondary particles on relatively warm and sunny days. This is also apparent for fine particles in Athens, producing a notable correlation with temperature, indicative of enhanced production of sulphate and organic carbon particles during the summer months. The correlation between solar radiation and particles was not significant (Table 5).

CONCLUSIONS

Sources and factors affecting $PM10/PM_{2.5}$ levels in 3 urban traffic locations in Athens, Birmingham and Madrid were identified. In Athens and Madrid traffic sources dominate both fine and coarse particles, either by direct emissions or by inducing dust resuspension, establishing a status of poor air quality regarding particle pollution. Moreover, secondary particle formation is a factor to consider, especially during favourable atmospheric conditions. In Birmingham non traffic sources are the dominant in the location examined.

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CHILDREN'S PM_{10} AND NO_2 EXPOSURES IN RELATIONSHIP TO AMBIENT CONCENTRATIONS DURING AN AIR POLLUTION EPISODE IN TURIN, ITALY

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Ambient air pollution has been set as one of the regional priority goals in European Union children's environment and health action plan. Ambient air quality surveillance against limit values and optimization of environmental safety for selected population groups may lead to vastly different emphases in policy formulation. In the current work PM_{10} and NO_2 exposures of 101,563 children below age of 15 living in the 120 km² metropolitan area in Turin, Italy are modelled and compared with levels observed at air quality monitoring sites.

The high-end short-term exposures are characterized by modelling episode-day exposures during the worst day of a 4-day stagnation in January 2003 in Turin. The exposure model is based on 1-km² spatial resolution and non-personalized time-activities of preschool and school children. Mean episode day exposures of NO_2 and PM_{10} of school children in the metropolitan area of Turin predicted by using air quality and exposure models only were substantially underestimated (~60%), but when accounting for the differences in predicted and observed ambient air quality, the fixed monitoring data represented the children's mean exposure quite well (observed ambient 111 µg m^{-3} versus 104 µg m^{-3} mean exposure).

Exposure modelling provides a powerful tool for policy optimization. Underestimations of ambient levels by air quality models in episodic conditions (59% for the current episode day) need and can be handled using observed data and scaling procedure when estimating true exposures. Air quality warning systems and consequent adjustments in the time-activities of the children as well as ventilation patterns used in buildings, including homes and schools, can reduce adverse exposures during episodes.

1. INTRODUCTION

Air pollution is associated with the development of serious cardio-vascular and respiratory diseases, including development of asthma in children (Gordian et al., 2006). Exposures of children are modified by (i) their mobility in the variable concentration fields, (ii) filtration of pollutants by the building envelopes, and (iii) indoor sources of air pollution. While air quality monitoring is subject to the limitations set by the small numbers of monitoring stations and the substantial variability of concentrations in urban environments (Sauli-Zajani et al., 2004), better spatial coverage is achievable with air quality models, which benefit also from the recent advances in the integration with urban micrometeorological modelling (Baklanov et al., 2006). The current work combines these with population exposure modelling.

Turin, one of the largest industrial centres in the Northern Italy, is located in the river Po valley. With a population of about 900,000 people the city is influenced by intense road and railway traffic. The city is frequently affected by very low winds and severe air pollution episodes. Air quality management in such a setting needs to consider options to lower exposures of sensitive population groups. One of the regional priority goals of the Children's Environment and Health Action Plan for Europe (CEHAPE, 2004) is to reduce acute and chronic respiratory disorders, including asthmatic attacks in children and adolescents due to outdoor pollution. The listed means to achieve this goal include pollution-free school zones and alert systems. The vulnerability of children to air pollution exposure is related to several differences between children and adults. (World Health Organization, 2005a)

2. METHODOLOGY

The exposure model used is based on microenvironment approach (Hänninen et al., 2003; Kruize et al., 2003). Exposures (*E*) are calculated as the time-weighted average concentration (*C*) over the microenvironments visited (indexed by *i*). According to equation 1, time-weighting is done using personal time-activities as fractions of time (f_i) spent in each microenvironment, implicitly defining the averaging time:

$$E = \sum_i f_i \times C_i \qquad (1)$$

Three microenvironments were defined: (i) indoors, (ii) in traffic, and (iii) outdoors. No separation was done between residence and school indoor environments. The small time fractions spent in other indoor environments, like shops or visits to friend's home, were not separately modelled. Times spent in the microenvironments were estimated by using typical daily timetables of school children in Italy.

Indoor concentrations were modelled using an infiltration model (Hänninen et al., 2005a,b, 2004a). Mean infiltration efficiency of $PM_{2.5}$ particles ranged from 0.59 in Helsinki to 0.70 in Athens; latter values from the Mediterranean climate were used in the current work as proxies for PM_{10} in lack of European PM_{10} specific data. The indoor microenvironment concentrations (C_i) were modelled according to equation 2:

$$C_i = F_{\mathrm{inf}} \times C_a + \sum_j C_{Sj} \qquad (2)$$

where F_{inf} is the infiltration factor and C_a the outdoor PM_{10} concentration used to estimate the indoor concentration of ambient origin ($F_{inf} \times C_a$). The concentrations (C_{Sj}) caused by various sources (j) were not considered in the current work. Concentrations experienced while in traffic were estimated using the fixed site monitoring station concentrations multiplied by coefficients observed in a number of studies reviewed by World Health Organization (2005b) (1.5 for both NO_2 and PM_{10}).

The exposure model included the whole metropolitan area (120 km²) with 101,563 children aged 14 or less. Children of age 0-14 years were assumed to be evenly distributed between the ages when combining time-activities of school children and children below school age (0-6 yr).

Spatial distribution of ambient air quality during an extreme episode condition on 13.-14. January, 2003 was estimated using the urban air quality information and forecasting system applied in Turin, developed in the framework of the FUMAPEX project at ARPA Piemonte in collaboration with ARIANET consulting. Only Tuesday, the worst day, is included in the results presented here. The system was built to provide hourly forecasts on pollutants defined in the current Italian legislation (PM_{10}, NO_2, O_3, CO, SO_2 and benzene). The system's main constituents are the prognostic non-hydrostatic meteorological model RAMS (Cotton et al., 2003) which downscales ECMWF weather forecasts, and the three-dimensional Eulerian model FARM (Calori et al., 2005) that accounts for the transport, chemical conversion and deposition of atmospheric pollutants. Emissions were estimated using results from the European EMEP inventory, the national Italian inventory CORINAIR, and the high resolution inventory for Piemonte region, and integrated to set up the database needed to perform the chemical and dispersion simulations. Resuspension was estimated as a source category in the emission inventory. Long range transport and secondary particulate matter are taken into account with initial and boundary conditions.

Concentrations were calculated for 1 km² grid and exposures of children living in each grid cell were estimated using the grid concentration data, time-activity, infiltration, and traffic concentration factors. All time spent outside home was assumed to occur during daytime (07-22). Effectively, the average exposure value for each grid cell (E) is calculated according to equation 3, where f is fraction of time during the daytime hours (7-22) spent in indoors (f_i), outdoors (f_o), and in traffic (f_t) and C is the concentration for daytime (C_d) and night time (C_n):

$$\overline{E} = \frac{14}{24}(f_i F_{INF} + f_o + f_t k_t)C_d + \frac{10}{24} F_{INF} C_n \qquad \mathbf{(3)}$$

When using an air quality model as source of concentrations, the model errors need to be estimated to get "best estimates of the true exposures". Policy development must be based on best understanding of the true exposures, and if the models have known deficiencies, these must be accounted for. In Turin the difference between modelled and monitored air quality at Consolata station was 46 vs 111 µg m^{-3}, ratio 0.414.

3. RESULTS AND DISCUSSION

The average PM_{10} exposure level (104 µg m^{-3}) is 93 % of the level observed at the Consolata monitoring site located in the centre of the city but approximately 250 m from the major road. Consolata reflects quite well the average exposures, but nevertheless more than 40% of the children are exposed to higher levels. In relationship to the corresponding WHO PM_{10} guidelines (2006) the exposures were 208 % of the daily guidelines, while the ambient levels at the monitoring sites exceeded the EU PM_{10} limit values by 223 %. Approximately 90 percent of the children living in the metropolitan area were exposed to levels above the WHO 24-hour PM_{10} guideline and corresponding EU PM_{10} limit value defined for the ambient air. Moreover, all tailpipe particles from traffic as well as combustion particles from local sources are in fact smaller than 2.5 µm in diameter and therefore application of lower guideline for $PM_{2.5}$ could be relevant.

Comparison of geographical distributions of exposures modelled in Turin and emission inventory data indicate that the higher modelled PM_{10} concentration levels occur in industrial and traffic areas, as confirmed by the highest levels observed in the Northern border of the area. Nevertheless, the highest exposure levels occurred in the South-Western downtown area, as a result of both the high population density and the presence of some industrial sources in the vicinity.

Potential exposure reduction policies could involve traffic bans, industrial stoppages and recommendations for reduced ventilation. Actions specific to children include closing schools and recommendations to avoid playing outdoors during episodes. A long-term policy option especially effective on children is to apply mechanical ventilation with particle filtration in schools.

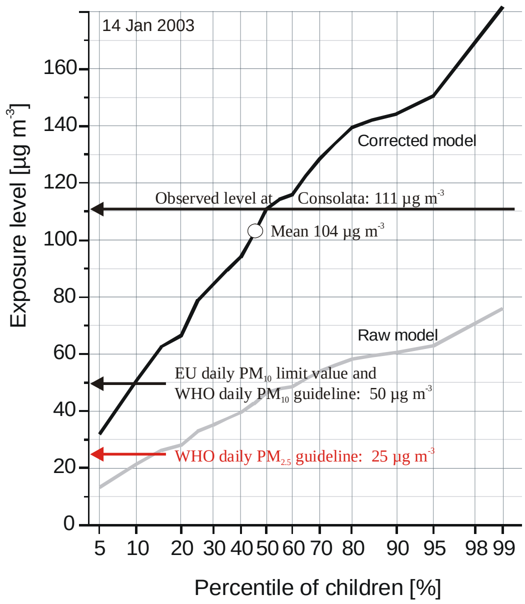

Figure 1. Exposure distribution of children in comparison to observed level at monitor (111 µg m^{-3}) and some guideline values. Raw model does not account for underestimation of episode levels.

Exceedances of EU limit values are substantially more common for PM_{10} than for NO_2. Moreover, also higher PM_{10} exposure levels indicate that PM_{10} is the main concern for public health. Current no-threshold model for PM dose-response suggests also that there are public health effects even below the limit value. The high spatial variability indicates that the episode is largely of local origin, supporting the used relative model scaling procedure.

4. CONCLUSIONS

Peak exposures of children were modelled for an episode day to study implications for health based environmental policy refinement. As shown by numerous studies earlier, relationship of personal exposures and outdoor levels at monitoring stations are very variable and different for different types of monitoring sites. In Turin the monitoring station Consolata, located in the downtown but not close to the major traffic arteries, represented the average exposures of the children quite well.

Urban air quality information systems have been originally developed to assess compliance to air quality standards, but can be utilized also to promote public health in combination with exposure models. Networking of end-users (metropolitan area and regional authorities), scientists (atmospheric chemists, physicists, meteorologists, modellers, environmental scientists, epidemiologists, etc.), and policy makers is needed for development and application of exposure modelling techniques to support health-based air quality management. The air quality model was able to capture the spatial variability of levels only partly; as was highlighted in the case of the peak episode. Therefore it is important to evaluate models against observations and to consider a model calibration procedure, when the necessary data is available.

Majority of the children were exposed to levels of health concerns in the case of an episode. Air quality management in episode situation requires reliable forecasting to allow for the actions like traffic restrictions, industrial shutdowns, and recommendations to the public via media to be prepared and implemented. However, based on the epidemiological data, health effects of long-term exposures outnumber those of high but short-term peak exposures. Therefore, additional focus in environmental policy development must be in reduction of long-term exposures. In practice this means optimization of air quality management decisions according to the quantitative estimates of exposures in alternative policy scenarios. Compliance to air quality standards is a weak optimising target for health based air quality management and it does not guarantee optimal environmental safety. This is emphasized by the fact that almost all recent epidemiology points to linear zero threshold exposure-response relationship. A long-term goal must be set to develop air quality legislation instruments towards better support to health-based decision-making.

5. ACKNOWLEDGEMENTS

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COMPARING THE IMPACT OF A ROAD TUNNEL VERSUS A ROAD VIADUCT BY MEANS OF AN INTEGRATED EXPOSURE ASSESSMENT

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ABSTRACT

Emissions and concentrations of PM_{10} and NO_2 were calculated for a road tunnel and a road viaduct, foreseen as a means to close the ring road around Antwerp, a major city in the north of Belgium. The assessment was carried out using the integrated MOBILEE approach. The MOBILEE methodology has been applied for a reference situation (2003), the ring road closure by means of a tunnel and the ring road closure by means of a viaduct. For the tunnel scenario, two different exhaust heights were considered (5 m. and 30m.). The exhaust height was found to be a crucial parameter. Compared to the impact of the viaduct, a tunnel with an exhaust at a height of 5 m. shows an *increase* in total exposure of 40%, whereas an exhaust height of 30 m. shows a *decrease* in total exposure of 5%.

1. INTRODUCTION

We calculated the emissions and concentrations of PM_{10} and NO_2 for a road tunnel and a road viaduct, foreseen as a means to close the ring road around Antwerp, a major city in the north of Belgium. One of the criteria on which the choice between these two road constructions will be based is a detailed air quality assessment. The results were evaluated against the limit values presented in the EU directives on ambient air quality assessment and management (1999/30/EC). The calculated PM_{10} and NO_2 concentrations were further used to assess the exposure of the population in Antwerp living in the vicinity of the planned constructions. Section 2 describes the MOBILEE methodology which was used for this study. Further exposure assessments were based on the impact pathway methodology, which is discussed briefly in this section as well. Section 3 summarizes the main results of the study and discusses briefly its main implications with respect to the construction works.

2. METHODOLOGY

The assessment was carried out using the integrated MOBILEE approach. This approach combines road transport emission calculations (Mensink et al., 2000) with dispersion and exposure modelling. The dispersion modelling is based on the coupling of two models: the street canyon model OSPM (Berkowicz, 1998) and the Gaussian model IFDM (Cosemans et al., 1997). OSPM is a street canyon model and calculates the contribution of the traffic emissions inside a particular street. IFDM is a Gaussian dispersion model and computes the background contributions. This includes not only the contribution from industrial stacks and domestic heating within a domain with a 20-30 km radius (larger Antwerp region), but also the concentration levels caused by the surrounding streets.

The exposure assessment is based on the impact pathway methodology (see Figure 1), as discussed extensively in the book by Friedrich and Bickel (2001). The impact pathway method follows the fate of pollutants along the steps in the DPSIR chain: Drive (human activities), Pressure (emissions), State (air quality and exposure), Impact (health, economic) and Response (policy). The evaluation of environmental impacts is based on the accounting framework of the European ExternE project. Using the ExternE methodology, estimations of the environmental damage costs related to the impacts can be provided (Friedrich and Bickel, 2001). The calculation framework for external costs results from a series of European research projects, starting in 1991 and still ongoing for further development, extension and refinement.

The MOBILEE methodology has been applied for a reference situation (2003), the ring road closure by means of a tunnel and the ring road closure by means of a viaduct. Three corresponding mobility scenarios were used to calculate the traffic emission. The mobility scenarios are based on the current situation (1), the closure of the ring road by means of a viaduct (2) and the closure of the ring road by means of a tunnel (3). In all scenarios the number of vehicles and the composition of the traffic fleet was identical and based on the situation in 2003. For the tunnel scenario, two exhaust heights have been studied. In a first case, the emission height was set at 5 m. In the second case an emission height of 30 m. was used. Note that all scenarios include a part in which a tunnel is running underneath the Scheldt river. The exhaust height for this part of the ring road was set at 5 m.

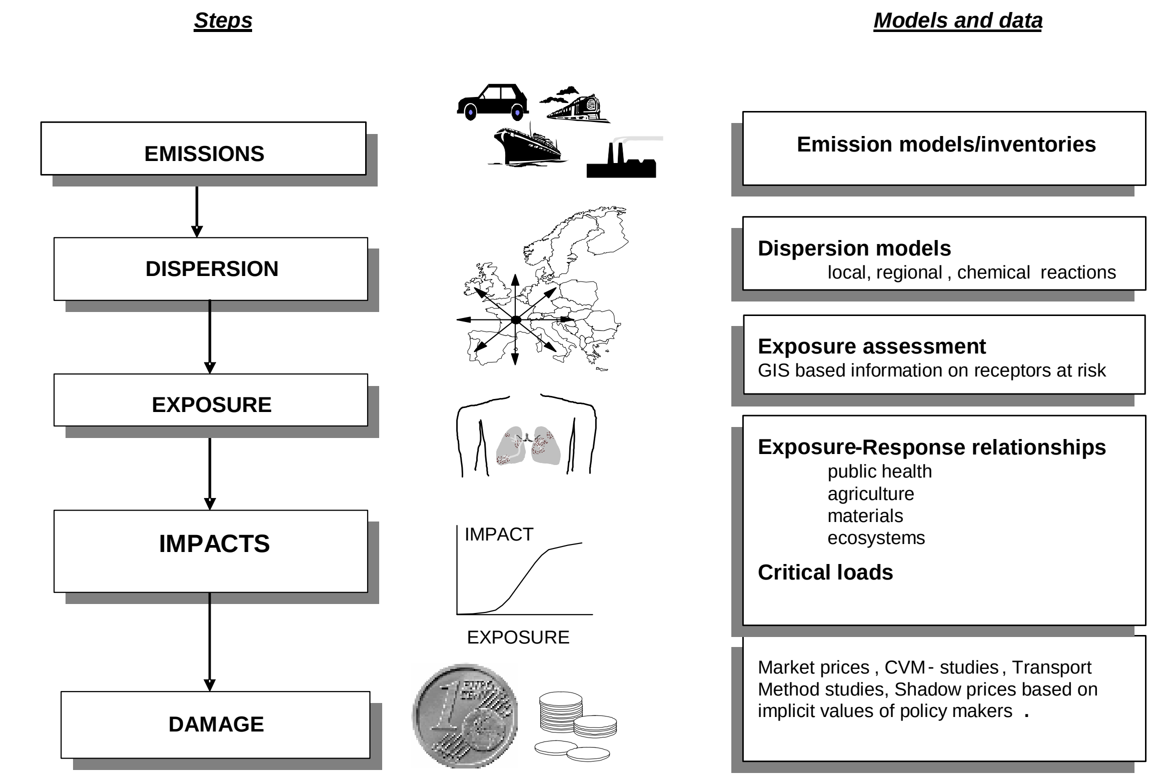

Figure 1: Integrated assessment framework: the impact pathway methodology.

3. RESULTS AND DISCUSSION

3.1 Emissions

Table 1 gives an overview of the annual emissions (2003) for NO_x and PM_{10} in the viaduct scenario and the tunnel scenario. Note that the differences are very small and associated with a slight modification in the trajectory. For comparison, Table 1 also shows the total annual traffic emissions in the neighborhood and the total annual traffic emissions in Flanders in 2003. Another remark is that the exhaust pipe of the tunnel does not include any filter installation

Table 1. Annual NO_x and PM_{10} emissions for the viaduct scenario and the tunnel scenario in comparison with emissions in the neighborhood and with total traffic emissions in Flanders.

Scenario	**NOx (tons/year)**	**PM_{10} (tons/year)**
Viaduct scenario	201,2	8,15
Tunnel scenario	197,7	8,0
Ring road neighbourhood	709	31
Traffic in Flanders	87488	4384

3.2 Annual PM_{10} and NO_2 concentrations

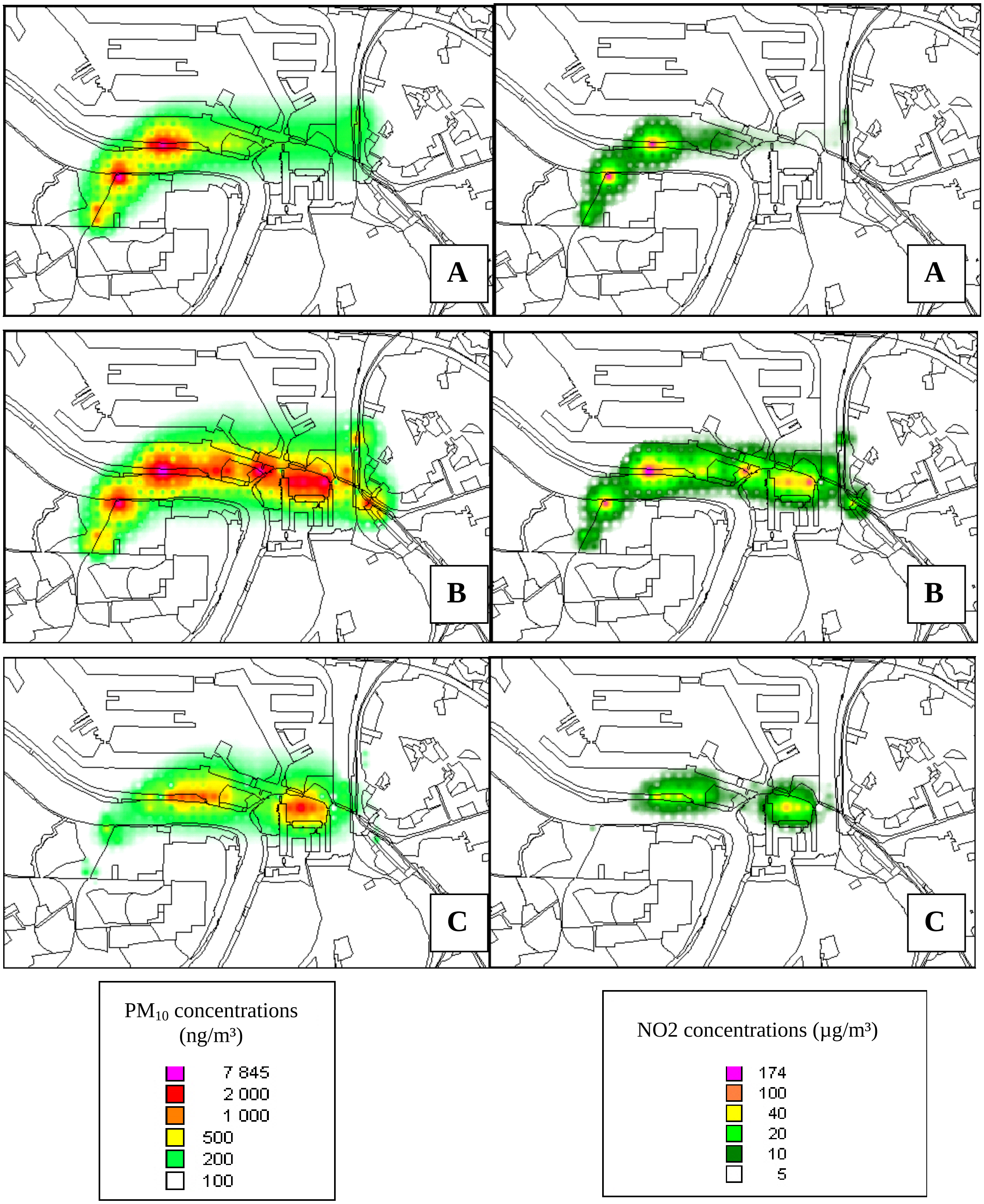

Figure 2. Calculated increase in annual PM_{10} concentrations (left panels) and annual NO_2 concentrations (right panels) for A) the viaduct scenario; B) the tunnel scenario with exhaust height of 5 m. and C) the tunnel scenario with exhaust height of 30 m.

