JOAQUIN JOINT PROJECT CONCLUSIONS

OCTOBER 2015

This report was drafted as a part of the Joaquin-project. This is an INTERREG IVB NWE project aiming to improve air quality in the Northwest European region.

Joaquin (Joint Air Quality Initiative) focusses on the air quality in Northwest Europe, the associated health effects an possibilities for improvement. The project comprises the measurement of some parameters showing a stronger correlation with health effects (ultrafine particles, particulate matter composition (metals, soot …) than the currently measured PM_{10} and $PM_{2,5}$ parameters.

The project will also evaluate measures currently available to policy makers. Certain measures will even be piloted in the participating cities. These findings will be presented to stakeholders and policy makers, whilst providing them with a tool to start working on these measures (decision supporting tool).

Finally, this project will also spread information on these novel parameters and air quality in general to both experts and the general public, that will enable them to better assess the air quality in their own region.

Duration: 01/05/2010-30/11/2015

Partners:
- Belgium (4): Vlaamse Milieumaatschappij (VMM), Intergewestelijke Cel voor het Leefmilieu (IRCEL-CELINE), Vlaams Agentschap Zorg & Gezondheid (VAZG), Stad Antwerpen
- France (2): École des Ingénieurs de la Ville de Paris (EIVP), Atmo Nord Pas de Calais
- The Netherlands(4): GGD Amsterdam, Provincie Noord-Holland, Rijksinstituut voor Volksgezondheid en Milieu (RIVM), Enery research Centre of the Netherlands (ECN)
- United Kingdom (6): University of Brighton, University of Leicester, Leicester City Council, London airTEXT, Greater London Authority (GLA), Transport for London (TfL)

More information on the project can be found on www.joaquin.eu.

1 Ultra-fine particle (UFP) Monitoring – Instrumentation, Data Collection and Interpretation

Although UFPs (particles <100 nm) contribute little to the mass of particulate matter (PM) in ambient air they are the dominant contributors to particle number and owing to their small size and ability to penetrate the respiratory system, are thought to be potentially more hazardous to human health than larger particles. These particles have not been routinely measured in air quality monitoring and there is not one reference technique which is currently used. A range of equipment was assessed for the purpose of characterising UFPs.

1.1 UFP measurements – How easy was the set up and maintenance of the UFP equipment and what data coverage was obtained?

TSI 3031 ultrafine particle monitor (UFPM) - six size channels ranging from 20-800 nm:
Easy to initially set up, is rack mountable and has relatively low maintenance requirements. Remote data access over Ethernet is easy to achieve. Problems were encountered including the software freezing requiring instrument reboot and anomalously low counts (consistent lower particle counts with a correlation in temporal behaviour, but with a constant factor ~4 offset) recorded by University of Brighton instrument.

TSI 3783 water based condensing particle counter (EPC) - measuring from 7-1000 nm
Easy to initially set up and again is rack mountable. Maintenance includes four-weekly wick changing water top ups and instrument is sensitive to deviation from horizontal when filled with water instrument to avoid flooding. Problems encountered included University of Leicester experiencing problems with decreasing pulse height over the year for reasons unknown, rectified by yearly servicing.

TSI instruments require connection to the sample conditioning system (TSI ESS) requiring four-weekly maintenance.

Grimm Scanning Mobility Particle Sizer (SMPS)+C 5420 with L-DMA 45 size classes 10-1100 nm
Relatively easy to initially set up and again is rack mountable. It did, however, require somewhat more knowledge and assistance from the manufacture than typical for an air quality monitoring instrument. Maintenance includes four-weekly flow check and butanol handling (draining the waste liquid and adding clean liquid). The radioactive source of the SMPS can cause regulatory issues, although it can even be used in mobile trailers. Transportation and permit costs of the radioactive source need to be taken into account.

TSI 3550 NSAM – particle surface area deposition in lung region 10-1000 nm (with 1µm cyclone).
Very easy initial set up, cyclone requires weekly cleaning. Software very limited with only RS-232 data output and only exports data into readable format when sampling terminated. It is a questionable choice for use for long term particle monitoring especially where near real time data output is required.

The consumables (filters etc.) for all instruments represent a relatively high expense (~€4000/annum for the three TSI monitors) and all require return to base yearly calibration at a high cost (€2000-€4000 per instrument).