From figure 2 we can learn that the emission exhaust height in the tunnel scenario is a decisive parameter. This becomes clear when comparing plots B and C. In both figures the coloured areas correspond with the parts of the tunnel scenario where an “open” tunnel exists, i.e. a part of the tunnel that is not covered. The highest concentrations are obtained for the tunnel variant with an exhaust height of 5 m. The expected total increase in PM_{10} concentrations in case of the tunnel variant with an exhaust height of 30 m. is approximately 1 µg/m³. Locally the NO_2 concentrations are expected to rise in this scenario with 20 µg/m³.

3.3 Exposure calculations

The exposure results were obtained by using a GIS map with detailed information of the population density in the Antwerp area. Thus exposure is evaluated in a static way. Note that most people who will be affected by the new road are living east of the tunnel or viaduct. In terms of exposure assessment, PM_{10} is the dominant parameter. However, an evaluation of the population exposure for the three situations demonstrates that the differences are not very significant in absolute terms, because of the high background concentrations for PM_{10}. Compared to the impact of the viaduct, a tunnel with an exhaust at a height of 5 m. shows an *increase* in total exposure of 40%, whereas a tunnel with an exhaust height of 30 m. shows a *decrease* in total exposure of 5%. A further consideration with respect to the tunnel situation is the possibility to apply filter installations in order to further reduce the concentrations.

Note that the exposure calculations have been based on data for 2003, whereas the inauguration of this new part of the ring road is foreseen in 2015. By this time the fleet composition, the traffic emission factors and possibly the population distribution might have been changed considerably. This effect has not been quantified nor been taken into account by the study.

4. CONCLUSIONS

The MOBILEE methodology has been applied to study different scenarios for the ring road closure in Antwerp, a major city in the north of Belgium. Three mobility scenarios were used to calculate the traffic emission: a ring road closure by means of a viaduct and a ring road closure by means of a tunnel, varying the exhaust height between 5 m. and 30 m..

The evaluation of the population exposure for the three situations demonstrates that the differences are not very significant in absolute terms, because of the high back ground concentrations for PM_{10}, being the dominant parameter in the exposure assessment. A tunnel with an exhaust height of 5 m. shows an *increase* in total exposure of 40% when compared to the viaduct scenario. A tunnel with an exhaust height of 30 m. shows a *decrease* in total exposure of 5% when compared to the viaduct scenario.

It should be taken into consideration, that the tunnel variant offers the possibility to install filter equipment to further reduce the PM_{10} concentrations. However, the open parts of the tunnel, i.e. the parts where the tunnel is not covered, are mostly contributing to the decrease in air quality or the increase in exposure. This will not be altered by introducing filter equipment.

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CHARACTERISTICS AND INFLAMMATORY POTENTIAL OF WEAR PARTICLES FORMED IN A ROAD SIMULATOR

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ABSTRACT

Inhalable particles (PM_{10}) in the ambient air is recognized as one of the most severe air pollutants. In Sweden, wear particles from pavements, tyres and gritting periodically cause very high PM_{10} concentrations causing the environmental quality standard for PM_{10} to be surpassed in many Swedish municipalities. Since little is known about the mechanisms causing particles to induce health effects, knowledge about source specific particle characteristics and health effects is needed in order to be able to develop cost-effective measures to mitigate particle emissions. At VTI (Swedish National Road and Transport Research Institute) a road simulator was used as a wear particle generator and wear particles with a low contamination from other particles sources can be studied and sampled for further in vitro studies. This paper presents some results from the WearTox project, where wear particles from two different pavements where characterised and their inflammatory potential in human cells were investigated.

1. INTRODUCTION

Wear particles from road pavements and tyres strongly contribute to episodes with very high concentrations of inhalable particles in outdoor air in Sweden. These episodes normally occur during dry periods in winter and spring. During the winter season in Sweden, studded tyres are commonly used except in the southernmost parts where friction tyres tend to be more common. Even though pavements have been improved since the 80s and studs nowadays are mainly made of lightweight alloys instead of steel, about 100 000 tons of pavement is worn each winter season in Sweden. Also, winter sanding in urban areas contributes to dust formation, both through vehicles grinding sand into dust, but also through increased pavement wear by sand (Kupiainen et al., 2003). During dry periods in winter and early spring, abraded pavement and sand are ejected into the air by vehicle turbulence and cause particle concentrations to vastly exceed the environmental quality standard set for inhalable particles.

Numerous studies have shown that the concentration of inhalable particles in ambient air is associated with mortality and different kinds of respiratory health problems in the population (Schwartz et al. 1996; Schlesinger 2000, Brunekreef and Forsberg, 2005). However, the mechanisms and properties that make particles more or less toxic are poorly understood. To find optimal measures against high PM_{10} concentrations, it is important to study source specific particle characteristics as well as to determine what properties of the parent material are important for the release of inhalable wear particles.

This paper presents some results from the WearTox project in which wear particle properties from two pavements were described and their inflammatory potential in human airway cells investigated.

2. METHODOLOGY

At the Swedish National Road and Transport Research Institute (VTI) a circular road simulator was used to generate wear particles (Figure 1). Particle sampling in the simulator hall makes it possible to sample pure wear particles, with very low contamination from ambient particles and tail-pipe emissions. In the experiments, PM_{10} particles from studded tyre wear of two Swedish pavements (one with granite and one with quartzite) were characterised and sampled for cell tests. The particle generation is described in the paper by Gustafsson et al. (2007) in the current proceedings. The airborne particles in the simulator hall were sampled using a PM_{10}-inlet and the particle size distributions were measured using APS (Aerodynamic Particle Sizer). Particles less than 1 µm was sampled using a 2 m and 6 mm in diameter copper tube and measured using a SMPS (Scanning Mobility Particle Sizer) placed outside the simulator hall. PM_{10} sampling was made using a high volume sampler (Sierra-Andersen/GMW Model 12000). Particles were studied using a Scanning Electron Microscope (SEM) (LEO Gemini 1550) with Emission Dispersive X-ray Spectroscopy (EDS) (Link ISIS).

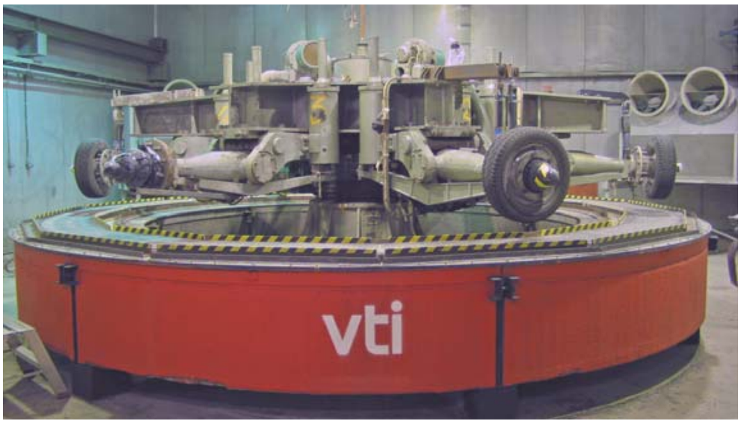


Figure 1. The VTI circular road simulator.

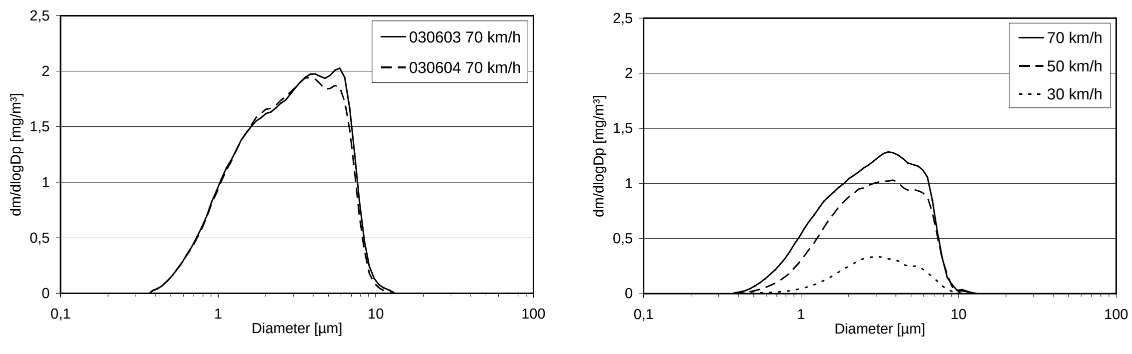
For comparison in the cell study particles from a street environment and subway particles were collected by The City of Stockholm Environment and Health Administration, during early spring at a highly trafficked street (Hornsgatan) and in a subway station (Mariatorget) in Stockholm, Sweden. Inhalable particles were sampled with a high volume sampler (Sierra-Andersen/GMW Model 12000) equipped with glass fibre filters (Munktell MG 160). Diesel particles (DEP) were generated (AVL MTC AB, Stockholm, Sweden) according to a standardized test protocol (70/220/EEC) and during the particle sampling the engine was run according to the European driving cycle (NEDC). A side flow was lead through a Teflon filter on which the particles were collected.

Cell study methodology is described in detail in Lindbom et al. (2006). Human monocytes were isolated from heparinized human whole blood and allowed to differentiate in to macrophages. The particles were resuspended in the cell growth medium and sonicated. The cells were then incubated with a final concentration of 10, 50, 100, 250 and 500 µg/ml for 18 h in a total volume of 1 ml/well. Cytokines (IL-6, IL-8, IL-10 and TNF-α) were measured with QuantiGlo kit (R&D-systems, Minneapolis, MN) using a Lumistar (BMG Labtechnology, Offenburg, Germany) luminometer, according to the manufacturers instruction.

3. RESULTS AND DISCUSSION

3.1. Particle characteristics

Mass size distributions of PM10 pavement wear particles peak at 2–8 µm for the two pavements studied (granite and quartzite, Figur 2). SEM photography of PM_{10} particles show that particles from pavement wear by studded tyres consist of freshly worn rock material with sharp edges. The granite PM10 have a somewhat more flaky appearance, while the quartzite PM10 is more granular (Figure 3). Particles originating from tyres should be present in the system, but is suggested to be too small to be detectable at this magnification and also too large to be collected in PM10. In Dahl et al. (2006) and in Blomqvist et al. (2007, in these proceedings) ultra-fine particles produced during the tests are discussed.



Figur 2. Mass size distribution of pavement wear particles caused by studded tyres. At left from the granite pavement and at right from the quartzite pavement.

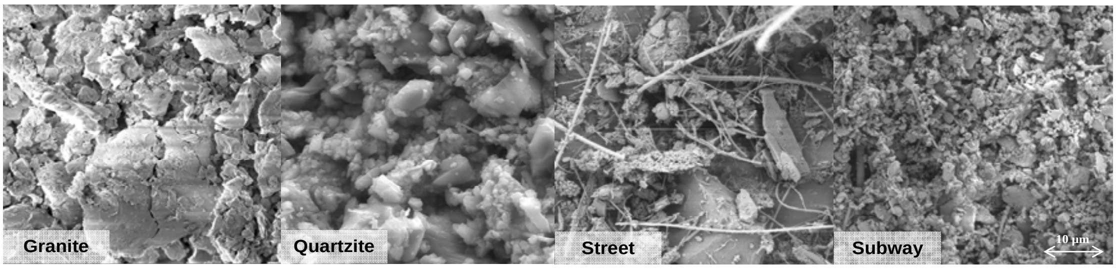
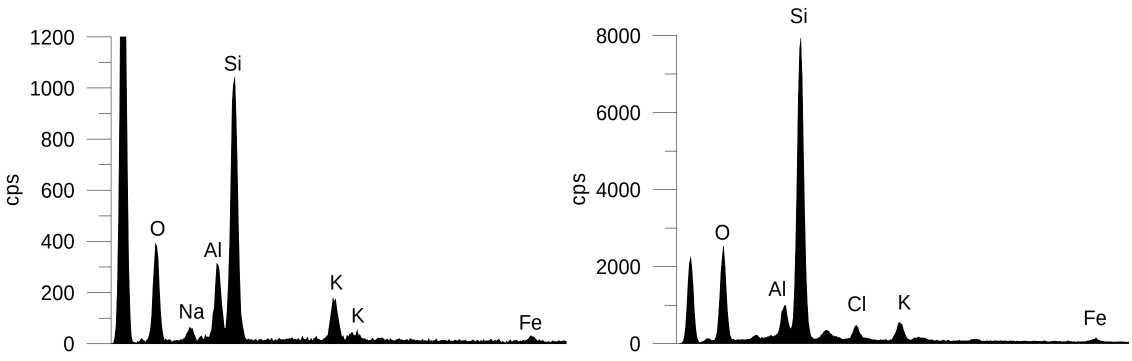


Figure 3. SEM photos of PM_{10} from combinations 1 (Granite, upper left), 2 (Quartzite, upper right), urban street (lower left) and subway (lower right). The magnification was 2000 times.

Also element spectra confirm that the wear particles mainly consist of minerals (Figur 4). Silica, oxygen, aluminium and potassium dominate the spectra, but while the granite PM_{10} contains some sodium the quartzite PM_{10} contains chloride.



Figur 4. EDS spectra of wear particles from the granite pavement (left) and the quartzite pavement (right).

3.2. Cell study

The cell study shows that all tested particles induce inflammatory responses in human macrophages. Wear particles from the granite pavement and street particles induces higher secretion of the cytokine TNF-α than the equivalent dose of wear particles from the quartzite pavement and subway particles and the difference is significant ($p<0.05$) (Figure 5). The same pattern was seen also with the cytokines IL-6 and IL-8. Generally the wear particles induced inflammatory response at least as much as the diesel particles. A more detailed analysis can be found in Lindbom et al. (2006).

The differences in inflammatory potential between the granite and quartzite pavement particles are difficult to relate to the particle characteristics. The particle size distributions are very similar, which suggests that other particle properties are more important than size for the observed effects.

The results imply that pavement wear particles, even though they primarily consist of stone, should be regarded as a potential health risk. Also, the obvious difference in inflammatory potential between a granite and a quartzite pavement shows that there is a potential to mitigate health effects from pavement wear particles through choosing rock materials with optimized health properties.

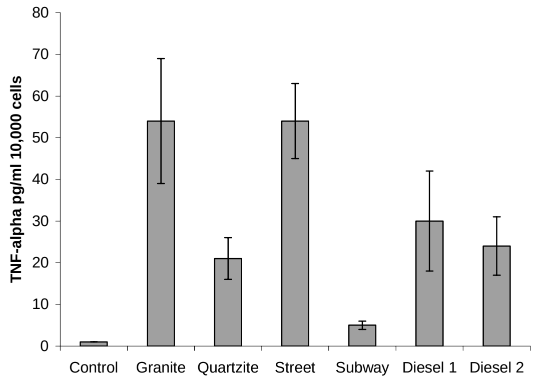


Figure 5. Secretion of TNF-α from macrophages into the cell growth medium exposed to 100 µg/ml of respective particle type for 18 hours. Diesel 1; particles water extracted, Diesel 2; particles methanol extracted. Cells only exposed to cell growth medium used as controls.

4. CONCLUSIONS

- PM_{10} particles produced when using studded tyres on a granite and a quartzite pavement mainly consist of freshly worn minerals.
- Granite pavement PM_{10} is more flaky and contains relatively more aluminium, sodium and potassium than quartzite PM_{10}, while quartzite PM_{10} is more granular and also contains some chloride
- Mass size distributions are very similar for granite and quartzite PM_{10}.
- Wear PM_{10} from a granite pavement is more inflammatory than PM_{10} from a quartzite pavement, as inflammatory as street PM_{10}, at least as inflammatory as diesel PM_{10} and more inflammatory than subway PM_{10}.

5. ACKNOWLEDGEMENTS

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INDOOR/OUTDOOR PARTICULATE MATTER CHEMICAL CHARACTERISTICS AND SOURCE-TO-INHALED DOSE RELATIONSHIPS -- URBAN-AEROSOL PROJECT: OVERVIEW OF RESULTS FOR OSLO, NORWAY

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ABSTRACT

Indoor-outdoor measurements have been performed in the Oslo metropolitan area at two different houses during summer and winter periods. The objective was to study and evaluate the mechanisms controlling the outdoor/indoor relationships and the composition/size distribution of indoor generated PM in the selected urban places and their consequences on exposure and internally PM deposited regional dose in the human respiratory tract. The results indicated that the outdoor concentrations of particles were enhanced during the winter and lower during the summer. The main sources of particles outdoors were vehicle exhaust, domestic heating and natural marine and crustal aerosols emissions. In the indoor environment particles were mostly affected by the penetration of outdoor air indoors –consequently the air exchange rate - and indoor activities. The respiratory tract dose of a typical individual exposed to the above concentrations was higher outdoors and enhanced indoor on days with indoor activities.

1. INTRODUCTION

People spend about 85% of their time indoors and therefore are exposed to PM and gaseous pollutants from both outdoor sources through infiltration of outdoor air and indoor sources (personal activities, cleaning, emissions from building materials and products, combustion – e.g. cooking, cigarette smoking, candle burning etc- and various other indoor activities) (Eurostat, 2004; Molhave et al., 1991; Su, 1996; Nazaroff et al., 2004). Exposure to indoor particulate indoor air pollutants has been associated with sensory irritation, respiratory symptoms, allergy, neurotoxic and other adverse health effects and increased mobility and mortality (Pope et al., 1995; Schlesinger, 1995; Molhave et al., 1991).

There are numerous recent papers on indoor air quality measurements and modelling studying the different aspects of the indoor air quality ranging from the characterisation of the indoor/outdoor ratio (I/O) of specific pollutants, indoor source strength, infiltration, deposition of pollutants on surfaces, chemical reactions and human exposure (e.g. Wallace, 2000; Weschler, 2004; Nazaroff et al., 2004, Lazaridis et al., 2006). In the current work we present the results from an indoor/outdoor study performed at two different accommodation places during summer and winter periods (2002-2003) in Oslo, Norway. The work is part of the European Commission project Urban-Aerosol (Characterisation of Urban Air Quality - Indoor/Outdoor Particulate Matter Chemical Characteristics and Source-to-Inhaled Dose Relationships) which comprises indoor/outdoor measurements in Athens, Oslo, London, Hannover, Prague and Milan. The objective of the measurement study was to determine the physico-chemical properties of indoor and outdoor particulate matter associated with actual human exposure. In this work we present an overview of the results regarding the indoor concentration of particles and gaseous pollutants and their relationship to outdoor concentration, indoor sources/activities, air exchange rate, meteorological conditions and chemical processes indoors, the evaluation of possible sources of particles both indoors and outdoors and the consequences of the above factors on the exposure and internally PM deposited regional dose in the human respiratory tract.

2. METHODOLOGY

Indoor-outdoor measurements have been performed in the Oslo metropolitan area at two different accommodation places during summer and winter periods. The selected urban places were one apartment in the Oslo centre and a house at the suburbs. Measurements included one week of monitoring during each season at each place (04/03/02 – 18/03/02 and 03/06/02 – 17/06/02 for the house and between 26/08/02 – 09/09/02 and 13/01/03 – 27/01/03 in the apartment). Particulate matter measurements were performed using both offline and real time instruments. Specifically, TEOM instruments were used for continuous measurements for PM_{10} together with a Scanning Mobility Particle Sizer (SMPS) model 3934C, and an Aerodynamic Particle Sizer (APS) model 3320 for particle number distribution measurements. Integrated PM_{10} and $PM_{2.5}$ measurements were also conducted using the URG Versatile Air Pollutant Sampler (VASP), dichotomous samplers (Dichotomous Partisol Plus (DPP) sequential air sampler, model 2025), Kleinfiltergerät instruments (Low Volume Sampler LVS3.1) and impactors (8 stage Berner type low pressure cascade impactor) for the aerosol size distribution outdoors. In addition a denuder/filter pack system has been employed in order to study the volatile inorganic aerosol components in conjunction to the related trace gases, indoors and outdoors. Samples were collected either on Teflon filters (47 mm diam. Pall Zefluor 2 μm) or on pre-fired quartz filters (Whatman QM-A) analyzed for inorganic water soluble ions (IC) and EC/OC (thermo-optical analysis) respectively. Further, gaseous precursors for secondary aerosol formation were measured. In particular O_3 and NO_2 were measured using automatic chemiluminescence instruments (Ozone Analyzer, model 400A; ML®9841A Nitrogen Oxides Analyzer) and air samples have been analysed (GC/MS) for Total VOC and specific organic compounds content. The air exchange rate was evaluated using sulphur-hexafluoride gas (SF_6) and meteorological parameters were measured. The measurements were performed in the presence and absence of human activities (smoking, cooking and dusting) which were registered in an activity diary.

In addition the human exposure and the received dose in the human respiratory tract were evaluated. The actual human dose is estimated as the product of aerosol concentration in air, the ventilation rate, and the deposition fraction of particles in the respiratory tract separately for the fine and coarse mode. The deposition fractions of particles in the respiratory tract were derived by the ICRP66 model (ICRP, 1994). The required physiological parameters are set according to the reference values for the "adult Caucasian male" characteristics given in the ICRP66 model (ICRP, 1994). The subject is supposed to be always under "light exercise" (outdoor exposure scenario) and consequently the ventilation rate equals to $1.5m^3/h$. Furthermore, particles are considered to be spherical (shape factor is 1) and have a typical density of $1g/cm^3$.

3. RESULTS AND DISCUSSION

Descriptive statistics of the measured parameters at each season (real time instruments) are presented in Table 1. The outdoor concentration of particles and nitrogen oxides is higher during the winter measurement periods whereas O_3 concentrations were enhanced during the summer. During the winter period concentrations are affected by emissions from domestic heating and heavy traffic especially at the Oslo centre site (Colletts gate apartment) while during the summer O_3 production is promoted. Indoor concentrations have been typically lower than the outdoors for the gaseous compounds and exhibited large positive correlations indicating that the infiltration of outdoor air is their main source indoors. On the other hand the indoor concentration of particles varied in respect to season and indoor activities. Specifically the indoor concentration of particles was lower than the outdoor one except for the case of specific indoor activities usually between 9:30 - 12:30. In the absence of indoor sources, particle concentrations indoors were correlated with outdoor concentrations (e.g. $R^2 = 0.65$ for the June 2002 period) whereas during days with indoor activities the PM concentration variability indoors increased and the I/O ratio reached values up to 50 (cigar smoking). In addition the concentration of total particles was higher indoors than outdoors during the winter due to the indoor activities, the operation of a heating system and the deficient ventilation for energy saving. On the other hand the indoor concentration was lower than the outdoor in the summer at both sites due to the increased ventilation and the enhanced secondary aerosol formation in the outdoor environment.

The effect of meteorological parameters on the I/O concentration ratio was generally less pronounced and overshadowed by the variability in all the other parameters affecting the particulate matter and gaseous compounds concentration in both environments. Only ozone concentrations were correlated with temperature during the June 2002 measurement period (Pearson R = 0.7). The air mass origin however affected both the indoor and outdoor concentrations. Specifically, when air masses originated from western locations the concentration of marine particles was enhanced, whereas, when the air masses originated from eastern and northern areas the concentration of the secondary and crustal component of aerosols was enhanced (according to chemical composition data and back-trajectory analysis). In addition their indoor concentration was increased when the air masses originated from eastern directions.

Outdoor PM concentrations were influenced by marine and crustal aerosols and anthropogenic pollution from both local (vehicle emissions, domestic heating) and remote sources. The concentration of nitrate, chloride, sodium, calcium and magnesium ions was higher during the winter periods due to the enhanced effect of local traffic (direct emissions and resuspension of road side dust) whereas sulphate and ammonium concentrations were higher during the summer periods due to the enhanced secondary aerosol formation (as indicated by the OC/EC concentration ratio and the correlation of outdoor PM_{10} with NOx and O_3). The average outdoor concentration of most inorganic species was higher than indoors whereas the organic mass was higher indoors, especially during days with activities.

The concentration of TVOCs, and pollutant groups in TVOCs, specifically aromatic compounds, terpenes aldehydes and acids are presented in table 2. During the measurements butanol concentration was enhanced due to its use in DMPS monitors at the sampling sites. The concentration of TVOCs outdoors was low (19/1)

Table 1. Concentrations of PM_{10}, NOx, O_3, NO_2, NO and values of meteorological parameter during the 4 sampling periods (STD: standard deviation).

	PM_{10}[1]		NOx[2]		O_3[3]		NO_2		NO		Temperature (^oC)		Air Velocity ($m\ s^{-1}$)	Relative Humidity (%)	
	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor		Outdoor	Indoor
Steinborgveien house															
June 2002															
Mean	9.08	8.83 (5.73)[4]	20.31	26.97	56.60	11.75	16.79	20.09	2.39	4.58	17.11	33.25	1.39	57.91	25.91
Median	8.17	6.07 (5.68)	15.20	20.05	56.60	9.80	13.35	17.40	1.30	6.23	16.45	32.30	1.30	54.85	25.40
STD	3.97	21.04 (2.23)	16.93	16.26	24.93	8.79	11.60	8.14	4.40	41.60	4.57	4.52	0.74	20.22	4.46
5'th percentile	4.21	1.56 (1.74)	6.50	10.60	16.12	5.40	5.30	9.34	0.30	0.40	10.57	26.40	0.50	30.07	20.07
95'th percentile	16.33	16.87 (9.48)	50.39	58.73	96.40	25.24	39.39	33.16	8.30	18.20	24.80	40.04	2.80	91.84	33.60
March 2002															
Mean			27.11	22.65	52.26	21.95	21.75	19.56	4.07	3.32	1.37	21.45	1.97	63.52	19.77
Median			17.17	17.20	56.57	23.17	16.15	16.22	0.55	0.80	1.60	21.70	1.60	61.80	19.50
STD			30.37	21.74	21.37	8.35	16.99	13.50	10.86	8.04	3.16	2.08	1.28	19.22	3.33
5'th percentile			4.41	5.20	8.81	5.43	4.59	5.08	0.10	0.10	-4.40	17.20	0.39	36.13	14.90
95'th percentile			82.80	68.76	79.63	33.10	60.99	48.59	23.45	20.78	6.51	24.60	4.50	93.31	25.91
Colletts gate apartment															
January 2003															
Mean	12.34	13.47 (9.30)	74.10	64.20	21.30	7.56	39.21	33.32	22.83	20.21	1.24	21.40	0.69	84.59	24.16
Median	10.42	8.91 (7.93)	43.80	42.90	17.00	6.20	34.90	31.90	4.15	3.85	2.30	21.50	0.50	89.40	24.90
STD	8.32	19.84 (6.81)	88.08	70.27	17.80	5.90	27.56	17.32	43.52	38.87	3.27	1.01	0.58	13.29	3.96
5'th percentile	4.13	2.88 (2.70)	5.30	8.10	1.40	3.36	4.64	7.46	0.40	0.40	-5.91	20.00	0.10	52.69	16.69
95'th percentile	28.44	29.26 (18.14)	248.59	192.73	52.33	14.12	93.78	59.15	103.76	94.23	4.91	23.00	1.90	96.60	29.80
August/September 2002															
Mean	5.19	12.40 (10.85)	26.71	19.04	40.33	6.74	20.19	15.26	4.54	2.61	18.11	27.29	0.70	69.16	41.56
Median	4.94	7.63 (7.33)	18.60	16.00	39.80	6.40	16.10	14.00	1.40	1.30	17.50	27.20	0.70	71.90	41.80
STD	1.56	17.56 (15.70)	27.23	11.28	22.78	2.69	14.59	5.89	10.99	4.78	3.66	1.25	0.45	16.89	3.66
5'th percentile	3.11	1.35 (1.35)	6.88	9.24	5.64	3.60	5.78	8.50	0.30	0.20	12.90	25.30	0.10	38.14	35.00
95'th percentile	7.81	35.72 (27.83)	81.54	39.60	79.04	10.60	53.64	27.26	18.73	9.47	24.98	29.60	1.58	91.82	46.58

Table 2. Concentrations of TVOCs and specific pollutant groups during the 4 sampling periods (toluene equivalents ($\mu g/m^3$).

	Indoors TVOCs, of which:	Aromatic Compounds	Terpenes	Aldehydes	Acids	Outdoors TVOCs	I/O
06/06/2002	1497.3	3.1	21.8	27.3	8.0	110.5	13.6
09/06/2002	1407.7	1.7	2.7	21.7	9.0	108.1	13.0
07/03/2002	120.4	4.4	2.8	23.5	3.8	37.2	3.2
10/03/2002*	141.6	3.3	0.0	11.5	0.3	35.2	4.0
16/01/2003	457.4	45.3	36.6	24.3	3.8	40.5	11.3
19/01/2003*	457.6	53.3	106.5	21.8	3.0	100.9	4.5

[1] TEOM Instruments
[2] ML® 9841A Nitrogen Oxides Analyzer
[3] Ozone Analyzer, model 400A
[4] Values in parenthesis refer to indoor concentrations during the non-activity hours

* No indoor activities

or slightly elevated due to car exhaust. In the indoor environment the concentrations were quite normal during the March 2002 period but at very high level (exceeds comfort level) during the June 2002 period and the January 2003 period. At the Steingorgveien house infiltration of the outdoor air indoors, wall-painting and floor covering (wood) emissions were the dominating sources of VOCs indoors. The neighbour house was painted during the measurement period therefore elevated concentrations were observed both indoors and outdoors. At the Collets gate apartment elevated concentrations of limonene and hexanoic acid were found in the sample probably due to the storage of citrus-fruits. Other sources are emissions from wall painting, wood and floor covering. The indoor concentrations were slightly enhanced on days with no indoor activities since no ventilation occurred and concentrations of TVOC built up in the indoor environment. The contribution of aldehydes and terpenes to TVOCs is also indicative of the recent renovation of the apartment.

The received human dose was estimated during the June 2002 measurement period at the Steinborgveien house. The daily dose indoors was less than the outdoor in the absence of activities. In addition the dose of Cl^-, Mg^+ and Ca^{2+} was dominant in the upper respiratory system along with the dose of K^+ and Na^+ particles since these particles are found in the coarse mode. On the other hand NH_4^+ and SO_4^{2-} particles dose is equally distributed in the lower and the upper region. A large part of the particles lung burden consists of carbonaceous particles. The dose of total carbonaceous particles accounted for the 23% of the average particle dose.

4. CONCLUSIONS

Indoor and outdoor measurements of PM and gaseous compounds were performed at Oslo. The results indicated that the indoor/outdoor concentration and chemical composition/size distribution of particles is affected by anthropogenic and natural sources outdoors, the contribution of which varies by season and air mass origin. In addition the air exchange rate and indoor activities (cooking, cleaning etc.) and sources (building materials, furniture) determine the indoor concentrations. Finally, the respiratory tract dose of a typical individual exposed to the above concentrations was higher outdoors and enhanced indoor on days with indoor activities.

5. ACKNOWLEDGEMENTS

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THE STOCKHOLM TRIAL – EFFECTS ON AIR QUALITY AND HEALTH

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ABSTRACT
The Stockholm Trial has resulted in a reduction in the volume of traffic in Stockholm's inner city. This has led to lower emissions of carbon dioxide, particles and nitrogen oxides, which in their turn in have lowered the contribution from traffic to total levels of particles (PM10) and nitrogen dioxide. The emissions of particles and nitrogen oxide from road traffic fell by 8%-12% in Stockholm's inner city. For all road traffic in the City of Stockholm this corresponds to between 3%-5%. This means that, with a congestion tax, both the average particle levels for the population of Stockholm and the nitrogen oxide levels would be some percent lower. In total for the entire Greater Stockholm area (1.44 million inhabitants, 35 x 35 km), it is estimated that between 25 and 30 fewer premature deaths would occur per year as a result of a reduction in long-term exposure to particles.

1. INTRODUCTION
Effects of traffic pollution on health is well documented. In Swedish studies NO_2 has been used as marker for traffic exhaust and showed acute effects on asthmatics (Forsberg et al., 1998), but also long-term exposure has been shown to be associated with air way sickness (Forsberg et al., 1997), lung cancer (Nyberg et al., 2000), cardiovascular mortality (Rosenlund et al., 2006) and incidence of adult asthma (Modig et al., 2006). Actions to reduce air pollution from traffic are therefore needed. On June 2, 2003, Stockholm City Council proposed testing congestion charging of traffic — the Stockholm Trial. On June 16, 2004 the Swedish Parliament, the Riksdag, adopted the Congestion Charge Law (SFS 2004:629). The law made it possible to charge a congestion tax in Stockholm up to July 31, 2006. The Stockholm Trial consisted of three parts: extended public transport, congestion tax and more park-and-ride sites in the city and the county. The objectives of the trial were:

- The number of vehicles in the congestion-charging zone during the peak periods of the morning and afternoon should be reduced by 10 to 15%.
- Traffic flows should improve on the most heavily trafficked roads in Stockholm.
- Emissions of carbon dioxide, nitrogen oxides and particles in inner city air should be reduced.
- People residing or staying in the inner city should experience an improvement in the urban environment.

The effect of the Stockholm Trial on emissions and levels of air pollutants is presented in this report as well as the estimated consequences for health.

2. METHODOLOGY
The report focuses on inhalable particles (PM10), and nitrogen oxides (NOx and NO_2), but estimates of emissions have also been carried out for other air pollutants, such as the greenhouse gas carbon dioxide. The estimates are based on traffic analyses carried out in connection with the trial. Before and during the trial air quality has also been measured at 20 or so sites in the Stockholm area. The effects of the Stockholm Trial on air quality and health have been assessed by making use of:

• Fixed continuous monitoring of air pollutants and meteorology in Stockholm with a high time resolution, i.e. hour by hour.
• Monitoring of air pollutants before and during the Stockholm Trial along main streets and approach roads, partially with lower time resolution, day or month.
• Estimates of emissions from road traffic with emission factors according to the National Road Administration and traffic data from traffic monitoring during the Stockholm Trial and the Road and Traffic Research Institute's model estimates.
• Estimates of levels and exposure with an air quality dispersion model (SMHI, Airviro).

3. RESULTS AND DISCUSSION
3.1 Effects on emissions
Compared with the situation in 2006 with no Stockholm Trial, it is estimated that nitrogen oxides in the Greater Stockholm area (1.44 million inhabitants, 35 x 35 km) would decrease by approx. 55 tons, of which most would be a result of a reduction in emissions in Stockholm's inner city (Table 1). For particles, PM10, the corresponding reduction would be approx. 30 tons, of which about two thirds would be a result of reductions in emissions in the inner city. Both particles formed as a result of road surface wear (primarily due to the use of studded tyres), and particles emitted from exhaust pipes would have decreased. It is estimated

that carbon dioxide emissions would have fallen by approximately 41,000 tonnes. For the Greater Stockholm area the percentage reductions in emissions would be approx. 1%-3%, for Stockholm city approx. 3%-5%, and for the inner city approx. 8%-14%. The emissions also include the results of the extended bus traffic associated with the Stockholm Trial (e.g. direct buses to and from inner city).

Table 1. Estimated reductions in emissions from road traffic in Stockholm for a situation for 2006 with the Stockholm Trial.

	Inner city:		City of Stockholm:		Greater Stockholm*:	
	tons/year	per cent	tons/year	per cent	tons/year	per cent
Nitrogen oxides. NOx	45	-8.5%	47	-2.7%	55	-1.3%
Carbon monoxide. CO	670	-14%	710	-5.1%	770	-2.9%
Particles. PM_{10} total	21	-13%	23	-3.4%	30	- 1.5%
"road wear particles"	19	-13%	21	-3.3%	28	-1.5%
"exhaust particles"	1.8	-12%	1.8	-4.4%	2.1	-2.4%
Volatile organic compounds, VOC	110	-14%	120	-5.2%	130	-2.9%
benzene. C_6H_6	3.4	-14%	3.6	-5.3%	3.8	-3.0%
Carbon dioxide. CO_2	36,000	-13%	38,000	-5.4%	41,000	-2.7%

** defined as an area of 35 km x 35 km across central Stockholm.*

3.2 Effects on air quality

The average levels of nitrogen oxides (NOx) are estimated to fall by at most 5-10 µg/m^3 (microgrammes per cubic metre of air) and the levels of particles, PM10 by at most 2-3 µg/m^3. The greatest improvements in the air quality are estimated to be found along the Klarastrandsleden bypass, Centralbron Bridge, Valhallavägen and Sveavägen, and at the entrances to the Söderleds Tunnel (Figure 1). The levels of air pollutants increase in an area around the tollfree Essingeleden Södra Länken bypass. But as a whole, considerably more people in Stockholm experienced reductions in air pollutants and better air quality compared with those who experienced increased levels.

On Hornsgatan it is estimated that levels of nitrogen oxides (NOx) at street level would fall by approx. 7%-8%, levels of nitrogen dioxide (NO_2) by approx. 3%-4% and levels of particles (PM10) by 5%. The improvement is sufficient that the environmental quality standard (to protect public health) as regards the mean annual value for particles, PM10, is not exceeded on Hornsgatan. On the other hand, environmental quality standards are still being exceeded as regards high daily mean values both for particles, PM10, and nitrogen dioxide. On Sveavägen it is estimated that levels of nitrogen oxides (NOx) at street level would fall by 3%, levels of nitrogen dioxide (NO_2) by approx. 1%-2%, and levels of particles (PM10) by 4%. The improvement is sufficient for the environmental quality standard of the annual mean value for nitrogen dioxide, NO_2, not to be exceeded on Sveavägen. Just as on Hornsgatan, however, the environmental quality standards for high mean daily values are still being exceeded for both particles, PM10, and for nitrogen dioxide.

On Norrlandsgatan the levels of nitrogen oxides (NOx) at street level are estimated to have fallen by 11%, the levels of nitrogen dioxide (NO_2) by approximately 5-6% and the levels of particles (PM10) by 7%. The improvement is sufficient for the environmental quality standard for the annual mean value for nitrogen dioxide, NO_2, not to be exceeded on Norrlandsgatan. Here, too, the environmental quality standard is, however, exceeded as regards high daily mean values both for particles, PM10 and for nitrogen dioxide. On S:t Eriksgatan (south of S:t Eriksbron Bridge) the air quality is estimated to be unchanged at street level. A little more traffic and somewhat higher emissions are balanced by the fact that the urban background level of air pollutants has fallen. The environmental quality standard for mean annual values is being met, but the standard for high mean daily values of particles, PM10 is being exceeded.

On Valhallavägen (NW of Lidingövägen) the levels of nitrogen oxides (NOx) at street level are estimated to have fallen by 12%, and the levels of nitrogen dioxide (NO_2) and particles (PM10) by approximately 7-8%.

The improvement is not sufficient to meet the environmental quality standard for high mean daily values of particles, PM10, on Valhallavägen.

Along the Essingeleden bypass the environmental quality standard for protecting public health is also being exceeded. The increased traffic on this road, with the Stockholm Trial, is estimated to increase daily mean levels by approximately 3 µg/m3 (micrograms per cubic metre of air) for nitrogen oxides, NOx, and up to approximately 2 µg/m3 for particles, PM10. In order to meet the environmental quality standards for particles along the Essingeleden bypass, major reductions in traffic emissions are needed.

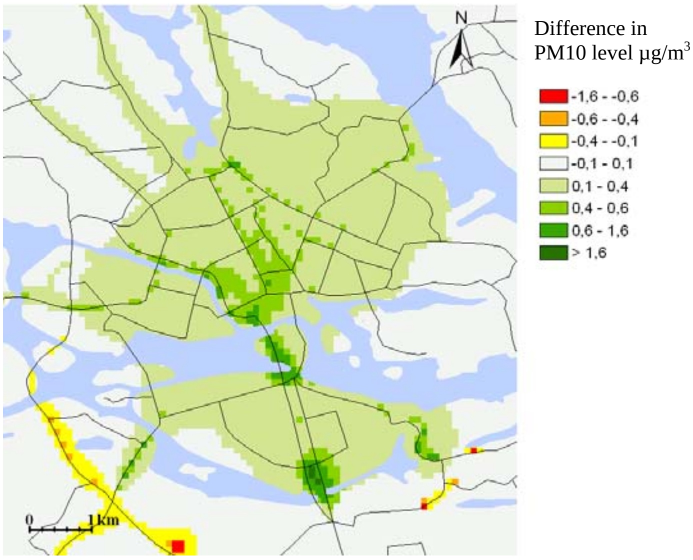

Figure 1. Changes in levels of particles (PM10, mean annual levels) with the Stockholm Trial compared with levels without the congestion charge for 2006. Within the green areas the levels have fallen, within yellow to red areas there is an increase in levels. In the inner city changes refer to rooftop height (not street canyon).

Comparisons between ***observed*** levels of air pollutants that have been measured during the Stockholm Trial 2006 (January to July 2006) with corresponding months in 2003, 2004 and 2005 show that the variations in levels of pollutants between different years can be significant. This depends to a great extent on the fact that meteorological conditions are very important when studying a short period of time. Particle levels in the air, for example, depend to a high degree on the humidity of the road surfaces. During the spring of 2006 Stockholm received a large amount of precipitation and the snow melted late, which caused particle levels to be unusually low. The influence of the weather means that the total levels measured during the Stockholm Trial cannot provide quantitative information on how significant the reductions in traffic emissions have been for levels of air pollutants. In the long term, for example if the Stockholm Trial becomes permanent, air quality in Stockholm will be affected most by reductions in emissions.

A more detailed analysis of the measurements on the inner city streets of Hornsgatan and Sveavägen during the Stockholm Trial 2006, shows that the contribution of traffic emissions to nitrogen oxide levels has decreased. However, contributions to emissions from the new direct buses could at certain times be proven in the measurements on Sveavägen.

3.3 Effects on health

Recently studies have been presented which use the differences in the level of exposure to exhausts measured as nitrogen dioxide (NO_2) or nitrogen oxides (NOx) in cities. Studies of this kind now exist from Holland, New Zealand, France and Norway (Hoek et al., 2002; Scoggins et al., 2004; Filleul et al., 2005; Nafstad et al., 2004). These studies have arrived at very similar exposure response factor for the importance of traffic emissions for mortality. They found that the increase in mortality is 12%, 13% and 14% per 10 µg/m^3 increase in nitrogen dioxide, NO_2, respectively, in the studies from Holland, England and France. The Norwegian study, carried out on adult men, has been considered the most relevant for the consequence analysis of the effects of the Stockholm Trial. In that study they arrived at an increased premature mortality of 8% per 10 µg/m^3 increased level of nitrogen oxides, NOx. If one accepts the exposure-response function for NOx from the Norwegian study (8% per 10 µg/m^3) and a mortality frequency of 1,000/100,000 inhabitants per year, then the reduction in level in Stockholm's inner city, with its approx. 350,000 residents, is expected to result in approx. 20 to 25 fewer premature deaths per year. We have then taken into account that even the low mortality among younger people is influenced to the same extent calculated as a percentage by lower levels, as other studies have shown that particles even affect mortality in children. The fact that younger people are included has however negligible significance on the result. Using the same assumptions for the entire assessment area (approximately Greater Stockholm) with 1.44 million inhabitants (including the inner city), the reduction in level is estimated to result in avoiding approximately 25 to 30 premature deaths per year.

4. CONCLUSIONS

The reduction in traffic volume has reduced the contribution from traffic to the total levels of particles (PM_{10}) and nitrogen dioxide. The emissions of particles and nitrogen oxides from road traffic in Stockholm's inner city are estimated to fall by between 8% and 12%. For road traffic in the City of Stockholm this corresponds to between 3% and 5%. The reductions in emissions of primarily nitrogen oxides would have been higher without the extended bus traffic during the Stockholm Trial. The reductions in emissions overall mean that the interim target for the Stockholm Trial, that emissions of air pollutants should fall, has been achieved.

The average levels of nitrogen oxides (NOx) are in some places estimated to fall by up to approx. 2-3 $\mu g/m^3$. The greatest improvements in air quality are estimated to occur along the Klarastrandsleden bypass, Centralbron Bridge, Valhallavägen and Sveavägen, and at the entrances to the Söderleden Tunnel. Greater levels of air pollutants are found around the Essingeleden and Södra Länken by-passes, but considerably more Stockholmers now have reduced levels of air pollutants and better air quality compared with those who have higher levels. Average particle levels for the population of Stockholm (Greater Stockholm) are estimated to be a percentage point or two lower with congestion charging.

In total, for the whole of the Stockholm area (1.44 million inhabitants, 35 x 35 km), it is estimated at between 25 and 30 fewer premature deaths each year will result from long term exposure to particles. Emissions from road traffic in Stockholm also cause certain illnesses, exacerbates respiratory problems and leads to allergies among certain sensitive individuals. These are reduced with the congestion tax. Environmental quality standards to protect public health will be met to a greater extent than before with the reductions in emissions. The effects of this trial are however not sufficient for environmental quality standards to be met everywhere within the Stockholm region.

Comparisons between pollutant levels measured during the Stockholm Trial (the period January to July 2006) with the corresponding period in 2003, 2004 and 2005, show that variations in pollutant levels between different years can be significant. This results in large part from different meteorological conditions. Overall measured levels during the Stockholm Trial therefore cannot provide a quantitative answer of importance of the traffic reductions for the levels of air pollutants.

5. ACKNOWLEDGEMENTS

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HEALTH RELATED MEASUREMENTS AND EXPOSURES FROM TONG LIANG, CHINA

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ABSTRACT

Chemically speciated $PM_{2.5}$ and particle-bound polycyclic aromatic hydrocarbon (PAH) measurements were made at three sites near urban Tong Liang, Chongqing, a Chinese inland city where coal combustion is used for electricity generation and residential purposes outside of the central city. Ambient sampling was based on 72-hr averages between 3/2/2002 and 2/26/2003. Elevated $PM_{2.5}$ and PAH concentrations were observed at all three sites, with the highest concentrations in winter and the lowest in summer, reflecting a coupling effect of source variability and meteorological conditions. The $PM_{2.5}$ mass estimated from sulfate, nitrate, ammonium, organics, elemental carbon, crustal material, and salt corresponded with the annual average gravimetric mass within ±10%. Carbonaceous aerosol was the dominant species, while positive correlations between organic carbon and trace elements (e.g., As, Se, Br, Pb, and Zn) were consistent with coal-burning and motor vehicle contributions. Ambient particle-bound PAHs of molecular weight 168–266 were enriched by 1.5 to 3.5 times during the coal-fired power plant operational period. However, further investigation is needed to determine the relative contribution from residential and utility coal combustion and vehicular activities.