Station	Start	Stop	Observations	PM$_{10}$		PM$_{2.5}$		NO$_2$		NO		BC		TNC		PNC		Total availability
			#	NA's	%	NA's	%	NA's	%	NA's	%	NA's	%	NA's	%	NA's	%	%
Amsterdam (AD1S)	01/04/2013	31/03/2015	35040	848	98	1074	97	1698	95	1698	95	240	99	6034	83	9660	72	91
Antwerp (AP1S)	01/04/2013	31/03/2015	35040	3287	91	2560	93	2478	93	2478	93	734	98	4583	87	5765	84	91
Leicester (LE1S)	01/04/2013	31/03/2015	35040	-	-	11008	69	5014	86	5008	86	4849	86	13500	61	12731	64	75
London (LO1S)	01/04/2013	31/03/2015	35040	2600	93	11634	67	6814	81	6786	81	17678	50	24881	29	25451	27	61
Lille (LL1S)	01/04/2013	31/03/2015	35040	1076	97	2712	92	645	98	940	97	-	-	-	-	-	-	96
Wijk aan Zee (WZ1S)	01/04/2013	31/03/2015	35040	836	98	1030	97	1032	97	1032	97	-	-	-	-	-	-	97

Table 1 - Total particle number concentration (TNC) and size-specific particle number concentration (PNC). N.B. - TNC and PNC measurements started in Oct 2013 for LE1S, TNC, PNC and BC measurements started in Apr 2014 in LO1S

Instrument coverage is shown in Table 1. Coverage for the two UFP instruments at the UK sites appears disproportionately low owing to delays in the beginning data collection. If these periods of no

data are excluded then the average coverage for the six sites for TNC is 84% and 81% for PNC, compared the more usually monitored pollutants of NO_2 at 92%, PM_{10} at 94% and 90% for $PM_{2.5}$.

Overall the instruments ran well, although occasional problems were encountered during the monitoring period with some periods of extended data loss. Over the span of the project data coverage was reasonable (81-84%) although below the more commonly used NO_x and particle monitoring equipment. In conclusion whilst the instruments represent feasible additions to monitoring networks they may require more maintenance and expertise than traditional air quality monitors in order to obtain the best data coverage.

1.2.1 UFP data collected by GRIMM SMPS compared to the UFPM

Figure 1 shows the correlation between half-hourly particle number concentration for each of the channels for the UFPM and SMPS monitor, taken from the two instruments in the mobile station and collected over a one month period. The correlation between particles less than 30 nm and greater than 200 nm show relatively poor correlation (R^2 = 0.39 and 0.54, respectively) indicating a problem with one of the instruments measuring in these size ranges. Other comparison campaigns indeed showed that the UFPM is less accurate in measuring particles >200 nm, so that the UFPM class from 200 to 500/800 nm should be considered as indicative only. The other channel bins (particles range 30-200 nm) show good correlations indicating both monitors are reliable for quantification of particles in this range.
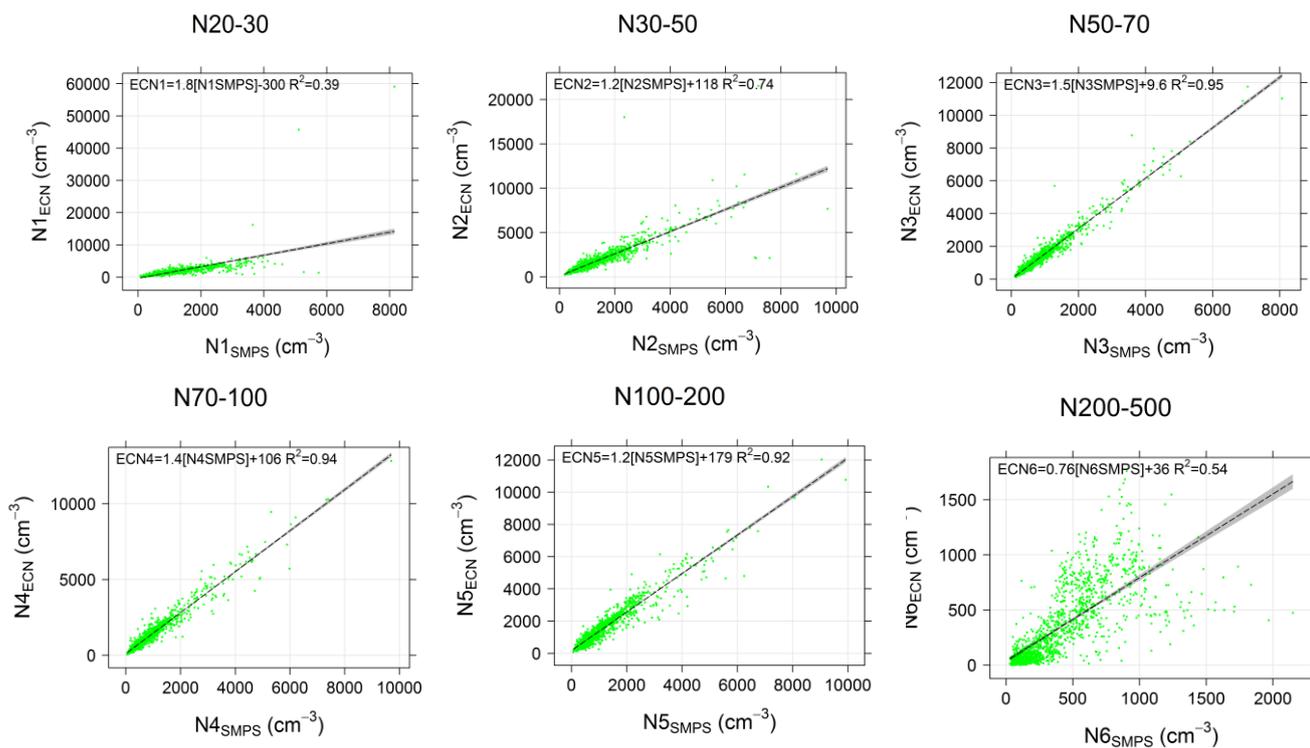