1. INTRODUCTION

The extensive use of coal as an energy source and high-sulfur petroleum products contribute to high concentrations of suspended particulate matter (PM) and sulfur dioxide (SO_2) in China (WHO, 2001). Fine particles can be inhaled and deposited in the human respiratory system, or translocated to the lymphatic or circulatory system, resulting in adverse health effects. In addition to SO_2, coal combustion without adequate control measures also emits fly ash, which contains toxic metals and carbonaceous material including gas- and particle-phase polycyclic aromatic hydrocarbons (PAHs). Several studies have demonstrated an association between specific PAHs and early genetic damage during breast and lung carcinogenesis.

The Tong Liang Infant Environmental Health and Molecular Epidemiology (TLIEHME) Study was initiated to assess the health effects of energy-related air pollution on newborns in China (Perera et al., 2005). The goal of the study was to investigate possible improvements to the health of children resulting from reductions in coal-burning emissions. This study collected and analyzed blood samples from newborns before and after a scheduled permanent shutdown of the coal-fired power station in March 2003 (~150 samples for each period) to test a hypothesis that carcinogens in fetal and newborn blood are lower when the mother's exposure to airborne chemicals from coal-burning is reduced. Ambient $PM_{2.5}$ (particles with aerodynamic diameters less than 2.5 micrometers [µm]) mass, elements, ions, carbon, and PAHs were measured in Tong Liang between 3/2/2002 and 2/26/2003. The objectives of this monitoring were to: 1) investigate the influence of a coal-fired electricity generator on local air quality; and 2) determine the spatial and temporal variation of concentrations and human exposures in this urban area. Reported here are PM, PAHs, and other pollutant concentrations that might be attributed to the power plant emission. Since the power plant operated only part of the year, effects of its emissions can be evaluated by comparing the ambient concentrations between the operational and non-operational periods.

2. METHODOLOGY

MiniVol samplers (Airmetrics, Eugene, OR, USA) equipped with tandem PM_{10} (particles with aerodynamic diameters less than 10 µm) and $PM_{2.5}$ impactors were used to acquire particles on filter substrates for subsequent chemical analysis at the Desert Research Institute (Reno, NV, USA). Integrated 72-hour samples (1 p.m. to 1 p.m. local time) at a flow rate of 5 L/min were taken continuously at three sites to represent human outdoor exposure in urban/commercial/residential areas in the central (Site A), northern (Site B), and southern (Site C) parts of Tong Liang. Site A was located on a second-story patio attached to an occupied apartment near the Tong Liang town center in an urban/commercial area of moderate population density. Site B was on a second-story rooftop at the existing Tong Liang Environmental Protection Bureau's air quality monitoring station, ~1.5 km north of Site A, in a densely populated urban area where emissions from cooking and refuse burning are prominent. Site C was on a sixth-story rooftop in a commercial/residential area, ~2 km south of Site A and ~1 km north of the local coal-fired power-generating station. The three sites were selected to contrast different microenvironments and distances from the power plant. Detailed chemical analyses were described in Chow et al.

(2006), except for non-polar organics, including PAHs, *n*-alkanes, hopanes, and steranes, which were analyzed on quartz fiber filters by the in-injection port thermal desorption (TD)-Gas Chromatography/Mass Spectrometry (GC/MS) method (Ho and Yu, 2004).

3. RESULTS AND DISCUSSION

Annual Averages. Annual average $PM_{2.5}$ varied by <20% among the three sites, with 123.2±65.4 µg/m^3 at Site C (closest to the power plant), 115.0±52.7 µg/m^3 at Site B, and 107.9±43.2 µg/m^3 at Site A. Every-sixth-day samples (n=61) at Site A (following the U.S. Environmental Protection Agency's [EPA] sampling schedule for compliance networks) reported an annual average $PM_{2.5}$ of 112.2±69.9 µg/m^3, differing by <10% from the annual average 72-hr measurements at Site A. This shows that a sixth-day sampling frequency is sufficient to represent annual averages. The U.S. National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ are 15 µg/m^3 for an annual average and 65 µg/m^3 for a 24-hour average. Tong Liang's annual averages were 7 to 8 times higher than 15 µg/m^3 for $PM_{2.5}$ mass. Twenty-four-hour $PM_{2.5}$ mass concentrations ranged from 30.2 µg/m^3 (6/13/2002) to 418.5 µg/m^3 (12/16/2002), with 75% of them exceeding daily average $PM_{2.5}$ of 65 µg/m^3.

Figure 1 shows that major chemical components explain 91%, 102% and 110% of the $PM_{2.5}$ mass at sites A, B, and C, respectively; indicating a nearly complete mass closure by the measured species. Carbonaceous material (the sum of OM and EC) was the largest $PM_{2.5}$ component, ranging from 52.8 µg/m^3 (Site A) to 71.8 µg/m^3 (Site C), and accounting for 49–53% of $PM_{2.5}$ mass. OC/EC ratios between 3 and 4 are consistent with contributions of primary emissions from combustion sources other than diesel-fueled vehicle exhaust. Higher OC/EC ratios may be associated with secondary organic aerosol (SOA) or biogenic contributions.

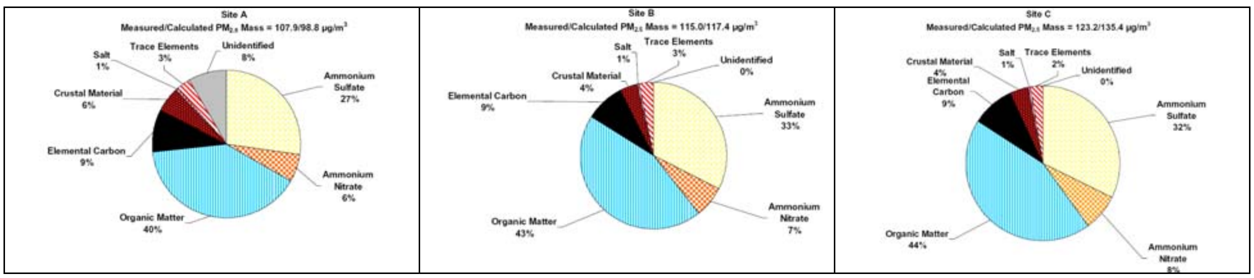

Figure 1: Annual average (3/2/2002-2/26/2003) $PM_{2.5}$ material balance at three sampling sites in Tong Liang, China. Ammonium sulfate = $1.38 \times SO_4^{=}$; Ammonium nitrate=$1.29 \times NO_3^-$; OM = 1.4 × OC (to account for the associated H, N, and O); Salt = 2.54 × Na^+; and Crustal Material = ([2.2 × Al] + [2.49 × Si] + [1.63 × Ca] + [2.42 × Fe] + [1.94 × Ti]) to account for the associated O with the mineral form of these elements. Other trace elements include 35 of the 40 elements (Na to U, excluding Na, Mg, Al, Si, Ca, Fe, and Ti).

The second-largest contributor is ammonium sulfate [$(NH_4)_2SO_4$], which ranged from 29.4 µg/m^3 (Site A) to 43.7 µg/m^3 (Site C) and accounted for 27–33% of the $PM_{2.5}$ mass. $SO_4^{=}$ originates mostly from the oxidation of SO_2 emitted during the combustion of sulfur-containing bio- or fossil fuels. SO_2 to $SO_4^{=}$ conversion can be very rapid in moist environments. The high $SO_4^{=}$ levels are related to the extensive use of sulfur-rich coal in China. Ammonium nitrate (NH_4NO_3, 6.7 to 10.6 µg/m^3) accounted for 6–8% of $PM_{2.5}$. Nitric acid (HNO_3) and NO_3^- are predominantly formed in the atmosphere through oxidation of reactive nitrogen oxides (NO_x) emitted from high-temperature combustion in engines or boilers. NH_4^+ is the dominant cation associated with NO_3^- and $SO_4^{=}$ in $PM_{2.5}$. Na^+ is only 2–3% of NH_4^+ in terms of molar abundance. The molar ratio of NH_4^+ over $SO_4^{=}$ and NO_3^- indicates a full neutralization at sites A and C. At the northern Site B, however, $PM_{2.5}$ was slightly acidic.

Trace-species concentrations at Site C were higher than levels at the other two sites by $\geq$50%. Crustal material (CM) ranged from 4.7 to 6.1 µg/m^3 and accounted for 4–6% of the $PM_{2.5}$ mass. Coal-combustion-related As and Se were detectable, averaging 30-40 ng/m^3 and 10-40 ng/m^3, respectively. The maximum As level of 0.12 µg/m^3 was found on 05/19/2002 at Site A and on 07/12/2002 at Site C. These levels are much higher than those found in U.S. cities where coal combustion emissions are negligible. Two bio-accumulative heavy-metals, cadmium (Cd) and mercury (Hg), which are probable carcinogens and neurotoxic, were detected at annual averages of 22 to 28 ng/m^3 and 14 to 19 ng/m^3, respectively. Similar ambient levels of Cd and Hg have been reported for northern hemisphere urban and rural areas. The peak 72-hr Cd concentration was 14 ng/m^3 (5/13/2002 at Site A). Average strontium (Sr) concentrations ranged from 30 to 40 ng/m^3 with a maximum of 120 ng/m^3 at Site A on 5/13/2002, indicative of impacts from nearby (within 30 km) $SrCO_3$ production. Average K^+ concentration levels ranged from 2,600 to 3,600 ng/m^3. Although K^+ is often used as a marker for vegetative burning, it may also result from other combustion sources, such as waste incineration. Residual oil-related nickel (Ni) and vanadium (V) levels were less than 1.5 ng/m^3.

The total *n*-alkane concentrations at Site A, B, and C were 726, 955, and 1,058 ng/m^3, respectively. The main sources of *n*-alkanes include petroleum residues and biogenic emissions (e.g., plant wax and wood burning). Hopanes and steranes are known molecular markers of aerosol emissions from fossil fuel utilization, ranging from 9.9 ng/m^3 (Site A) to 15.9 ng/m^3 (Site C) for hopanes, and 3.9 ng/m^3 (Site A) to 5.8 ng/m^3 (Site C) for steranes. The annual average sum of particle-phase PAH concentrations ranged from 225 ng/m^3 (Site A) to 286 ng/m^3 (Site C). PAHs such as, chrysene (18-21 ng/m^3), benzo[b,j,k]fluoranthene (37-46 ng/m^3), benzo[e]pyrene (30-68 ng/m^3), indeno[123-cd]pyrene (8.1-8.9 ng/m^3), benzo[ghi]perylene (8.7-9.9 ng/m^3), and picene (1.5-2.5 ng/m^3) are useful markers for coal combustion, natural gas combustion, and gasoline vehicle emissions. Concentrations of carcinogenic PAHs, such as benz[a]anthracene, benzo[a]pyrene, dibenzo[a,h]anthracene, ranged from 2.6-17 ng/m^3. Highest PAH concentrations were found at Site C: 6.7-24.8% and 2.4-12.6% higher than Site A and Site B, respectively. These levels were one to two magnitudes higher than those found in U.S. cities but close to levels measured in Nanjing, China where coal combustion emissions are also dominant. The differences between those values and the monthly average PAH measured by the solvent extraction method (Chow et al., 2006) were within ±6.9%. The satisfactory agreement of PAH measurements in the inter-method comparison demonstrates the comparability of the TD method in the determination of non-polar organic compounds in the aerosol filter samples.

Temporal Variations. Figure 2(a) presents the time series of 24-hr $PM_{2.5}$ measurements at Site A. Highest monthly averages occurred in December and January (>200 μg/m^3). Monthly averages were <150 μg/m^3 between April and May. From 5/10/2002 through 11/10/2002, when the coal-fired power plant near Site C was not operating, the $PM_{2.5}$ concentration remained relatively low; however occasional high levels (>150 μg/m^3) were still recorded. This annual cycle partially reflects synoptic meteorology, which is largely driven by the surface radiative effect of the Asian continent. In summer, strong surface heating causes low-level convergence, producing substantial precipitation and PM scavenging. In winter, surface radiative cooling dominates, leading to subsidence that suppresses the atmospheric boundary layer (ABL) depth, accompanied by lower precipitation and stagnant conditions that enhance pollutant accumulation. Cold, dry weather begins in late December and persists through early February, increasing demand for heating through residential or utility coal combustion. With increased precipitation in summer, hydropower production replaces coal-fired boilers to meet electricity demands. The use of coal for commercial and residential cooking, however, continues throughout the year. To investigate the change of air pollutants with and without the operation of the local coal-fired power plant, seasonal variations of $PM_{2.5}$ were examined.

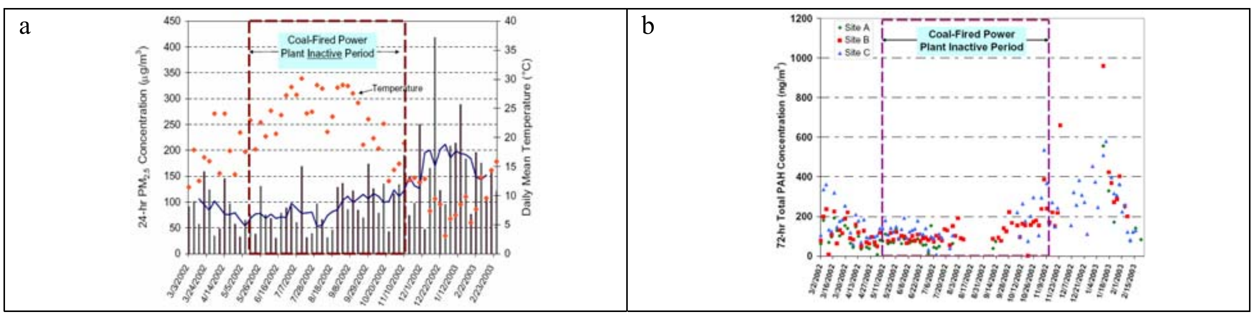

Figure 2 (a) Time series of 24-hr $PM_{2.5}$ measurements taken every sixth day at Site A in Tong Liang, China from 3/2/2002 to 2/26/2003 (bars). Also shown is daily mean temperature from the Chongqing surface measurements (diamonds). The solid line indicates the monthly moving averages of $PM_{2.5}$ concentration. (b) Time series of 72-hr total PAHs measurements taken at Site A, B, and C in Tong Liang, China from 3/2/2002 to 2/26/2003.

The power plant remained active during winter (December to February) and 80% of spring (March to May). Overall coal-burning activity was more intense in winter. Geopotential height decreased by > 75% from the winter to summer. A strong prevailing wind was observed during summer at 2,000 m above ground level (AGL). Seasonal variations exceeded the spatial variations for major $PM_{2.5}$ components. OM concentrations in winter (82.6–107μg/m^3) were more than twice as high as those in spring (33.8–42.2 μg/m^3) and summer (30.7–40 μg/m^3); despite the possibility of long-range transport and SOA formation in summer. The trend for EC is similar, with an OC/EC ratio between 3 and 4. The EC level during winter was 6 to 9 times that found during other seasons.

Although SO_2 oxidation efficiency is expected to be higher during summer, the highest $(NH_4)_2SO_4$ of 63.5 μg/m^3 occurred at Site B in winter. This might be explained by the combination of enhanced coal combustion coupled with a shallower ABL. Similar levels (within ±10%) were found for carbonaceous aerosol and $(NH_4)_2SO_4$

between spring and summer. This implies that contributions from local power plant emissions to $PM_{2.5}$ were low during the spring. NH_4NO_3 and CM also peaked during winter and show appreciable spring-summer differences. Lower temperatures during winter favor NH_4NO_3 over HNO_3. Winter NH_4NO_3 concentrations were 15–18 $\mu g/m^3$, compared with 5–7 $\mu g/m^3$ in spring and 3–4 $\mu g/m^3$ in summer. CM was relatively low, but its spring highs reached 7.3 $\mu g/m^3$ in contrast to the summer lows of less than 4 $\mu g/m^3$. Coal-combustion-related trace elements, including Zn, As, Se, Cd, and Hg were enriched during winter, with the highest concentrations at Site C. Zn reached nearly 0.24 $\mu g/m^3$ in winter. At Site C, As and Se were strongly correlated with each other and with OC and $PM_{2.5}$ mass ($r \geq 0.87$) for the spring period, implying contributions from a common source, possibly the local coal-fired power plant and/or residential coal burning. Multiple source contributions explain the lower wintertime correlations of As and Se with $PM_{2.5}$ ($0.76 < r < 0.84$). When the power plant was inactive during summer, lower correlations were found between $PM_{2.5}$ and coal-combustion-related trace elements.

Figure 2 (b) presents the time series of 72-hr total PAHs concentration at the three sites. During the study, precipitation was reported on 105 days, or ~1/3 of the sampling period. Intense rains occurred in summer, resulting in instrument malfunction and occasional missing data. The power plant began operation during mid-November (11/11/2002), but the PAH concentrations in fall (September and October) were relatively high, likely due to contributions from other combustion sources. The power plant operations did not correspond with increases for most low molecular weight (MW) PAHs (i.e., naphthalene [128] to fluorene [166]). Medium MW PAHs (168–266) increased by 1.5 to 3.5 times during the power plant's operational period. High MW PAHs (>266) were not enriched during the operational period. The distinct temporal trends of indeno[123-cd]pyrene and benzo[ghi]perylene are consistent with contributions from vehicle exhaust and other combustion sources in the area. The temporal trend of benzo[b,j,k]fluoranthene and benzo[a]pyrene tracked OC and $PM_{2.5}$ mass, with concentrations threefold higher during winter than summer. The concentrations of benzonaphthothiophene increased by 1.6 to 2.4 times in winter, due to higher use of sulfur-containing fuels than in summer. Seasonal changes of PAHs are dominated by the power plant operations and also widespread commercial and residential coal combustions. The potential impact of the local power plant's emissions is examined through the PAH ratios between its operational and non-operational periods.

4. CONCLUSIONS

Elevated ambient $PM_{2.5}$ and PAH concentrations were found in Tong Liang, an inland city in China during the year-long study conducted from 3/2/2002 and 2/26/2003. The annual average $PM_{2.5}$ mass concentrations in Tong Liang ranged from 107.9–123.2$\mu g/m^3$. The major components in $PM_{2.5}$ include OM, EC, $(NH_4)_2SO_4$, and NH_4NO_3. Carbonaceous material accounted for ~50% of the $PM_{2.5}$ mass with an OC/EC ratio between 3 and 4. Temporal variations of $PM_{2.5}$ mass and its major components, as well as PAHs, exceeded their spatial variations. These concentrations were highest during winter, with lowest concentrations found during summer. Along with coal-fired power plant emissions and extensive residential heating and cooking by coal combustion, a shallow inversion layer and low precipitation during winter enhanced the accumulation of pollutants. The $PM_{2.5}$ and PAH concentrations in spring were only slightly higher than those in summer, despite the fact that the coal-fired power plant was operating. Good correlations between OC and combustion related trace elements were observed in winter and spring, but not in summer. Elevated carbonaceous material, including PAH, resulted from a mixture of coal combustion, vehicle exhaust, and vegetative burning, and was influenced by meteorological conditions. Ambient PAHs of medium MW (168–266) were 1.5 to 3.5 times higher during the power plant's operational period. The two high MW PAHs, indeno[123-cd]pyrene and benzo[ghi]perylene, showed opposite seasonal trends and anti-correlations with EC, indicative of influence from other sources, possibly vehicle exhaust, vegetative burning, or regional-scale sources.

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SIMULATING THE IMPACT OF URBAN SPRAWL ON AIR QUALITY AND POPULATION EXPOSURE: A CASE STUDY IN THE GERMAN RUHR AREA

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ABSTRACT

Numerical simulations were performed to evaluate the effect of urban sprawl on air quality and associated human exposure. Working on a highly urbanised area in the German Ruhrgebiet, the atmospheric model AURORA was applied, under conditions representative of the area as it is today. A fair agreement was obtained between simulated and observed pollutant concentrations. Subsequently, an urban sprawl scenario was defined and implemented using spatial modelling techniques, relocating 10 % of the urban population to the cities' periphery. The resulting modified land cover and population density maps of the area were then used as input for traffic and atmospheric dispersion simulations, representative of urban sprawl. In a final step, a calculation was made of human exposure to air pollution. For ozone, urban sprawl was found to lead to a slightly increased exposure. For primary particulate matter the exposure decreased, but when accounting also for secondary particulate matter, the exposure was higher than for the reference situation.

1. INTRODUCTION

Cities exert significant pressures on the environment owing to high levels of consumption of resources related to, among other things, car traffic and building heating. The proximity of these activities to people's living space leads to situations with increased risk to human health. A related problem is that of urban sprawl. Indeed, over the past decades there has been a tendency for people to leave cities to settle in the surrounding greener areas, which has induced an enhanced transport demand. The work described here investigates the effects of urban sprawl on air quality, including human exposure, at the scale of a large urban area and its surroundings. The study area consists of a highly urbanised region in the Ruhr area, located in central North Rhine-Westphalia, in the north-western part of Germany, with a total population in excess of 5.5 million.

The remainder of this paper is organised as follows. In Section 2, a description is given of the atmospheric model and the simulations that were carried out, including a validation study. Section 3 then describes the impact of urban sprawl – through its effect on the modified patterns of land use, traffic, and associated emissions – on simulated air quality and human exposure. Section 4 presents the conclusions.

2. THE ATMOSPHERIC MODEL

Regional air quality was simulated using the AURORA modelling system (Mensink et al., 2001), which employs meteorological fields (wind vectors, diffusion coefficients, radiation, temperature and humidity) produced by the meso-scale atmospheric model ARPS (Xue et al, 2000) to calculate dispersion of pollutants emitted by traffic, industry, and building heating. Advection is treated using the Walcek approach, and diffusion is solved with the Crank-Nicholson scheme. Chemical reactions are calculated with the Carbon-Bond IV model, which was upgraded to include the effects of biogenic isoprene. Particulate matter was calculated as the sum of primary and secondary contributions, the latter consisting of sulphate and nitrate aerosol converted from gaseous species. In this study, the land surface in the ARPS model is represented by the scheme of De Ridder and Schayes (1997), modified to better represent urban surfaces as described in De Ridder (2006).

The simulation domain is shown in Figure 1. Land use-dependent parameters were specified as a function of land cover type, which were interpolated from the CORINE land cover map. Terrain height was interpolated from the Global 30 Arc-Second Elevation Data Set (GTOPO30). Sea surface temperature was derived from NOAA/NASA Pathfinder Advanced Very High Resolution Radiometer (AVHRR) SST imagery. Vegetation abundance was specified as a function of the Normalised Difference Vegetation Index (NDVI) contained in imagery from the VEGETATION instrument onboard the SPOT satellite platform. In order to account for the effect of large-scale atmospheric features, the ARPS model was nested in 6-hourly analysis fields of the global model operated by the European Centre for Medium-Range Weather Forecasting (ECMWF). AURORA was nested within pollutant concentration fields of the CHIMERE transport-chemistry model.

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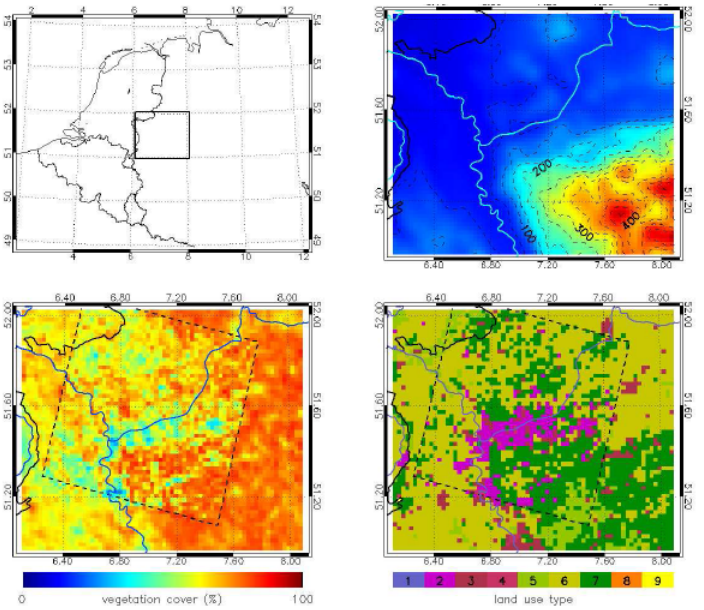

Figure 1. Simulation domain, as indicated by the black square on the upper left panel, with centre at longitude 7.1 degrees and latitude 51.5 degrees. The upper right panel shows orography in metres. The lower left panel gives percentage vegetation cover, and the lower right panel land use types, the purple colours corresponding to urban land use, the green to pastures, crops, and forests.

A three-week period, 1-20 May 2000, was selected to perform the AURORA simulations on. This period was characterized by the presence of a blocking anticyclone over southern Scandinavia, producing weak south-easterly winds, clear skies, and moderately high temperatures over the Ruhr area. The nice weather ended abruptly on the 17th, when a cold front swept over the area. Emissions from industry, shipping and building heating were obtained from the 'Landesumweltamt Nordrhein-Westfalen', the local environmental administration. Traffic-related emissions were calculated using the MIMOSA model (mensink et al., 2000), which uses the COPPERT III methodology to calculate geographically and temporally distributed traffic emissions using traffic information (including fluxes of vehicles and their speeds), which were provided by a traffic flow model. Apart from the above-mentioned anthropogenic emissions, biogenic emissions from forests (isoprene) were also calculated. The simulations carried out here focused on ground-level ozone and PM_{10}, both pollutants having major effects on human health.

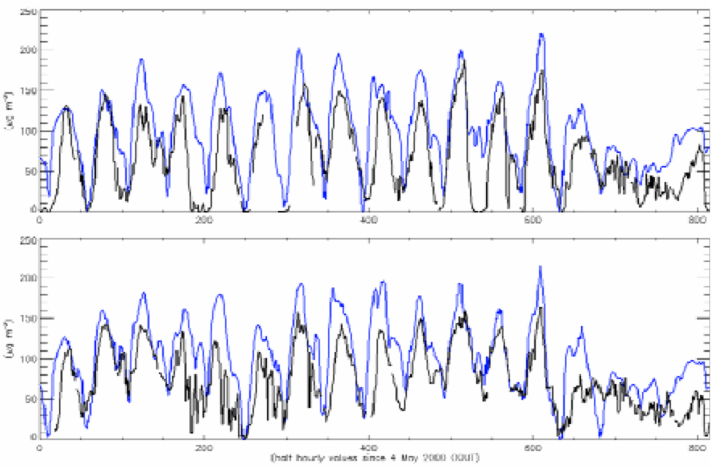

Figure 2. Simulated (blue line) as compared to observed (black line) ground-level O_3 concentrations for the stations Bottrop (upper panel) and Essen (lower panel).

Air quality simulations performed with the AURORA model for the reference situation were validated by comparing model results with available observations from two stations in the area that measured ozone Figure 2. Even though the simulations overestimate the ozone peak concentrations somewhat, the diurnal cycle as well as the behaviour of the model over the entire three-week period is rather satisfactory. In particular the difference of nighttime concentrations between the two locations, which is due to the titration effect (reduction of ozone by no emissions) caused by the intenser traffic at bottrop, is well captured by the model, meaning that the spatial distribution of traffic emissions as well as the chemical processes accounted for in the model perform correctly. Also, the abrupt decrease of ozone concentrations towards the end of the simulation period, caused by a frontal passage, is relatively well simulated.

3. IMPACT OF URBAN SPRAWL

Based on land use maps for the reference case (i.e., containing the current land use patterns), spatial modelling techniques were applied to simulate changes in land use, population, and job density, according to an urban-sprawl scenario. The purpose was to relocate 10 % of the current population to areas near the city. As far as the re-distribution of land use is concerned, spatial simulations were done using the so-called ‘potential model’, which models the probabillity of transition to a given land use type of all the grid cells in the domain. In the present study, a natural area has a high probability of being converted into a built-up area if it is mainly surrounded by already existing built-up areas; otherwise it does not change its state. Already built-up categories remain, and newly created built-up areas are always of the non-industrial type, while existing industrial areas do not change. The resulting land use map for the urban-sprawl scenario was used to update the corresponding spatial distribution of people and jobs. The latter information was then used as input for the traffic model, which simulated corresponding traffic flows and vehicle speeds on a network of road segments for the study area.

Subsequently, the AURORA model was run on the urban-sprawl scenario, using the modified land use characteristics as well as the correspondingly modified traffic flows as inputs. As an example, the associated simulated percentage change of ground-level ozone is shown in Figure 3. Owing to the dominant south-easterly wind direction during this episode, increased ozone values are simulated in a plume extending at the north-west side of the urban agglomeration. The titration effect, on the other hand, which is the consequence of increased traffic emissions, slightly depresses ozone concentrations in the central portion of the domain, which is where the highest population densities occur.

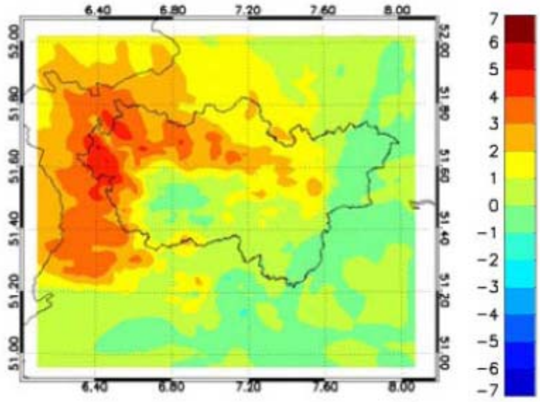

Figure 3. Simulated changes of ground-level ozone concentration in the Ruhr area, following the implementation of the urban-sprawl scenario.

In a final step, a calculation was made of human exposure to air pollution, by multiplying fields of gridded population density with simulated pollutant concentrations. More specifically, the changes between the exposure in the reference versus that in the urban-sprawl situation were evaluated, the main results being the following:

1. Exposure to ozone was found to increase very slightly. Even though the domain-average ozone concentration increased, most of the increase took place in the urban plume, away from the most densely populated areas in the city. Furthermore, the titration effect mentioned above caused a slight decrease in the densely populated city core. Both effects compensating each other, the net effect was very small indeed.
2. With respect to particulate matter, distinction was made between primary and total (i.e., primary + secondary) particulate matter. When considering primary particulate matter only, the exposure was

found to decrease for the urban sprawl situation, despite the increased emissions and domain-wide concentration values of this substance. Even though this may seem surprising initially, this may be explained by the fact that in the urban-sprawl scenario a significant share of the population is relocated to areas outside the dense conurbation, that is, to areas where pollutant concentrations are lower. As a result, exposure is reduced.

3. However, when looking at the total particulate matter, i.e., also including secondary aerosol, it was found that the exposure increased. The cause is that secondary PM, unlike primary, is formed (from its gaseous precursors) at a certain distance from the corresponding emission sources. Consequently, secondary aerosol is much less a ‘local’ pollutant than primary particulate matter, thus also affecting the share of the population in the domain that was relocated to the urban periphery in the urban sprawl scenario.

4. CONCLUSIONS

A brief overview was given of an interdisciplinary study aiming at contributing to the understanding of the relation between city compactness and (exposure to) air pollution. The selected study domain consisted of a cluster of cities in the German Ruhr area, and the case investigated was one of urban sprawl. The methodology was based on evaluating the simulated differences in air pollution exposure between the current situation, and an artificial situation representing the situation in which 10 % of the area’s population was relocated to the areas surrounding the cities.

As a main result it was found that, following the increased emissions (mainly from traffic), human exposure to most pollutants increased, very little so in the case of ozone, and more significantly in the case of particulate matter. However, when considering primary particulate matter only, a decrease in population exposure was found.

5. ACKNOWLEDGEMENTS

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RELATION BETWEEN AMBIENT AIR AND BREATH VOLATILE ORGANIC COMPOUNDS

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ABSTRACT

Breath analysis is a non-invasive tool that can be used to measure body impact following exposure to air pollutants. A method for collecting and analyzing exhaled breath was developed and used to compare individual exhaled breath volatile organic compounds (VOCs) with ambient air VOCs. Exhaled breath was collected in Teflon bags and analyzed using thermal desorption gas chromatography-mass spectrometry. Repeatability of this method was examined following analysis of 10 breath samples of each of three subjects. An environmental health study including 56 3-year-old children in Flanders showed that abundances of VOCs in exhaled breath were clearly related to these in ambient air indicating that interpretation of abundances of VOCs in exhaled breath should always be in relation to its presence in ambient air. It was also demonstrated that a number of VOCs were either significantly retained or cleared by the body.

1. INTRODUCTION

Monitoring the influence of environmental pollution on human health has increased remarkably during the past decades. So far, methods to evaluate exposure to or impact of these environmental pollutants mainly involved the collection and analysis of biological media such as blood or urine. Measuring biomarkers in breath, however, is a very attractive approach to monitoring environmental impact because it is non-invasive and makes repeated sampling possible (Kharitonov 2001). Breath contains valuable information because the pulmonary alveolar membrane consists of a thin barrier separating the air in the alveoli of the lung from the blood in the capillaries. The structure of this membrane allows diffusion of many volatiles. Consequently, inhalation of VOCs - present in the ambient air – will result in absorption of these VOCs by the pulmonary blood supply followed by subsequent distribution of these VOCs throughout the body. Following exhalation, the alveolar air will subsequently be enriched by the VOCs contained within the pulmonary blood to an extent determined by the concentration of the VOCs in the blood and the blood-gas partition coefficients (Kharitonov 1997). This blood-gas exchange model has resulted in extensive research with regard to the breath content and the potential to use breath analysis as a diagnostic tool able to link breath components with pathologies and their early onset. Almost 30 years ago Pauling and co-workers (1971) reported that normal human breath contains a mixture of several hundred volatile organic compounds (VOCs). Since then new techniques have been developed and explored that allow detection of > 1000 volatiles in human exhaled breath (Amann 2005). Most studies, however, focus rather on singular compounds in exhaled breath as a potential biomarker for specific biological effects. Recently, profiling of VOCs in exhaled breath is gaining interest with regard to biomarker research (Basanta 2006 and Phillips 2000). The aim of this study is to develop a method for collecting and analyzing exhaled breath of a variety of human subjects (children, patients, healthy controls) using gaschromatography – mass spectrometry. The method will be used firstly to compare individual exhaled breath VOCs with ambient air VOCs and secondly to find exhaled breath VOC patterns specific for oxidative stress.

2. METHODOLOGY

In order to find suitable combinations of compounds or specific ratios of compounds that relate to specific health effects (metabolic profiles), analytical methods are required that are able to detect, identify and possibly quantify as many VOCs as possible in exhaled breath. Such screening methods, however, often require large sample volumes because of the low concentrations of most VOCs present in the exhaled breath. For this reason a thermal desorption gas chromatography – mass spectrometry (GC-MS) method was further developed using sorbent tubes to preconcentrate large amounts of exhaled breath.

Sampling and Sample Preparation

Exhaled breath of study subjects was collected in Teflon bags. A 3-way valve was used to facilitate the collection of breath for children and for patients with severe airway obstruction. A Gillian personal sampler was used to draw the content of this sampling bag over a sorbent tube containing 3 cm Carbograph1TD/3 cm Carbopack X. For each breath test an equivalent amount of ambient air - present in the sample space which the subjects occupied during the breath test - was sampled on a sorbent tube.
Although breath consists of a relatively ‘clean’ sample matrix compared to blood or urine, the high CO_2 content and humidity can turn out to be a serious challenge with regard to GC-MS.

Because moisture trapped onto the sorbent tubes was found to interfere with GC-MS output, sorbent tubes were purged with 500 mL Helium (50 mL/min) prior to analysis to expel the moisture.

Thermal Desorption Gas Chromatography-Mass Spectrometry

Sampled VOCs were recovered from the adsorbent traps by thermal desorption (Markes International Ltd.). Analysis was performed by GC (HP6890 series) - MS (HP5973 Mass Selective Detector). The column was an RTX 502.2 Crossbond phenyl methyl polysiloxane phase (105 m long, 0.32 mm ID and 1,8 µm film thickness). Thermal desorption, gas chromatography and mass spectrometry parameters are summarized in table 1.

Table 1. Summary of analytical settings

Parameter Desorption Unit	**Setting**	**Parameter Gas Chromatograph**	**Setting**
Primary Desorption Flow	20 mL/min	Column Pressure	140 kPa
Primary Split	Splitless	Initial temperature held 1 min	35 °C
Primary desorption Temperature	350 °C	Final temperature	270 °C
Primary desorption Time	15 min	Temperature ramp	5C°/min→200°C
Cold trap volume	0.02 mL		70°C/min → 270 °C
Cold trap temperature	-10 °C	**Parameter Mass Spectrometer**	
Cold trap packing	Carbograph1TD		
	Tenax TA	Scan mode	EI auto
Trap heating rate	MAX	Temperature	350 °C
Secondary desorption temperature	325 °C	Transfer line temperature	175 °C
Secondary desorption time	3 min	Scan range	25-200 amu
Prepurge time	1 min	Scan frequency	2.14 scans/s
Prepurge flow	20 mL/min		

3. RESULTS + DISCUSSION

Repeatability Experiment

Repeatability of this method was evaluated by determining coefficients of variance for 56 VOCs present in exhaled breath of 3 subjects. Three subjects were asked to fill a 56 L Teflon bag with exhaled breath. 10 x 5 L of exhaled breath of each of the 3 subjects was captured on sorbent tubes and samples were subsequently analyzed. Coefficients of variance for these 56 VOCs (among which 29 VOCs included in the standard gas mixture) are summarized in Table 3. Coefficients of variance for these VOCs were well within acceptable range with 89 % of the coefficients being ≤ 30 %. Multiple ANOVA indicated that coefficients of variance were both subject ($p < 0.00$) and component ($p < 0.00$) dependent (Table 2).

Table 2. Multiple ANOVA results for coefficients of variance

Source	Sum of Squares	Df	Mean Square	F-ratio	P-value
Subject	4068	2	2034	16.3	0.00
VOC	18334	71	258	2.1	0.00
Residual	17690	142	125		
Total (corrected)	40092	215			

Table 3 also shows abundances for VOCs in exhaled breath and ambient air for 2 non-smoking subjects and 1 smoker. VOCs for which areas under the curve in exhaled breath are at least twice these in ambient air are marked blue. These compounds represent VOCs which are most likely to be metabolites produced by the body and some well known metabolites include 2-methyl-1,3-butadiene, acetone, dimethyl sulfide, dimethyl disulfide, ethanol and 1-propanol. VOCs marked grey represent those substances that are most likely to be retained by the body. Data for subject 3 are somewhat different to these of subjects 1 and 2 as it seems that most VOCs (especially the 29 HAPs that were included in the standard gas mixture) are rather produced by the body than retained. A possible explanation could be that smokers are more exposed to these substances and that build up of these VOCs lead to a release of these compounds rather than a steady state situation. Of course a larger sample is needed to test this hypothesis more fully and draw the right conclusions.

Table 3. Coefficients of variance and mean areas under curve (AUC) for 56 VOCs present in ambient air and exhaled breath of 3 subjects.

VOC	subject 1 (non-smoker) CV (%)	subject 1 Mean AUC[1] (counts x 10^4)	subject 1 Mean AUC[2] (counts x 10^4)	subject 2 (non-smoker) CV (%)	subject 2 Mean AUC[1] (counts x 10^4)	subject 2 Mean AUC[2] (counts x 10^4)	subject 3 (smoker) CV (%)	subject 3 Mean AUC[1] (counts x 10^4)	subject 3 Mean AUC[2] (counts x 10^4)
1-pentene	9.2	11.1	12.1	10.8	7.3	5.8	9.3	84.2	27.8
pentane	6.9	36.0	39.9	13.1	12.0	10.9	9.3	35.8	23.6
2-methyl-1,3-butadiene	2.0	11889.4	203.7	5.1	6205.2	33.1	1.7	16190.6	217.5
1-hexene	3.6	6.3	6.8	16.5	3.9	3.6	5.2	30.7	4.2
hexane	3.6	13.2	14.1	14.9	9.9	9.0	20.5	26.3	18.1
ethyl acetate	19.0	249.4	1348.0	45.3	104.6	318.6	4.0	14806.3	137.8
2-methyl hexane	12.0	33.7	41.5	10.0	32.0	24.8	19.8	171.1	78.3
3-methyl hexane	5.1	25.3	25.0	9.5	32.0	28.6	18.0	118.7	82.7
1,1,1-trichloroethane	6.0	5.1	6.5	5.5	10.0	10.0	27.5	4.9	19.6
cyclohexane	3.2	46.6	52.8	10.5	49.9	43.9	16.6	79.4	65.5
2,2,4-trimethyl pentane	2.3	51.7	51.9	12.2	75.8	67.0	17.2	106.6	0.0
heptane	1.8	12.3	14.9	8.1	7.8	6.5	13.0	23.2	14.2
benzene	3.4	273.8	483.9	15.5	327.5	449.1	5.1	5020.9	463.2
trichloroethylene	2.9	3.0	4.7	8.9	5.9	8.6	8.5	25.9	8.9
methylcyclohexane	2.5	31.6	39.2	9.3	20.1	17.2	16.6	36.1	25.8
octane	1.7	31.0	41.7	16.4	9.3	6.1	11.5	34.2	9.8
toluene	3.0	815.1	1292.1	13.9	504.7	685.4	10.2	7752.0	2049.8
tetrachloroethylene	1.4	22.0	13.6	9.5	15.5	14.6	21.7	43.6	15.3
nonane	3.2	53.6	64.0	21.0	7.4	5.2	12.8	16.6	7.4
ethylbenzene	6.2	107.9	223.1	18.2	62.3	127.0	10.4	547.2	196.5
m/p-xylene	6.3	220.0	453.7	19.2	115.8	255.5	12.4	940.3	423.7
o-xylene	6.3	94.8	196.4	20.3	41.9	89.4	17.3	202.1	143.4
styrene	9.3	42.3	196.4	21.3	13.8	29.0	4.3	202.7	56.1
a-pinene	4.5	210.9	325.4	12.3	63.8	20.3	23	77.3	74.2
decane	2.0	237.2	316.8	16.4	32.9	25.4	13.7	78.8	50.1
propylbenzene	4.2	78.4	170.9	35.4	23.0	33.7	15.3	80.0	50.5
1-ethyl-2-methyl benzene	3.9	93.5	218.9	21.7	22.3	36.2	20.0	55.1	53.6
1,2,4-trimethyl benzene	5.1	326.9	804.7	25.5	65.6	108.5	27.7	130.3	168.9
1,2,3-trimethyl benzene	5.0	92.2	210.7	24.4	17.0	28.3	18.6	45.3	43.1
ethanol	10.4	1495.4	2443.2	16.8	1085.1	182.3	10.2	28124.7	126.7
acetone	3.0	43032.3	2673.7	6.3	148773.2	420.2	2.9	52176.3	285.4
dimethyl sulfide	9.8	67.8	8.6	8.9	29.3	0.0	8.3	160.7	0.0
1-propanol	8.7	449.0	28.7	17.3	166.6	10.0	1.4	1406.4	2.4
2-butenal	13.4	510.3	18.9	23.9	36.4	9.8	6.9	219.4	12.5
2-butanone	7.6	214.4	243.8	9.3	94.6	74.8	1.9	11111.8	11.1
2-methyl furan	11.6	96.1	6.1	16.2	42.5	2.1	10.5	149.9	4.8
thiophene	64.3	5.0	1.6	8.9	5.3	3.5	6.5	36.2	2.5
2-pentanone	3.1	278.6	28.9	8.4	87.7	9.3	3	1753.9	13.2
1-methylthiopropane	8.2	57.4	0.6	10.8	3.7	0.0	6	207.9	0.0
3-penten-2-one	46.2	167.6	4.8	40.7	4.9	1.1	28.4	87.1	2.0
dimethyl disulfide	31.1	105.3	14.3	43	8.9	1.7	30.4	83.8	1.8
3-methylthiophene	60.5	271.0	3.1	27.9	72.7	3.3	48.1	64.6	3.3
hexanal	27.9	27.9	462.8	66.1	9.6	2.6	42	17.3	17.1
4-methyl octane	4.9	34.1	41.5	15.1	9.3	8.7	18.2	7.6	5.5
3-furaldehyde	30	15.4	1.4	31.3	3.4	0.4	23.2	11.1	0.3
2,6-dimethyl octane	3.8	28.7	28.4	11.2	5.9	5.0	23.1	11.0	6.8
2-heptanone	10.9	18.0	32.4	51.2	6.9	1.1	9.9	25.2	1.2
2-methyl nonaan	6.4	20.9	22.4	14.9	5.0	4.5	17.3	9.9	8.2
camphene	8.5	28.9	59.2	23.4	14.7	9.8	4.8	72.1	18.2
1-ethyl-3-methyl benzene	5.4	292.9	677.2	22.6	14.7	20.3	22	179.9	17.3
b-pinene	10.4	47.0	168.5	34	17.1	0.9	17.2	17.8	5.2
dimethyl sulfone	21.8	318.2	7.8	53.5	59.2	0.0	29.9	271.7	1.8
phenol	18.5	572.7	113.4	37.6	58.2	3.8	18.6	161.8	0.0
D-limonene	4.9	434.6	840.5	9.9	155.4	11.1	4.7	691.9	70.6
eucalyptol	11	10.3	16.5	31.3	6.8	0.0	13.5	3.7	0.0
methyl-ethenyl benzene	13.1	78.0	157.1	29.4	24.7	5.7	17.5	47.7	15.4

mean AUC breath ≥ 2 x mean AUC ambient air

mean AUC ambient air ≥ 2 x mean AUC breath

CV (coefficient of variation) > 30 %

[1] breath

[2] ambient air

Environmental Health Study

An environmental health monitoring study performed in Flanders allowed for collection and analysis of 7 L exhaled breath of 56 three-year old children living in urban and rural areas in Flanders. Using a standard gas mixture of 52 VOCs, 43 of these 52 VOCs could be identified in breath and their abundance was compared to their abundance in the ambient air the children inhaled previous to the breath test. A logarithmic (with base 2) transformation was applied to normalize both ambient air and breath abundance data.

A paired student T-test at significance level of 0.01 was performed taking differences between abundances of VOCs in ambient air and exhaled breath. Table 4 shows the results of this T-test.