Figure 1 - Comparison between ECN UFPM and ECN SMPS size classes (one month data). N1 to N6 represent the individual particle size classes of the size-resolved instruments.

1.2.2 How does the data produced by total particle counters (e.g. EPC) compare to size-resolved measurements (SMPS and UFPM)?

As Figure 2 demonstrates the magnitude of the measured total particle number concentrations varies with the different monitors. An evaluation from the comparison between EPC and SMPS (right panel Figure 2) found the EPC has low variability and showed high correlation (R^2=0.92) with the SMPS. In addition, in comparison with total number contribution UFPM (20 ~ 500 nm) found relatively good correlation indicating that the EPC covers the particle number concentration ranges of UFPM. Overall, it can be concluded that EPC is a good monitor for measuring total particle number in urban area with low uncertainty.
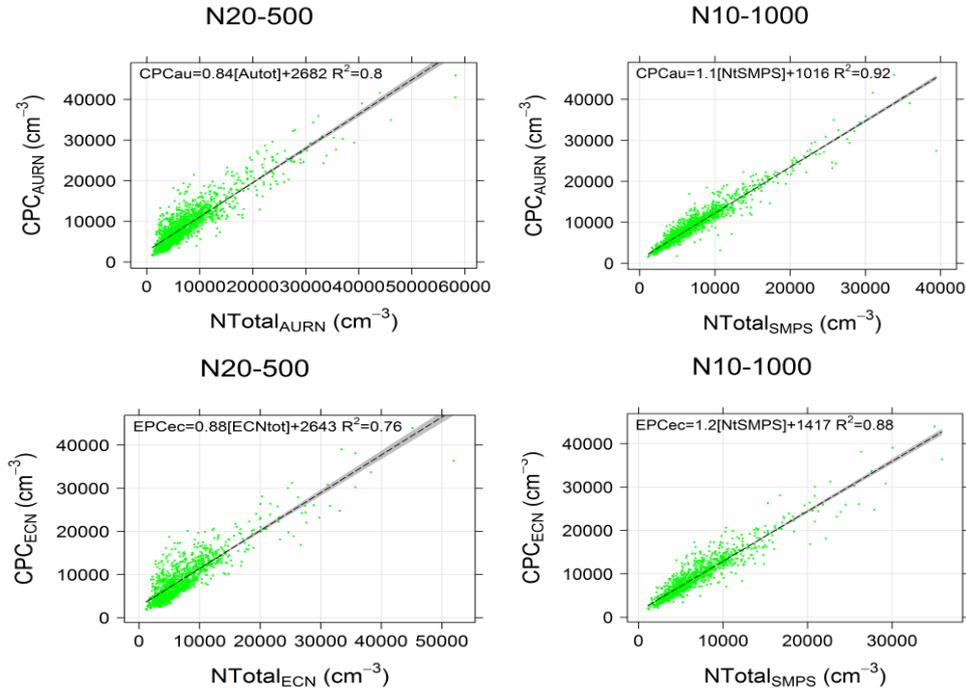
Figure 2 – comparison between EPC, UFPM and SMPS for AURN and ECN monitors (one month data).

The comparison between the size-resolved instruments (UFPM and SMPS) and total particle counter (EPC) were also performed at each site for the entire monitoring period. In order to compare the total number concentration of the EPC monitor against the size-resolved monitors (UFPM and SMPS), the measured total number concentration (TNC) was plotted against the sum of the concentrations of all size bins of respectively the SMPS in Amsterdam and Antwerp and the UFPM in Leicester and London (Figure 3). The relation between the EPC and the size-resolved instruments was evaluated by calculating the coefficients of divergence (COD) and Spearman Rank correlation coefficients (r) for all considered monitoring sites (Table 2). The correlation analysis provides information on the overall trend in association between the instruments, while the COD analysis shows differences in absolute concentrations and is defined as:

$$COD_{xy} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{C_{i\,x} - C_{i\,y}}{C_{i\,x} + C_{i\,y}}\right)^2}$$