Table 4. Retention (negative t value) /release (positive t value) of VOCs by the body

VOC	p-value	t	VOC	p-waarde	t
isobutane	0.000	3.76	chlorobenzene	0.007	-2.80
2-methyl butane	0.000	4.20	butane	0.008	2.76
2-methyl-1,3-butadiene	0.000	-24.46	2,2,4-trimethyl pentane	0.074	-1.82
dichloromethane	0.000	14.93	heptane	0.084	-1.76
chloroform	0.000	5.32	propylbenzene	0.093	-1.71
3-methyl hexane	0.000	-4.64	3-methyl heptane	0.108	-1.63
1,1,1-trichloroethane	0.000	6.51	hexane	0.130	-1.54
cyclohexane	0.000	-3.88	methylcyclohexane	0.153	-1.45
octane	0.000	-4.19	2-methyl heptane	0.157	-1.43
tetrachloroethylene	0.000	-5.39	2-methyl pentane	0.235	-1.20
nonane	0.000	-4.19	trichloroethylene	0.245	-1.18
styrene	0.000	-14.57	ethylbenzene	0.256	1.15
decane	0.000	-6.13	1,3-Butadiene	0.381	0.88
1-hexene	0.001	-3.47	3-methyl pentane	0.536	-0.62
2-methyl hexane	0.002	-3.21	benzene	0.622	-0.50
1,2-dichloroethane	0.002	3.28	methylcyclopentane	0.631	-0.48
α-pinene	0.003	3.07	pentane	0.648	0.46
carbon Tetrachloride	0.004	3.01	toluene	0.754	-0.32
m/p-xylene	0.004	3.02	1,2,3-trimethyl benzene	0.842	-0.20
1-pentene	0.005	2.92	m-methyl toluene	0.882	0.15
o-xylene	0.006	2.86	1,2,4-trimethyl benzene	0.900	-0.13
ethylacetate	0.007	2.78			

4. CONCLUSION

A sampling and thermal desorption gas chromatography – mass spectrometry method was developed that allows monitoring of C_5-C_{12} VOCs in exhaled breath of subjects as young as 3 years old. A repeatability experiment demonstrated that the method can be considered reliable for at least 56 VOCs present in exhaled breath with 90 % of the coefficients of variance being less than 30 % (and 85 % ≤ 20 %). Bearing in mind that a coefficient of variance of 20 % is normal for standard chemical analysis methods and that we are evaluating a screening method rather than a method optimized to monitor a small selection of compounds, we can conclude that coefficients of variance up to 30 % are acceptable and even much better than those for other whole organism bioassays. The noninvasive nature of breath sampling and analysis makes it more convenient than monitoring blood or urine; however several factors, including the high concentration of water vapor, the short biological half-lives of many absorbed vapors, etc…make screening methods difficult to validate. In these experiments issues such as breakthrough, background, stability of compounds on sorbent tubes and stability of mass detector signal over time were avoided by comparing samples of ambient air and exhaled breath sampled, prepared and analyzed at the same time following the same procedure and correcting for background. However, before a fully validated method can be presented there are still important issues about stability, reproducibility, variability and sensitivity that need to be addressed.

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EXPOSURE TO ROAD TRAFFIC AND RESPIRATORY SYMPTOMS

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ABSTRACT
Air pollution impact on respiratory diseases development has been widely discussed by the scientific community. The aim of the present work was to study this impact in a children population living in Barreiro city, Portugal. To assess children's living environment, a questionnaire was performed to primary school children's parents. A traffic study was also carried out, with a field campaign and through numerical models. Data, such as nitrogen oxides (NO, NO_2, NO_x), carbon monoxide (CO), ozone (O_3) and Sulphur Dioxide (SO_2), from air quality monitoring stations were also considered.
From the 108 children who attend the studied primary school, 28 have respiratory problems. These children with respiratory problems live in older houses, with more humidity and near busy streets or industrial areas, also smoking mothers in gestation period showed some weigh in children respiratory problems. Males are more affected by these kinds of problems. Concerning the street where the school is implanted, light-duty vehicles represented the major fraction of traffic flow. Through simulation work it was possible to conclude that the school is seriously affected by the road emissions.

1. INTRODUCTION
In the last decades, traffic, air pollution and respiratory problems, especially in children's in scholar age, have increased. Since this was noticed, several studies were carried out to assess the possible relation between these factors. It is now accepted that air pollutants can trigger allergies and respiratory problems, particularly in children already with the disease (Pekkanen J., 1997). However it still is a complex issue, since it isn't easy to confer causes to some respiratory diseases such as asthma and bronchitis, as well as due to the limited understanding on cellular and molecular levels and also because of the complex temporal and spatial pattern of human exposure to air pollution. The fact that the intervenient factors can interact between themselves, turn the relation even more complex. The purpose of the study is to investigate air pollution effects in a children population.

2. METHODOLOGY
The chosen population consisted of 6 to 10 years old children, who frequent the same primary school in Barreiro city, Portugal. A questionnaire was made to assess children's living environment and it was asked the teachers collaboration, to hand over the questionnaires to children's parents.

The school was selected since it's near a busy street, Bocage Avenue. To study road traffic pollutant emissions, a field campaign was carried out in Bocage Avenue. Vehicle counting was made according to the vehicle class - light-duty gasoline vehicles (LDGV), light-duty diesel vehicles (LDDV), heavy-duty diesel vehicles (HDDV), buses running on diesel (Bus) and motorcycles (MC) and the campaign was performed in the rush hour (8:00-9:00am; 5:00-6:00pm and 6:00-7:00pm). Counting was made in the crossroads to study these stops weight on traffic and on pollutants emission. Meteorological conditions were also registered, namely temperature, wind speed and direction.

Figure 1 shows the counting points in Bocage Avenue, as well as the school location and the air quality monitoring station (AQMS) used in this study.

Figure 1: Field campaign counting points

The results from the field campaign, as well as other variables like type of fuel, considering 30% of LDV to be diesel vehicles, vehicle velocity and street characteristics were introduced in MOBILE 6.2 to calculate CO, NO_x, PM, SO_2 and VOC's vehicle emission factors. This factor is a representative value that tries to relate vehicle emissions to the quantity of pollutant released to the atmosphere, due to this source. These emission factors, as well as road and building characteristics, were introduced in ADMS-Urban 2.0 to calculate pollutants dispersion.

Pollutants measured by the AQMS near the studied road were used to assess air quality index (AQI) near the school. This index allows classifying air quality and how can interact with human health. The index is calculated through an arithmetic average of each pollutant and is associated to a colour scale, being the worst pollutants responsible for the index attributed (IA, 2007).

3. RESULTS AND DISCUSSION

From the 108 school children whom parents complete the questionnaire, 28 present respiratory problems. From this 28, 63% are male, according with (Carlos A Camargo, 2003) and 61.5% have symptoms since earlier age. Associated to the respiratory problems, 77.8% have allergies. Lifetime prevalence of 5 respiratory problems, such as wheezing, cough, asthma, bronchiolitis and bronchitis, were also obtain through the questionnaire, being cough and wheezing the most presented one. None of the children had asthma. 63% of these children are followed by specialists in this disease.

Comparing the questionnaire answers of children with and without respiratory problems, some assumptions can be considered, like the fact that children with respiratory problems live in older houses (more than 10 years), with more humidity and near busy streets or industrial areas. Also the method used to clean the floor seems to have some influence, since when water vacuum cleaner is used children tend to haven't respiratory problems, quite the opposite of broom use. Considering gestation period, smoking mothers showed also some weigh in children respiratory problems.

However, and considering that it was a small sample, some factors known to affect respiratory system, such as smoking parents, pets and floor materials didn't increase the sample with respiratory problems.

Concerning Bocage Avenue field campaign, table 1 shows the results of vehicle counting in each point. Some counting points have more than one value, corresponding to crossroads.

Table 1: Vehicle counting.

Counting Points		LDV	Bus	HDV	MC
1	Bocage Avenue	2147	25	24	19
		2758	29	132	18
2	Bocage Avenue	2096	26	68	14
		1413	24	50	16
3	Bocage Avenue	3488	92	84	28
		1214	44	16	16

LDV are the major fraction of traffic flow in Bocage Avenue, followed by HDV, since the industrial area is near the city centre. The value of HDV on the 1st counting point is derived from the crossroad with a highway that goes in direction to the industrial area.

This counting's were used in Mobile 6.2 to calculate emission factors for CO, NO_x, PM, SO_2 and VOC's from combustion process. PM are produced also from brakes lining, tyres, car body, re-suspension of road and soil dust (Marko, 2005). The model calculates PM from exhaust pipe, brakes and tyres, emitted by LDGV, LDDV, MC, HDDV and Bus (Coelho, 2005) and also SO_2, VOC, CO and NO_x contribution from combustion process. Figure 2, 3, 4 and 5 show the results of PM per vehicle, PM considering traffic counting, pipe pollutants per vehicle and in total, respectively, considering traffic counting in Bocage Avenue.

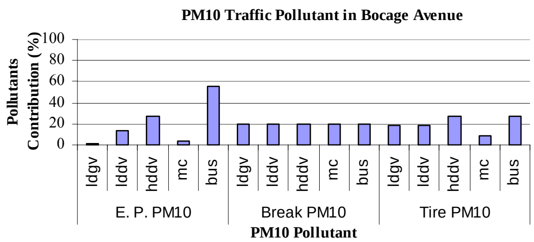

Figure 2: PM_{10} per vehicle in Bocage Avenue

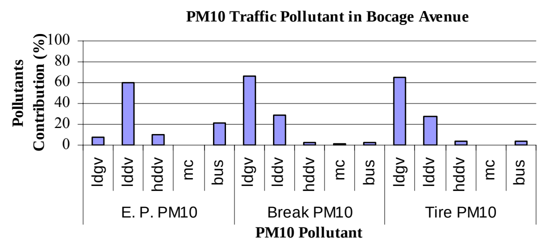

Figure 3: Total PM_{10} in Bocage Avenue

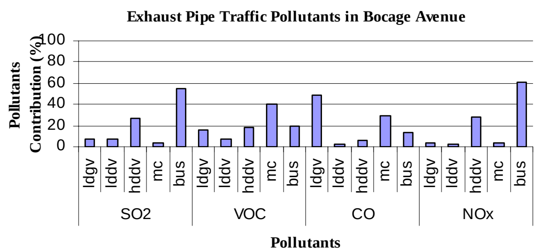

Figure 4: Exhaust pipe pollutants per vehicle

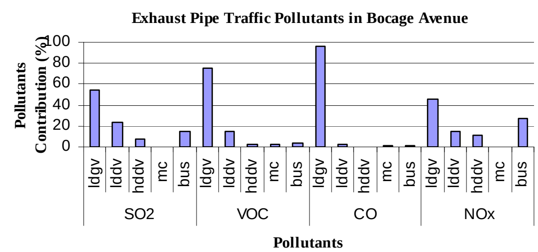

Figure 5: Total Exhaust pipe pollutants

According to the emission factors calculated by MOBILE 6.2, presented in figures 2, PM seems to be more significant for buses and HDDV than for LDV. PM from exhaust pipe are higher for diesel vehicles, particularly for buses, due to incomplete combustion and fuel composition. As a result of higher weigh, buses and HDDV release more PM from tires. Considering the total amount of vehicles, it is noticed the greater importance of LDDV regarding exhaust pipes and LDGV in relation to breaks and tires. This is mainly due to the larger number of light-duty vehicles and the importance of diesel vehicles concerning PM form exhaust pipe.

Other exhaust pipe pollutant emissions, such as SO_2 and NO_x have the same behaviour as PM considering each vehicle, but in total, LDGV have higher contributions, mostly due to the superior fuel consumption and combustion temperature of this type of vehicles. As a result of the greater number of LDGV and because diesel engine produces lower emissions of carbon monoxide and hydrocarbons (DTI, 1996), these pollutants contribution is substantially higher for LDGV.

The sum of the emission factors for all PM kind of source was used to quantify PM emitted by the vehicle:

$$PM_{10} = Exhaust\ \ pipe + Brake + Tire \tag{1}$$

where PM_{10} stands for the total emission factor (g/km), while exhaust pipe, brake and tire denote the contributions of the different source to the total emission factor. This value, as well as other pollutants calculated ones, were used in ADMS-Urban dispersion model. Figure 6 shows the result of PM dispersion in Bocage Avenue, with the most frequent meteorological conditions. In the picture is also represented the school and the AQMS locations.

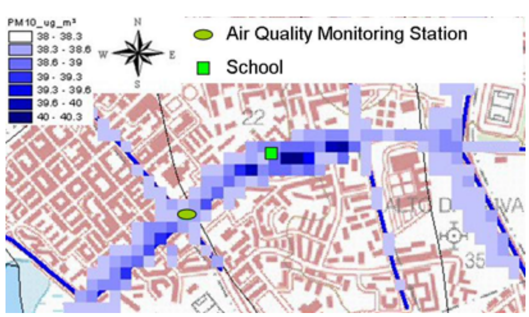

Figure 6: Field campaign counting points

The school is seriously affected by the road emissions, mainly in playground and due to the week permeability of the building, specially the glasses permeability. However according to the average measures of the AQMS, and also according to the simulation results, almost every concentration values are bellow the legal limit, corresponding to a very good AQI, except for O_3 and NO_x that correspond to a good AQI and PM with a medium AQI. Table 2 shows the simulation results of ADMS - Urban and measured by AQMS concentration values and the corresponding AQI.

Table 2: Air pollutants concentration, both predicted and measured.

	Pollutants (μg/m^3)						
	NO_x	SO_2	CO	PM	VOC	NO_2	O_3
ADMS	89	9	912	40	21	n.s.	n.s
AQMS	137	11	623	n.m.	n.m.	31	63

Very good AQI   Good AQI   Medium AQI

n.s. – not simulated; n.m. – not measured

Notice that the simulation was done only for traffic sources and in specific meteorological conditions (most frequent), wile the AQMS value is an average of 2003 to 2005 measurements and consider all type of sources.

4. CONCLUSIONS

In spite of the school location, only 28 from the 108 children who attend this establishment have respiratory problems. Interesting conclusions were made with the questionnaire, as the fact that children with respiratory problems live in older houses, with more humidity and near busy streets or industrial areas. Also smoking mothers in gestation period showed some weigh in children respiratory problems. Males are more affected by these kind of problems. However some factors known to affect respiratory system didn't increase the sample with respiratory problems.

Concerning the street where the school is implanted, as a source of traffic pollutants, the study showed that LDV represented the major fraction of traffic flow in Bocage Avenue, followed by HDV. Through simulation work it was possible to conclude that the school is seriously affected by the road emissions, in spite of low pollutant concentration values, except for PM that has a medium AQI.

5. ACKNOWLEDGEMENTS

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THE PAISARC PROJECT: ATMOSPHERIC POLLUTION, SOCIOECONOMIC DISPARITIES, ASTHMA AND MYOCARDIAL INFARCTION

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ABSTRACT

Health socioeconomic gradients are well documented in developed countries, but incompletely explained. A part of health inequalities might be explained by environmental exposures. The objective of PAISARC is to explore relationships between socioeconomic status (SES), air pollution exposure and health outcomes (asthma exacerbations and myocardial infarction onset) using a small area ecological design (IRIS). The setting is Greater Strasbourg, an eastern French urban area. We constructed a SES index from the 1999 national census data by Principal Components Analysis at the IRIS resolution. Air pollution data were modelled at the IRIS resolution on an hourly basis for the whole study period (2000-2005). Health data were obtained from different sources (local emergency networks, the local population-based coronary disease registry, medical insurance systems) according to the health outcome. Here, we present the first results concerning the construction of SES index and its relations with the distribution of health outcomes rates.

1. INTRODUCTION

In most industrialized countries, socioeconomic gradients in morbidity and mortality rates for respiratory and cardiovascular diseases are well documented (that is a lower socioeconomic status (SES) represents a higher risk factor for health outcomes). Socioeconomic inequalities in health can partly be attributed to biological and behavioural factors (smoking, alcohol consumption, physical activity…), psychosocial factors (stress, working environment), material living circumstances and access to health care. Another part of these might be explained by environmental exposures, especially exposure to urban air pollution. The objective of the PAISARC Project (Atmospheric Pollution, Socioeconomic Disparities, Asthma and Myocardial Infarction) is to explore the relationships between socioeconomic status, environmental exposure and health outcomes on an ecological basis. We study the relationships between short term exposure to atmospheric pollution and asthma exacerbations (PAISA project) on the one hand and onset of myocardial infarction (PAISIM Project) on the other hand. Such an approach was never employed in Europe for these health outcomes.

2. METHODOLOGY

Setting, Statistical Unit
The setting of our study is Greater Strasbourg, an eastern French urban area of 450,000 inhabitants distributed among 28 municipalities. The statistical unit is the IRIS, the smallest French geographic area for which socioeconomic data are available from the National Institute for Statistics and Economic Studies (INSEE). It is comparable in term of population (2,000 inhabitants on average) to census block which constitutes according to Krieger (Krieger et al., 2003) the most relevant geographic area to describe the socioeconomic inequalities in health. Greater Strasbourg is subdivided into 190 IRIS. Sixteen IRIS featuring a very small population were removed from the study (only 0.2% of the total population).

Socioeconomic Data
The demographic and socioeconomic data of the 1999 national population census have been supplied by INSEE for each IRIS. To synthesize information from these data, we decided to build a new census-based deprivation index by a Principal Components Analysis (PCA) at the IRIS resolution. For our analyses, we initially selected 52 quantitative standardized variables which described all the aspects of SES (Education, Income, Occupation …). To construct a single numerical index for all the IRIS, we decided to maximize the variance (the inertia) of the first component by deleting all the variables weakly correlated in the first component and the variables whose the contribution was lower than the average contribution. All the analyses were carried out using SAS® Software *(SAS Institute Inc., Cary, NC, USA).*
To compare our deprivation index with existing indexes, we also calculated the British Carstairs' (Carstairs & Morris, 1991) and Townsend's (Townsend, 1987) indexes which are the most popular deprivation indexes in the international literature. We estimated the convergence between our SES index and theses indexes with the Pearson's correlation coefficients (r).

The mapping of socioeconomic disparities in Greater Strasbourg using our SES index was realized with ArcGIS™ Software *(ESRI Inc., France)*.

Health Data

The PAISA Project: Asthma Exacerbations

Data on emergency visits of physicians for asthma attacks were obtained from two emergency and healthcare networks operating in Greater Strasbourg (SAMU and SOS Médecins) between January 1st, 2000 and December 31th, 2005 (n=4,729 cases, all ages). The distribution of cases by sex and by age group is presented in Table 1.

Data on respiratory drug prescriptions for 2004 were obtained from the 5 French Medical Insurance Systems (n=17,614 prescriptions for asthma exacerbations, age less than 40 to avoid confounding with prescriptions for COPD).

Table1. Distribution of emergency visits for asthma attacks by sex and age group

Age group	Males	Females	Unspecified Sex	Total
	N° cases (%)	N° cases (%)	N° cases (%)	N° cases (%)
All ages	1680 (35.5)	2369 (50.1)	680 (14.4)	4729 (100)
0-10	52 (3.1)	34 (1.4)	454 (66.8)	540 (11.4)
10-20	74 (4.4)	146 (6.2)	196 (28.8)	416 (8.8)
20-40	321 (19.1)	605 (25.5)	26 (3.8)	952 (20.1)
40-65	535 (31.8)	601 (25.4)	2 (0.3)	1138 (24.1)
65 +	673 (40.1)	963 (40.7)	1 (0.1)	1637 (34.6)
Unspecified age	25 (1.5)	20 (0.8)	1 (0.1)	46 (1.0)

The PAISIM Project: Onset Of Myocardial Infarction

Data on myocardial infarction in people aged 35 and 74 and recorded between January1st, 2000 and December 31th, 2003 were obtained from the local population-based coronary disease registry (n=1,193 cases, 35-74 age group). The distribution of cases by sex and by age group is presented in Table 2.

Data on emergency visits for acute coronary syndromes were also provided by the Strasbourg healthcare network (SAMU) for the period between January1st, 2000 and December 31th, 2005 (n=1,177 cases, all ages).

Table2. Distribution of cases of myocardial infarction in people ages between 35 and 74 years old by sex and age group

	Males	Females	Total
	N° cases (%)	N° cases (%)	N° cases (%)
Age group	912 (76.5)	281 (23.5)	1193 (100)
35-54	347 (38.0)	70 (24.9)	417 (35.0)
55-74	565 (62.0)	211 (75.1)	776 (65.0)

For each health outcome, we calculated incidence rates by age group (and by sex when it was possible) and Age-Standardized Incidence Ratios (SIR). To reduce the instability of extreme rates due to a sometimes very low number of incident cases by IRIS, we smoothed our data by an Empirical Bayesian approach using STIS™ Software *(Terrasseer Inc., USA)*.

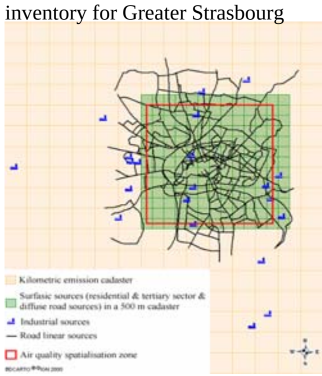
Air Pollution Data

For each IRIS, hourly ambient concentrations of five pollutants (PM_{10}, O_3, NO_2, SO_2 and CO) were modelled by the local air quality monitoring association (ASPA) for the whole period (January1st, 2000 – December31th, 2005), using a deterministic model, ADMS Urban. This model integrates emissions inventories (Figure 1), meteorological data and background pollution measurements as input parameters.

In future steps of PAISARC, an uncertainty assessment of the air pollution modelling will be carried out (INERIS), allowing to analyze the effects of modelling uncertainty on observed associations.

For each pollutant, we calculated average concentrations on the whole study period (2000-2005). The result is displayed on maps (Figures 2-6) using ArcGIS™ Software.

Figure1. ASPA's emissions inventory for Greater Strasbourg



Figures 2-6. Spatial distribution of average concentrations 2000-2005 of each pollutant across the study area: SO_2 (µg/m^3), PM_{10} (µg/m^3), NO_2 (µg/m^3), O_3 (µg/m^3) and CO (µg/m^3)

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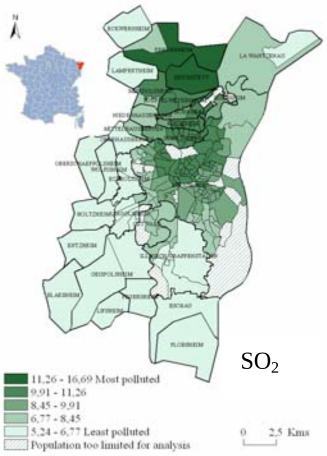

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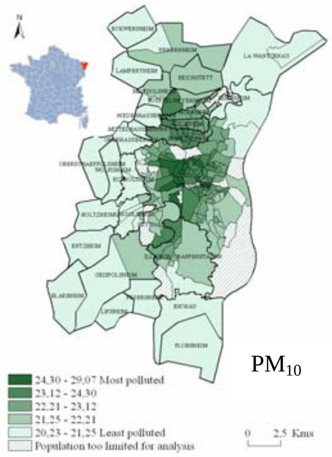

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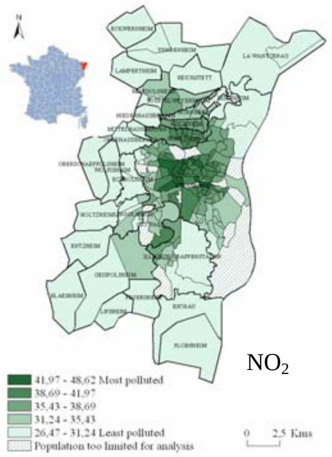

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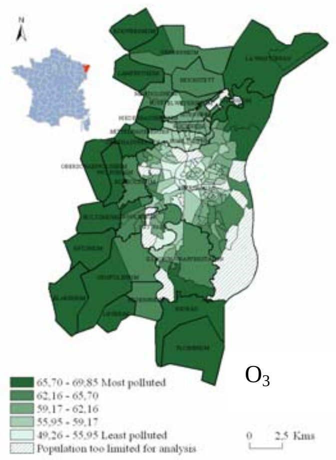

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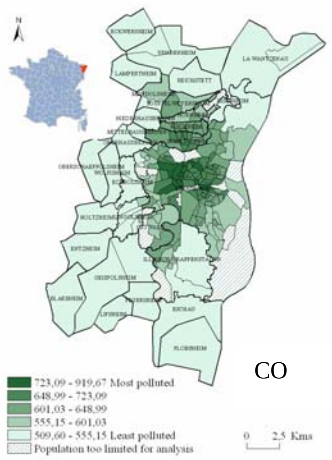


Statistical Analyses

The PAISARC Project targets 3 main and additional objectives.

1) Studying the associations between the distribution of health outcomes rates and the socioeconomic disparities in IRIS of residence (assessed by our SES Index).
2) Studying the associations between the spatial distribution of ambient pollutant concentrations across IRIS and the socioeconomic disparities (also assessed by SES Index).
3) Testing SES as a potential modifying factor in the relationship between air pollutants and health outcomes.

In this short paper, we present only the first preliminary results of analyses concerning the construction of SES index and its relations with the distribution of health outcomes rates.

For each data, the spatial autocorrelation was assessed by Moran's Index with STIS™ Software and taken into account in analyses if it was detected.

3. RESULTS AND DISCUSSION

SES Index

Nineteen variables were retained for the construction of SES index (66% of total inertia explained by the first component). They cover all the aspects of SES initially considered (unemployment, education, income, single-parent family, material wealth …). The IRIS with the lowest index's values are the most favoured ones while the IRIS with the highest index's values are the most deprivated ones (Figure 7). Mapping SES index highlights a socioeconomic gradient from the most favoured IRIS (suburbs) to the most deprivated ones (urban centre and intermediate districts). Our index presents a high internal consistency as measured by Cronbach's alpha coefficient (0.92).

It is strongly correlated with Carstairs' and Townsend's indexes (r=0.97; pvalue<0.01 and r=0.96; pvalue<0.01 respectively). A positive and significant spatial autocorrelation was detected in SES index (Moran's I = 0.54; pvalue<0.01).

Association Between SES Index And Distribution Of Health Outcomes Rates

Figures 8 and 9 present the Age-SIR of health outcomes according to SES index. A linear inverse gradient is observed between SES Index and Age-SIR of emergency calls for asthma (Figure 8); the more the IRIS are deprivated the more the Age-SIR are elevated (r=0.77; pvalue<0.01). For Age-SIR of myocardial infarction cases in men, the association is weaker (r=0.16; pvalue<0.05) (Fig. 9). Every health data source presents, after smoothing, a positive and significant spatial autocorrelation, which will be taken into account in subsequent analyses.

Figure 8. Age-SIR of emergency calls for asthma according to SES (Empirical Bayesian smoothing, all ages)

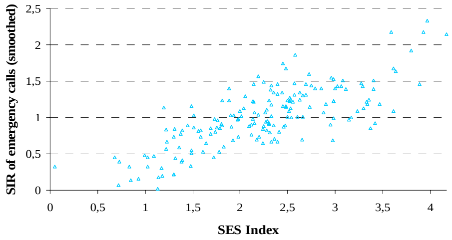


Figure 9. Age-SIR of Myocardial Infarction according to SES, registry data (Empirical Bayesian smoothing, males, 35-74 age group)

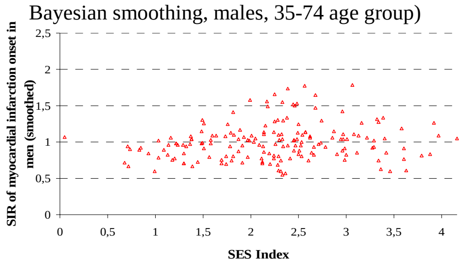
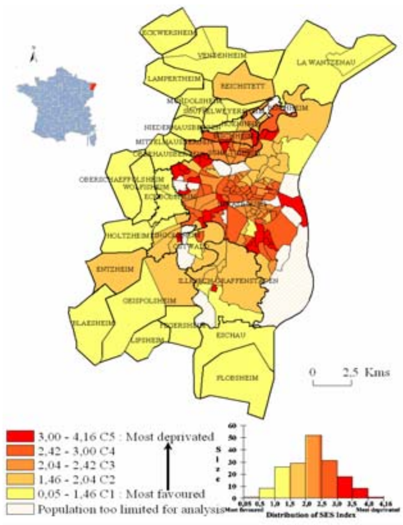


Figure 7. SES Index Distribution in Greater Strasbourg at the IRIS resolution



4. CONCLUSIONS

In the next step of PAISARC, we will study the distribution of ambient air pollutants concentrations according to SES index, taking into account spatial autocorrelation.

In the third step of PAISARC, we will use both case crossover and time series designs to test the SES as an interaction factor in the relationship between air pollution and health outcomes. These analyses will be adjusted for potential confounders (long-term trends, seasonality, meteorological factors, influenza episodes, pollen concentrations in air (for asthma only) …). Age and sex specific analyses will be carried out as far as possible. We will test different lags (0 to 5 days) between the exposure to pollutants and health outcomes.

We will also test the SES index's robustness on another area at the IRIS resolution to confirm its transposability in other French settings.

5. AKNOWLEDGEMENTS

The authors thank the French National Agency for Research (ANR) and the French Agency for Environmental & Occupational Safety (AFSSET) which fund this project.

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LINKING URBAN FIELD MEASUREMENTS OF AMBIENT AIR PARTICULATE MATTER TO THEIR CHEMICAL ANALYSIS AND EFFECTS ON HEALTH

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1. Introduction

Recent epidemiological studies have shown that atmospheric pollution caused by airborne particulate matter (PM) has a negative impact on human health. (Health Effects Institute (HEI) 2002, Pope et al 2002, and Brunekreef et al 2001) Its presence in the troposphere therefore represents an issue of major public concern and has led to increasingly detailed physico-chemical studies of the nature of PM in order to assess potential toxicological effects. (Anderson *et al* 1991, Pope *et al* 1995, Schwartz *et al* 2001). Chemical analysis shows PM to comprise of many inorganic (Trace metal/Inorganic ions), organic, and elemental materials, several of which are toxic.

Airborne PM encompasses a wide range of solid or liquid material with relatively small sizes and weights. Therefore particulates can remain in suspension for time spans greater than hours and enter human systems on a routine basis. Their chemical nature and physical properties depend mainly upon source although secondary reactions taking place in the atmosphere can also lead to their production. Many field studies have been performed worldwide to analyse inorganic/organic components of PM over a range of situations including roadsides, harbours and rural sites. However linking these analytical findings to mortality rates is not simple and requires much more detailed information about *in vivo* biochemistry than is currently available. In fact an understanding of possible relationships between the chemical content of PM, their sources and subsequent health effects has developed rapidly in recent years. For example it is clear that although damage is associated with total mass concentrations, it is also directly affected by key chemical components. It has been hypothesized that ambient PM toxicity might be associated with metal content, acidity, organic components and fine particles characteristics *i.e.* shape or size. This presentation focuses primarily on the effects of the metal components of $PM_{2.5}$ on lung toxicity. Metals which can be available in more than one stable valence state can catalyse an electron transfer and therefore demonstrate some capacity to generate oxidants. These oxidants or free radicals can induce alterations in the host respiratory system which ultimately lead to inflammation and lung injury. The current study is the first of its kind in Ireland to quantitatively analyse the chemical composition of ambient inhalable airborne particulate matter. It was motivated by the need to determine the concentrations of individual components within ambient PM over Cork City for different collected size fractions, as a prelude to the development of an appropriate source apportionment model and to cellular toxicity studies.

2. Methodology

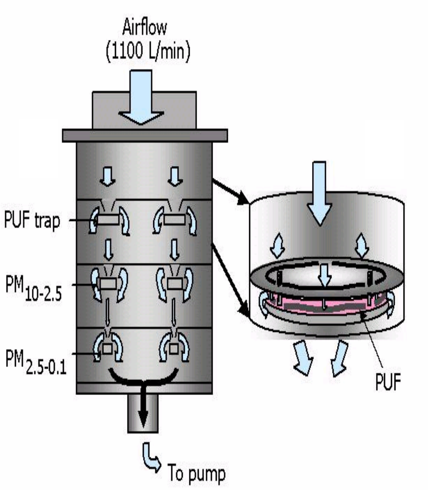

Figure 1. The three stage high volume cascade impactor, collecting $PM_{10-2.5}$ and $PM_{2.5}$ onto polyurethane foam substrate.

Samples of $PM_{10-2.5}$ and $PM_{2.5}$ have been collected using a High Volume Cascade Impactor (HVCI) shown in figure 1 at three sites throughout Cork; an urban, a background urban and rural site. The sampling campaign was scheduled to cover seasonal trends in pollutant concentrations and has since been completed giving us a sample library covering the three sites over four seasons. The collected samples have been chemically analysed for metal content using ICP-AES, organic markers have been determined using GC-MS-MS and inorganic ions have been quantified by ion chromatography. This presentation focuses only on metal content and linking this to adverse health effects.
To perform the chemical analysis and toxicology experiments, the PM was extracted from polyurethane foam collection substrate by methanol extraction technique based on a modified method of that by Salonen et al (2000).
20 metals comprising As, Sn, Hg, Mo, Zn, Sb, Pb, Cd, Ni, Fe, Co, Si, Mn, Cr, Mg, V, Ca, Cu, Ti and Al were analysed by ICP-AES after a microwave assisted acid digestion and water based extraction.
To investigate the biological effects of $PM_{2.5}$ at a sub cellular level, human epithelial pulmonary A549 cell line was exposed for three days to different concentrations of $PM_{2.5}$ (0, 5.5, 11.0, 22.0 $\mu g/cm^2$).
The intracellular reactive oxygen species (ROS) content was determined by pre-treating the cells with 2', 7'-dichlorofluorescein diacetate probe (DCFH-DA, Biosource) for 40mins. DCFH-DA penetrates the cells and is hydrolyszed to DCFH, a nonfluorescent compound that remains trapped within the cell. Oxidation of DCFH is induced by ROS in a stoichiometric reaction catalyzed in solution by peroxidases, yielding the fluorescent product DCF. The ability of $PM_{2.5}$ to produce ROS was determined according to the method of Tsubouchi et al. (2001) and the fluorescence intensity of the cells was monitored at excitation and emission wavelength of 485 and 535 nm, respectively with a fluorescent plate reader (Infinite M200, Tecan).

3. Results and Discussion

The primary objective of this work is to study the effect of metals on the production of ROS in $PM_{2.5}$ by determining the concentrations of ROS and metals. Both water soluble/transition and total metals were measured to evaluate their respective relationships with ROS after a 3 day exposure.

When total metals analysed were plotted against ROS production after a 3 day exposure for each site and season a poor agreement is seen for each site/season. This is shown in figure 2.

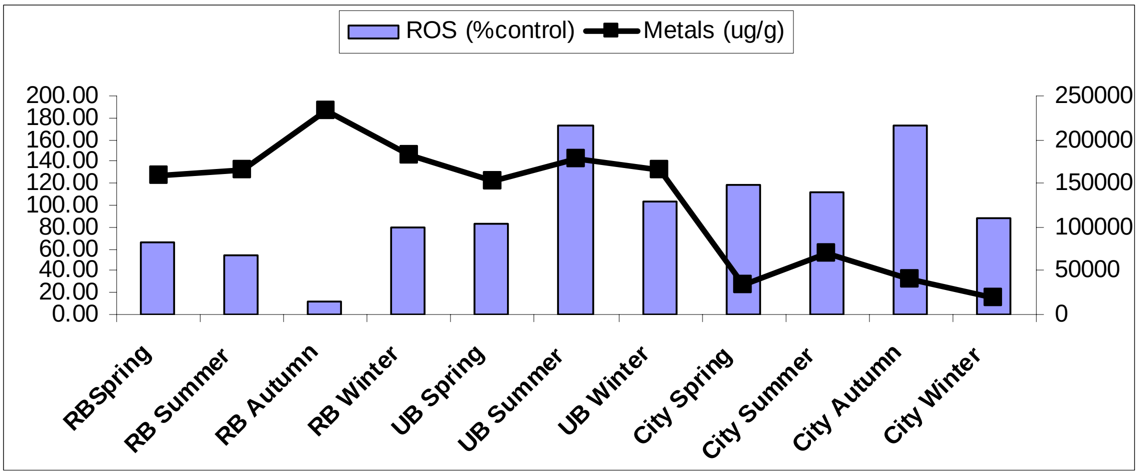

Figure 2. Plot of increase production of ROS as a percentage of control against total metals concentration shown over site/season. RB = rural background, UB = Urban background, and City =Urban

However, when transition metals concentration only are plotted against ROS production after a 3 day exposure for each site and season a very good agreement is observed shown in figure 3.

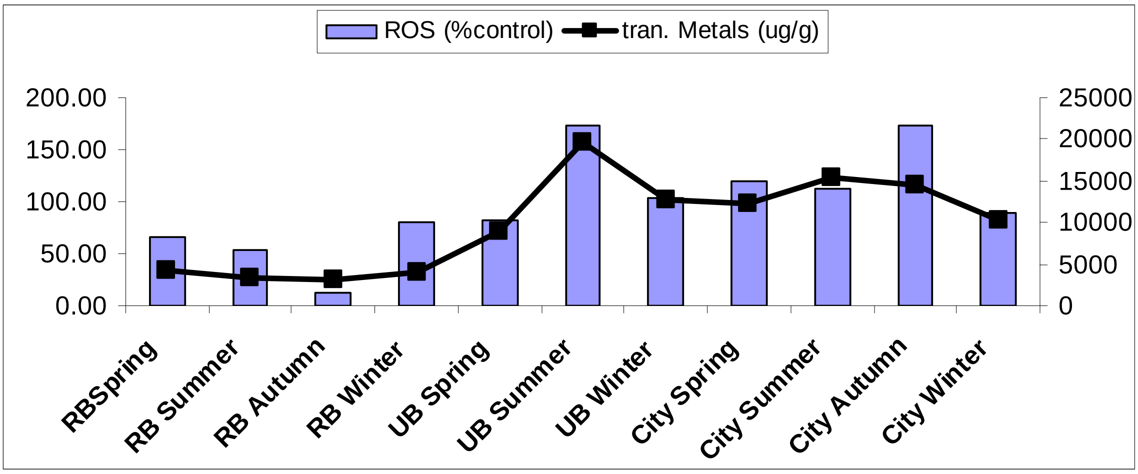

Figure 3. Plot of increase production of ROS as a percentage of control against total transition metals concentration shown over site/season. RB = rural background, UB = Urban background, and City =Urban

The correlation between ROS concentrations and both the total and water soluble metal concentrations was examined. Multivariate outliers were excluded from the linear least-squares regression analysis. At least 80% of the data collected are included in the regression analysis and ROS production as a percentage of the control were plotted against the concentration of individual metals in ng/m^3 A steep slope would mean that a marginal increase in metal concentration generates a significant increase of ROS. Hence, those metals with a large slope and r^2 value are ones which are more likely to participate in the reactions generating ROS. Linear correlation coefficients between selected transition metals and ROS production are shown in Table 1.Using this criterion, we found the total metals (acid digested) tend to have a lower ROS generating capacity than water soluble metals which tend to be more bioavailable. This is supported by *in vitro* studies that have shown that the water soluble (bioavailable) metal fraction of PM and its insoluble fraction can produce ROS independent of each other. Fenton mediated generation of hydroxyl-radicals has been shown to play a particular important role in the induction of oxidative DNA damage by the water-soluble fraction of PM as well as by transition metals.

Correlation Coefficients	Hg	Zn	Ni	Fe	Mn	Cr	V	Cu	Ti
ROS	0.7109	0.8308	0.8553	0.7602	0.7372	0.8349	0.7460	0.7984	0.5671

Table 1. Linear correlation coefficients between selected transition metals and ROS production**.**

Besides through the soluble metals, PM can generate oxidative stress directly from the surface of particles (*e.g* through activation of ROS generation in target cells, and through formation of ROS by inflammatory cells), this is in agreement with our exposure experiments where at time = 10mins an increase of ROS production as a percentage of control was observed almost immediately after the exposure of PM to the cells.

4. Conclusions

This study investigates the degree of bio reactivity of $PM_{2.5}$ on site, size, and seasonal differences. It has been shown that PM collected at three different locations throughout Cork (Ireland) and different seasons has a different degree of bio reactivity/potency to produce ROS. The chemical composition of $PM_{2.5}$ in ambient air shows strong location variability and the urban and urban background sites are influenced by higher emissions of the key chemical groups, such as metals. This fact is consistent with the higher anthropogenic emissions (industry, heating, traffic) in the urban areas compared with the rural background. In particular for the urban site where the chemical composition analysis determined a higher concentration of anthropogenic compounds during the cold seasons, $PM_{2.5}$ induced a significant increase in ROS production The results from this study, point to the conclusion that a number of water soluble transition metals maybe one group of species that can potentially lead to the generation of ROS and provides an indication of ROS–transition metal correlation over a very large number of samples. It can also be concluded that metal concentrations in ambient air over Cork city from metal concentration/inorganic point of view, to be a low to moderately polluted European city (with respect to ambient metal concentrations). In conclusion, this comprehensive study is the very first complete study of its kind encompassing all four facets of PM studies (Sampling, Analysis, Toxicology and Computer Modelling) and suggests that proactive measures should be considered to reduce particulate emissions from these transition metal emitting sources.

5. Acknowledgements

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Validation of AirGIS - a GIS-based Air Pollution and Human Exposure Modelling System

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ABSTRACT
This study describes in brief the latest extensions of the AirGIS system used in Denmark for exposure modelling and gives results of a validation with measured air pollution data. The system shows a good performance for both long term averages (annual and monthly averages) as well as short term averages (hourly and daily).

1. INTRODUCTION
Modelling of air quality is complementary to measurements since information on air quality is ideally required at many locations and in high spatial and temporal resolution. Model estimates are useful e.g. for assessment of compliance with air quality standards, for validation and optimisation of emission reduction strategies, for estimation of the personal exposure of groups of individuals in air pollution epidemiological studies or for air pollution forecast and information of the public.

The AirGIS system was developed at NERI to be able to calculate ambient air quality and human exposures at high temporal (hourly) and spatial resolution (address) to support air pollution epidemiology and urban air quality assessment and management.

AirGIS has been extended several times and in this paper we will present a detailed validation of the air quality modelling system against several years of measurements from permanent monitoring stations as well as against measurements from 204 locations from an exposure study in the Greater Copenhagen Area in Denmark that represented a wide range of traffic levels and different street geometries as well as urban and rural conditions.

2. THE AirGIS SYSTEM
For a detailed description of the system see Jensen et al. (2001) or the internet page: http://www.dmu.dk/International/Air/Models/AIRGIS/AirGIS.htm. Here only the basic features of the system are described.

The AirGIS system integrates air quality models, digital maps, national and local data registers and a GIS. One of its unique features is its ability to automatically generate the necessary input data for air pollution models (e.g. source information, geometric data) based on GIS data that would otherwise be tedious and very time consuming to generate for a large number of locations. Traffic information and buildings are available or under construction for the entire country. In principle the system is able to calculate air pollution at any address point in Denmark.

AirGIS is presently focussing on traffic-related air pollution and estimates the contribution from general urban traffic sources and the contribution from individual streets. The considered compounds are NO_x, NO_2, O_3, CO, benzene, PM_{10} and $PM_{2.5}$. Vehicle emission factors and vehicle fleet composition were updated in Feb. 2006 and used in this project. Historic emissions for NO_x and CO have been established to provide calculations back in time to about 1960.

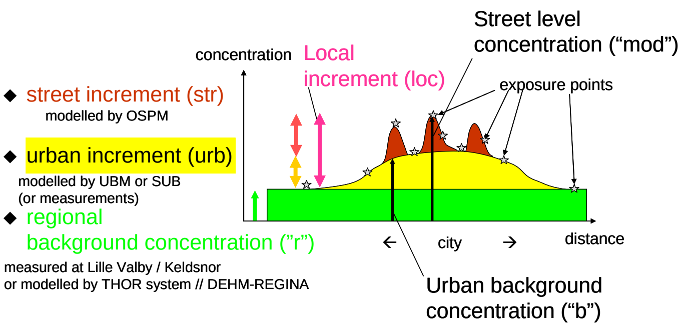

Figure 1 Illustration of the spatial variation of the three pollution levels in the vicinity of a city as considered in AirGIS and described in the text.

The AirGIS modelling system operates at three different pollution levels as illustrated in *Figure 1*. The regional background is considered as spatially homogeneous on the city scale. Depending on its location the exposure point is additionally influenced by emissions from all urban sources (urban increment) and contribution from the major street near the address (street increment). The exposure point represents the geocoded postal address location and concentration levels are modelled at a receptor point that represents a location at the facade of the building in question. The receptor height can be specified to take into account the different receptor heights of e.g. single family houses and multi-storey buildings.

- The regional background concentrations are obtained from either measurements at a regional background site or are modelled with a regional transport model (THOR system, Brandt et al. 2001).
- Urban emissions are represented in a 1x1 km^2 grid and dispersion is calculated with the Urban Background Model (UBM) (Berkowicz 2000b) or with a simplified urban background (SUB) procedure that requires much less calculation time and gives comparable results to UBM (Berkowicz et al. 2007). If available also measured data might be used as urban background. Regional background concentrations are input to modelling of the urban background concentrations.
- The local street contribution is calculated with the Operational Street Pollution Model (WinOSPM) (Berkowicz 2000a, http://ospm.dmu.dk). Urban background concentrations are input to modelling of the street concentrations.

All calculations in AirGIS are performed on an hourly basis. As far as available real measured meteorology and measured regional background data are used as input. This allows a calculation of short term averages and other statistics. Typically daily averages are used for correlation with health estimates as highest time resolution and averages over an exposure period, e.g. time a person was living at a certain address. AirGIS allows also the estimate of averages separated into working and non-working hours or other statistics, percentiles, running averages, highest daily average etc.

For historical calculations back to 1960 a standard meteorological input and synthetic background concentrations are used. For this type of estimates only long term averages (monthly, seasonal, annual) can be used for exposure estimates.

3. MEASURED DATA

Measurements at the following stations of the Danish Monitoring Network (http://www2.dmu.dk/1_Viden/2_miljoe-tilstand/3_luft/4_maalinger/default_en.asp) have been used for validation of modelled concentrations of NO_x, NO_2, CO and O_3:

- KELD – Keldsnor, South of the Island Langeland, Rural background
- HCOE – H.C. Ørsted Institute, Copenhagen, Urban background
- JGTV – Jagtvej, Copenhagen, Kerbside.

In the following figures and tables "str" denotes street, "b" urban background, "reg" regional background, "mod" model results, and "obs" measurements.

4. VALIDATION RESULTS

Annual and Monthly Means

The performance of the modelling system in reproducing long term trends at street and background level is shown in *Figure 2* for NO_2 and NO_x. When NO_2 is modelled the increasing share of direct NO_2 emissions has to be taken into account. In *Figure 2* (left) is shown that a constant value of 5% direct NO_2 emissions leads to an underestimation of the annual NO_2 concentrations at Jagtvej while an increase of direct NO_2 emissions from 5% until 1998 to 12 % in 2005 reproduces the constant trend in NO_2 concentrations much better.

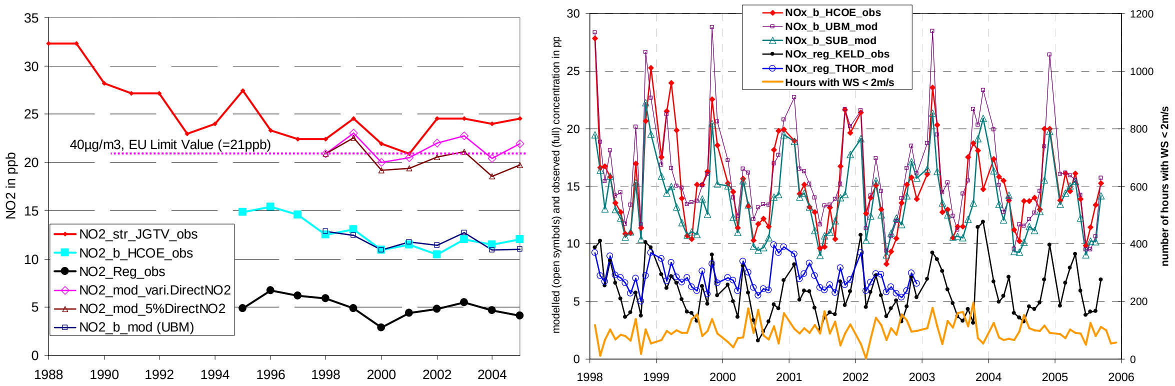

Figure 2 Annual trends for NO_2 1988-2005 (left) and monthly trends for NO_x 1998-2006.

The performance of the two background models used in AirGIS is shown in *Figure 2* (right) for NO_x using observed regional background. Both models reproduce the seasonal variation well with high correlation coefficient 0.84 and 0.79, for UBM and SUB respectively (Table 1). Also for other pollutants the background models show high correlations and reproduce both the averages and the variation in the data expressed as coefficient of variation (CoV= ratio of standard deviation and average). The present version of the THOR model performs well for O_3, while the CoV for NO_2 and NO_x is too small. Therefore we use observed regional background data as far as available and that only in case of missing data are substituted with modelled data. The variations in the urban background concentrations are influenced by the regional concentrations but also to a large extent by the meteorology. A large number of hours with wind speed below 2 m/s (plotted as well in *Figure 2*) causes typically a high urban concentration but can not explain all variation.

Table 1 shows the comparison between model and observations for different pollutants. The models show the best agreement for O_3, (R>0.9), followed by NO_x, NO_2 and CO.

Table 1 Validation for monthly means during 1998-2005, as shown e.g. in *Figure 2* (right). Av= average, CoV= coefficient of variation, Corr (R) = correlation coefficient R between model and observed values.

Location	Method	NOx			NO2			O3			CO		
		Av	CoV	Corr(R)	Av	CoV	Corr(R)	Av	CoV	Corr(R)	Av	CoV	Corr(R)
Keldsnor	r_obs	5.7	0.38		4.8	0.41		30.2	0.26				
THOR	r_mod	7.1	0.17	0.71	6.3	0.17	0.60	26.0	0.27	0.92			
HCOE	u_obs	15.1	0.26		11.8	0.20		25.1	0.27		0.28	0.24	
HCOE	u_UBM (r_obs)	16.1	0.27	0.84	11.9	0.19	0.80	23.9	0.34	0.93	0.19	0.20	0.80
HCOE	u_SUB (r_obs)	13.6	0.23	0.79	10.8	0.19	0.75	24.8	0.32	0.92	0.28	0.16	0.82

Daily means

For correlation with some health indicators (e.g. daily hospital admission) the daily concentrations of pollutants at a specific address point are of interest. The plots given in *Figure 3* give some examples of validation for NOx at street level and background level. As for the monthly averages the averages and variation are well reproduced by the modelling system. E.g. the correlation coefficient is in the range 0.8-0.9 for the street level and 0.7-0.8 for the background level.

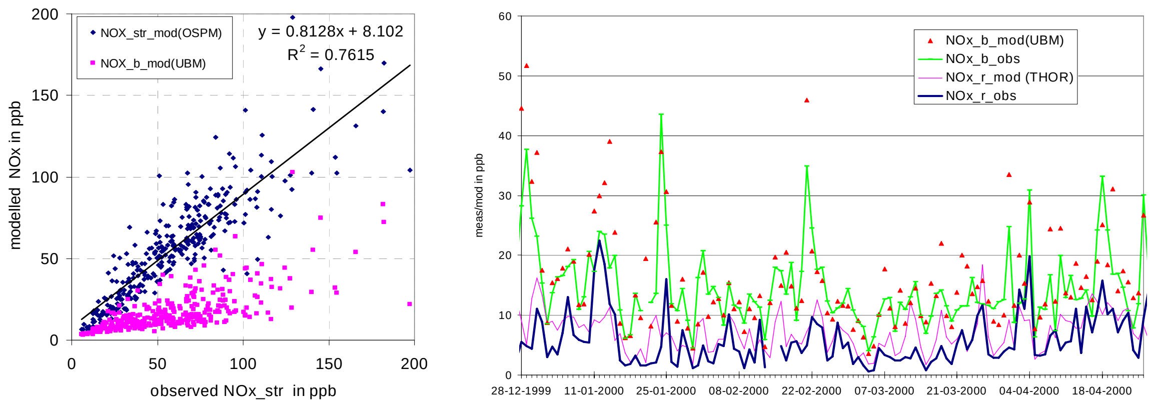

Figure 3 Validation of models for daily averages. Left: Scatter plot of daily street concentrations in 2003 modelled vs. observed for illustration also the modelled background concentration is plotted. Right: Time-series plot of modelled and measured daily NOx concentrations in urban regional background.

Diurnal variation and Hourly averages

The modelling system has been successfully validated for average diurnal variations and hourly data as well. Results will be shown in an extended version of this paper.

Comparison at many street locations

The comparisons presented so far have been performed for a few permanent monitoring stations that are well studied in beforehand. A valuable validation data set with measurements at 204 address locations was obtained in the Copenhagen Childhood Cancer project and was used before for validation of the OSPM model, where input data (traffic volume, building configuration) were obtained with questionnaires sent to

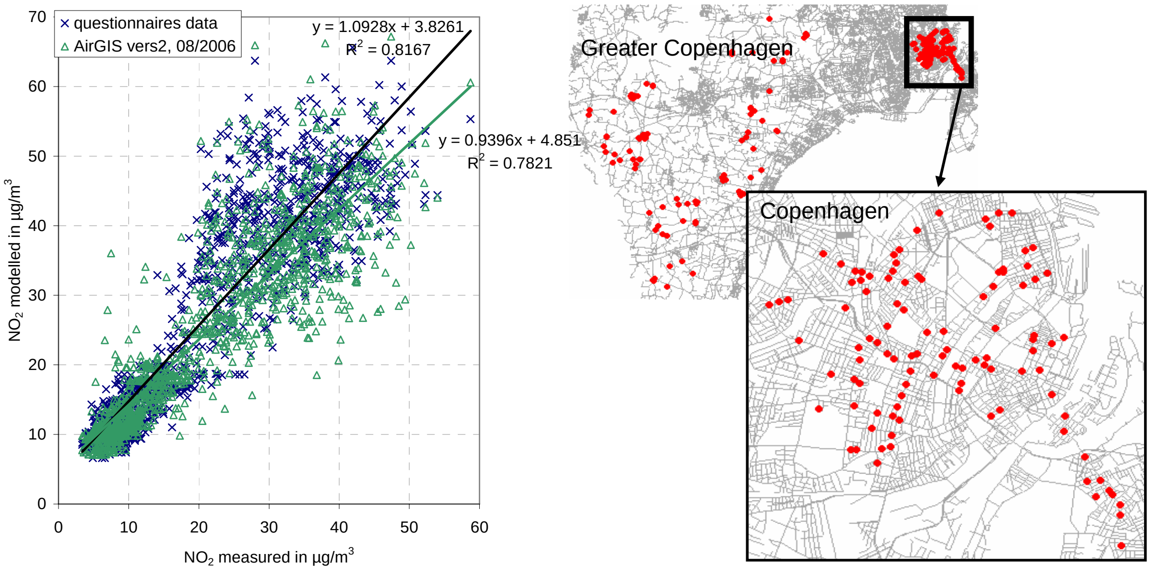

Figure 4 Left: Scatter plot for monthly averages of NO_2 modelled versus measured at the 204 addresses within the Childhood Cancer project (Raaschou-Nielsen et al. 2000). Shown are model results from 2000 based on input data from questionnaires as well as new calculations using the automated routines in AirGIS to create the input data. At each location 6 monthly periods are available. Locations were both inside Copenhagen and in surrounding rural areas (right).

municipalities (Raaschou-Nielsen et al. 2000). In *Figure 4* both the model results using input data based on the questionnaires and obtained with the AirGIS system are shown. This comparison will test the traffic data stored in the AirGIS data base and the routines to create the building configurations.

The comparison of modelled and observed data is slightly better (higher correlation coefficient) for the original data than with the automated procedure but the difference is small. The AirGIS system has the big advantage to be able to create the input data at virtually any location in much shorter time for many locations compared to the procedure by hand. A small loss in accuracy is observed. The reason for this is the higher number of outliers (points far away from the 1:1 line) in the AirGIS data. Main reasons here are differences in the traffic data bases and the differences in selecting the location of the receptor point in situations where the location is close to a street intersection. The original traffic data base was established based on questionnaire information for the specific locations obtained from the municipalities whereas the new traffic data base was established based on data from a number of different sources (data from the Danish Road directorate for main roads, traffic model data for the Greater Copenhagen Area, traffic mapping data obtained from municipalities that had prepared a traffic and environmental action plan, etc.)

CONCLUSION

The Air Pollution and Human Exposure Modelling System AirGIS has been validated against observations at permanent monitoring locations and a measuring campaign on a large number of streets. The system shows a good performance for both long term averages (annual and monthly averages) and as well as short term averages (hourly and daily) and is therefore well suited for air pollution and exposure assessment.

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MATHEMATICAL MODELING OF THE HEALTH IMPACTS INCURRED BY OPEN BURNING OF HOUSEHOLD WASTE IN RURAL SLOVAKIA

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ABSTRACT
This paper addresses the health risk incurred by two alternative waste management schemes – open burning of household waste in barrels and controlled municipal waste combustion. Using agricultural land use data and village population data we formulate three prototype villages, each representing about one third of the rural population. The two configurations of the controlled combustion are a municipal waste incinerator (MWI) and a modern waste-to-energy (WTE) plant. The CALPUFF model provides direct exposure data and the EMERAM model computes indirect exposure. Assuming 10% fraction of waste burned in the open, the cancer risk from open burning ranges from 10 to 80 times the commonly regarded de minimus value of one in a million. Cancer risks from the incinerator ranged from 7 to 371 in a million while the WTE risks were below 1 in a million. Sources of uncertainties are also briefly discussed.

1. INTRODUCTION
Uncontrolled burning is a common practice of waste disposal in rural areas of many countries. The extent to which this happens is rather uncertain as it probably varies from one country to another, depending on many factors. Several surveys and studies performed in recent years show that 16 – 48% of rural population practice open burning in the US, and 54% of all population does so in Mexico (Neurath, 2003). The European Union Dioxin Emission Inventory document (EC, 2000) assumes that only 0.25% of total domestic waste is burned illegally (means burned otherwise than in municipal combustors). At the same time these authorities admit that there are no data on the frequency with which this activity is performed. We believe that the European Union (EU) estimate understates the problem as it appears based on personal observation in rural Slovakia.
United States Environmental Protection Agency (EPA) study demonstrated that 2 to 40 households burning their trash daily in barrels can produce average polychlorinated dibenzo(p)dioxin and dibenzofuran (PCDD/F) emissions comparable to a 200 ton/day municipal waste combustor equipped with a modern cleaning system (Lemieux et al.,2000). The fact that emissions from open burning are released at the ground level, possibly resulting in decreased dispersion, makes the open burning of household waste a hot candidate for one of the top contributors to human exposure to a range of pollutants. The rural population of Slovakia is concentrated in densely populated villages made up of family houses with kitchen gardens. The principal question driving this study was: Is the open burning of waste in rural areas a serious problem from the point of view of health impacts on the population? As it is not so straightforward to decide whether the risks expressed as some absolute numbers are high or acceptable, the risks are related to those associated with actual cases of combustors in Bratislava.

2. METHODOLOGY
Formulation of Prototype Villages
The assessment of risks is based on a generalized concept of synthetic villages representing a statistical size distribution of the settlement clusters in rural Slovakia. We trisected the cumulative population distribution and established midpoint village populations, each representing one third of the total rural population of the region; hence each of the three prototype village sizes (MIN, MOD and MAX) represents about 30,000 persons in the region. Village areas are computed based on another regression approximation of village area on population, providing a linear relationship with R^2=0.845. To estimate how much waste is burned in each of the villages, the Slovak national average generation factor of 208 kilograms per person per year (SOSR, 2001) is used, averaged over 1992 to 1997. We assume that 10% of the waste generated in the villages is burned in barrels as a first approximation.

Formulation of Open Burning Emission Sources
We assume that the burning equipment and the composition of waste burned in villages is similar to the non-recycler scenario used the burn-barrel emission study performed by US EPA (Lemieux et al., 2000).
For the modeling purposes, some simplifying assumptions are necessarily adopted about the spatial and temporal configuration of sources. Based on observations, our study assumes burning of waste during daytime hours on dry days throughout the whole year as the most probable scenario. In order to save computer time, the sources are grouped as if approximately each 17 houses share one burn-barrel to which they bring their waste for burning. They are distributed uniformly throughout the villages on a 200 x 200m grid. The emissions are uniformly distributed over the 144 dry days of the year 2001 during the daytime period of 9 to 18 hour in winter and 8 to 20 in summer half of the year.

Receptor Configuration

There are two kinds of receptors considered in the village simulation: village receptors inside the village area where the burning takes place and agricultural receptors in the outer circle of the village where the croplands and pasture lands are located. In the air dispersion modeling, the agricultural receptor spacing is 500m (grid size) while village receptors spacing is 80m.
The parameters of the prototype villages, including emission sources and receptors are illustrated in Figure 1 and summarized in Table 1.

Figure 1. Design of synthetic villages – MAX village modeling domain with grouped sources (stars) and receptors (dots).

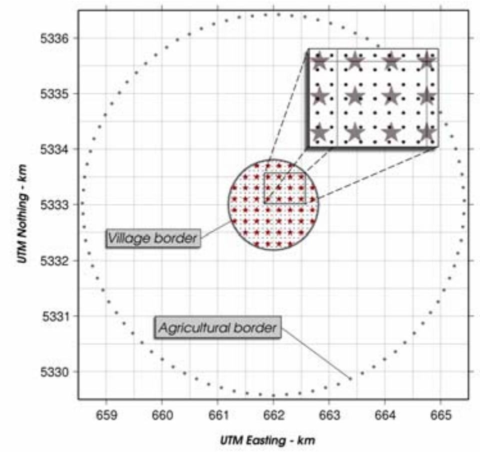


Table 1. Synthetic villages – summary of parameters

Village	Population	Area (m^2)	Radius (m)	Number of households	Number of sources	Sources spacing (m)	Total waste production ($t\ year^{-1}$)
MIN	959	582 000	419	239	13	200	199.5
MOD	1630	986 000	545	407	24	200	339.0
MAX	3601	2 170 000	809	900	52	200	749.0

Municipal Waste Combustor Hazard Identification

A public solid waste management company operates the municipal waste combustor located in Bratislava just about 2.5 km from the largest and densely populated residential city district of Petrzalka. After 23 years of operation as an incinerator, it was upgraded to a modern WTE facility in 2003. EPA's AP-42 documentation (US EPA, 1985) provides data on pollutant emissions and particle size distributions for waste combustion.
For the purpose of this study, discrete receptors were defined at the centroids of the census districts in addition to a regular rectangular mesh of grided receptors identical with the computational grid of the air dispersion model.

Exposure Scenarios and Environmental Pathways

The ultimate fate of the atmospheric emissions depends upon environmental partitioning of persistent pollutants. Quantitative descriptions necessitate the inclusion of different human exposure pathways. Table 2 summarizes the environmental pathways assigned to MWC and village burn barrels scenarios. Homegrown products are assumed to be grown in the kitchen gardens inside the villages. Commercial products are grown on the agricultural land around the villages and contribute to the transfer of risks from the open burning to urban areas of Bratislava (inter-zone risk transfers).

Table 2. Summary of exposure pathways for MWC and burn barrels.

EXPOSURE PATHWAYS[a]	POPULATION SUBGROUP (SCENARIO)			
	Urban resident & child	Suburban resident & child	Village resident & child	Fishing enthusiast
Inhalation	X	X	X	X
Soil ingestion	X	X	X	X
Maternal breast milk consumption	X	X	X	X
Homegrown fruit & vegetables	O	X	X	X
Homegown poultry and eggs	-	-	X	X
Commercial poultry and eggs	O	O	-	-
Commercial beef and dairy products	O	O	O	O
Consumption of water	-	-	-	-
Consumption of fish	-	-	-	X

[a] X – pathway from local contamination, O – inter-zone transfer or risks, - - pathway not included

Chemicals of Potential Concern (COPC)

COPCs to be studied were selected using two criteria. As our interest lies in the long term health effects, chemicals which are supposed to induce carcinogenic effects or to cause chronic functional disorders were selected. In the regulatory context this means the chemicals emitted have reference doses/concentrations and/or cancer slope factors that are referenced in scientific or regulatory literature. The second criterion was the availability of combustor emission data, and emission factors (burn barrels) for those chemicals. COPCs included in the study were thus reduced to PCDD/F, PCB, PAH and a selection of SVOCs and heavy metals; $PM_{2.5}$ was also included and treated separately, based on the analysis made by Levy and Spengler (2002).

Exposure Assessment Tools

Two computer models are used to perform the above calculations: CALPUFF (Scire et al.,2000) atmospheric dispersion model and EMERAM (Krajcovicova, 2007) – environmental media exposure and risk assessment model developed in the course of this study. Annual air concentrations and deposition fluxes enter EMERAM as inputs, together with the human exposure data. The model calculates the exposures to COPCs and associated health risks.

3. RESULTS AND DISCUSSION

We are most interested in the maximum risks and hazards in both cases (in other words, assess the impact of the burning activity on the most exposed individuals), the distribution of risks and hazards throughout the affected populations, the contributions of different pathways and chemicals to the total risks, and the degree of risk transfer from the rural agricultural areas to both populations.

Table 3 presents Incremental Lifetime Cancer Risks (ILCR) and Hazard Indices (HI) for each village and combustor scenario, and Figure 2 shows the population risk profiles.

ILCR in villages is mainly caused by polychlorinated biphenyls (PCB) through fish and breast milk ingestion pathways and acetaldehyde through plant ingestion. The remaining risks can be attributed to PCDD/F and PAHs. For the combustor cases the most important exposure pathway is plant ingestion. The largest contribution to the ILCR is associated with the PCDD/Fs and PAHs for the incinerator and PCDD/F and arsenic for WTE.

Sources of Uncertainties

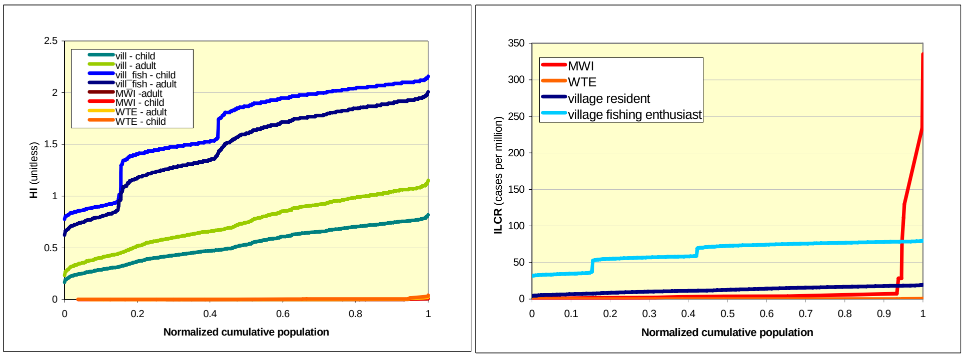
We are able to quantitatively assess the influence of uncertainties on the final risk for few of them and the direction of their influence for some. For some of them, however, we only know they exist, but, unfortunately, we do not have enough knowledge of each case to assess either their magnitude or the direction of their influence. They are discussed in detail in Krajcovicova (2007). Among those which are most influencing are the amount, composition, configuration of sources, receptors and burning conditions of the open-burned waste, which are likely to increase the resulting risks. Moreover, there are uncertainties associated with the dispersion and deposition modeling, among which the most influential is the user specified dry deposition velocity of 3 $cm.s^{-1}$, which is a highly conservative value recommended by US EPA (1998). Further uncertainties include the assessment of exposure parameters such as village farming habits, pond parameters and national statistics on the human consumption of different food items.

Table 3. Maximum and median values of ILCR and HIs for all settings and scenarios modeled in the study [c]

Settings	Scenarios	ILCR[a]		HI[b] – adult		HI[b] –child	
		max	median	max	median	max	median
OLO - MWI	Suburban resident	371	106	0.0	0.0	0.0	0.0
	urban resident	7	2	0.0	0.0	0.0	0.0
OLO – WTE	Suburban resident	0.8	0.2	0.0	0.0	0.0	0.0
	urban resident	0.0	0.0	0.0	0.0	0.0	0.0
MAX village	village resident	20	16	0.8	0.7	**1.2**	0.9
	village fishing enthusiast	**80**	**76**	**2.2**	**2.0**	**2.0**	**1.8**
MOD village	village resident	13	10	0.5	0.4	0.7	0.6
	village fishing enthusiast	**59**	**57**	**1.6**	**1.5**	**1.4**	**1.3**
MIN village	village resident	10	6	0.4	0.3	0.6	0.4
	village fishing enthusiast	**37**	**34**	**1.0**	0.9	**1.0**	0.8

[a] Incremental life-time cancer risk, [b] Hazard index, [c] Numbers in bold show the hazard indices with values equal or greater than 1

Figure 2. ILCR and HI profiles for all scenarios



4. CONCLUSIONS

The modeling results indicate that the risks rising from the open burning of waste in rural areas highly exceed those from a modern WTE facility. In absolute values, maximum ILCR from open burning is almost comparable to the obsolete incinerator. Moreover, we have to realize that we are comparing two very different phenomena. A combustor represents a highly concentrated way of waste disposal (mass of waste processed annually per square kilometer of the modeling domain is about 120 metric tons, compared with 15 tons/km2 processed in MAX village) emitting harmful chemicals from a tall stack. Consequently, high concentrations and deposition fluxes near the points of maximum impact rapidly decrease with the distance from the points. Median values and ILCR profiles tell us that all of the Bratislavsky Kraj rural population of a 100 000 is facing ILCR higher than 4 cases/million, with mean value of 15 per million, reaching in average 70 per million for fishing enthusiasts, while none of the Bratislava inhabitants faces risk higher than 1 per million due to the operation of WTE plant. Similarly, HI profiles expressing non-cancer health risks indicate that all rural fishing enthusiasts and about 20% of village children face HI higher than 1, while non-cancer hazards from both combustor cases are negligible. All the ILCR and HI values for rural population were computed assuming 10% of household waste is open-burned. Taking into account the uncertainties described in previous section, especially the one concerning the amount of waste open-burned, the open burning risks can conceivably reach values which are ten-fold times those discussed above. Another dangerous impact of open burning is supported by the premature mortality increases which are attributable to exposures to $PM_{2.5}$. These risks are considerably higher for open burning than for MWC.

These findings indicate that open burning of household waste imposes health risks to the rural population in Slovakia which are higher than those arising from the operation of a modern municipal waste combustor. It is an activity with a potential impact on human health which cannot be neglected, and is worthy of further study.

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