Where x and y respresent the considered instruments, C_i is the simultaneous half-hourly UFP number concentration and n is the total number of half-hourly measurements.
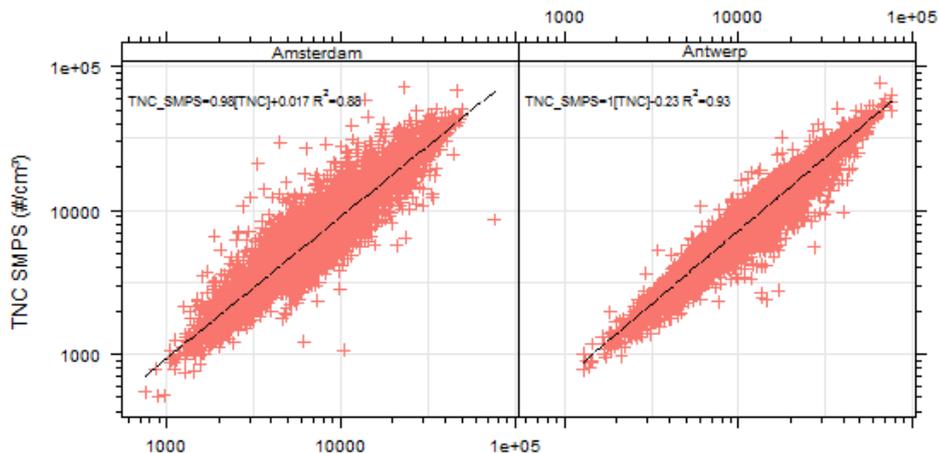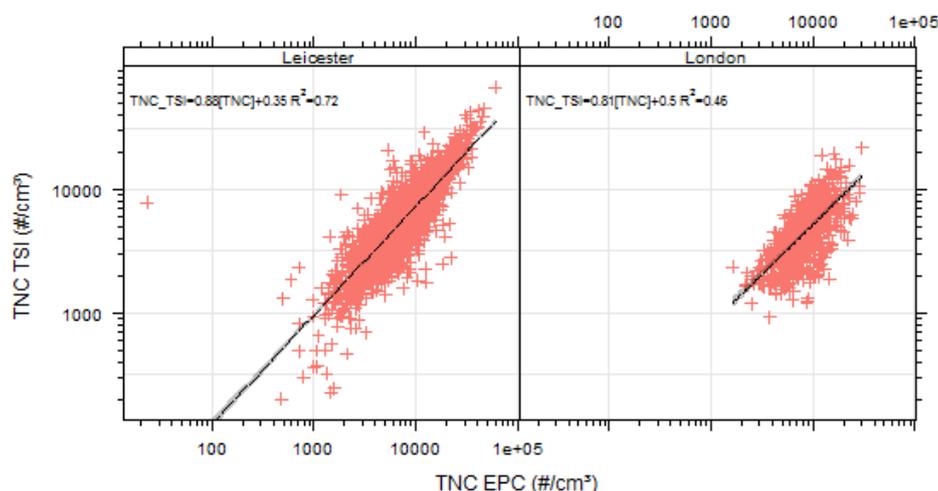
Figure 3 - Comparison of the total number concentration measured by the EPC and the total number concentrations obtained by the size-resolved UFP instruments (SMPS for Amsterdam and Antwerp (upper) and UFPM for Leicester and London (lower).

From Table 2, it becomes clear that the best associations (low COD and high r) are obtained between the SMPS and EPC instruments of Amsterdam and Antwerp. The observed COD and r differences can be explained by the sampling range of the individual instruments. While the SMPS quantifies >10 nm particles, the UFPM only samples particles larger than 20 nm, resulting in much less particle counts. As smaller-sized particles are fairly short-lived and thus determine much of the temporal variation in particle number concentration, part of the temporal variation will be underestimated when not quantified by the UFPM. This explains the weaker correlation coefficients for London and Leicester. Lowest association is obtained for London (COD=0.33, r=0.68) which might be due to the shorter monitoring period and the applied calibration factors.

	COD	Spearman Rank (r)
Amsterdam	0.10	0.93
Antwerp	0.17	0.96
London	0.33	0.68
Leicester	0.21	0.85

Table 2 - Coefficient of divergence (COD) and Spearman Rank correlation coefficients (r) between the half-hourly total number concentration quantified by the EPC and the size-resolved instruments, for all individual monitoring sites.

Looking at the regression plots in Figure 3, the total number concentration of the size-resolved instruments is always lower than the total number concentration measured by the EPC. Based on the regression coefficients forced through the origin (not shown), an 8%, 26%, 23%, and 46% lower particle number concentration is obtained with the size-resolved instruments for Amsterdam, Antwerp, Leicester and London respectively. This corresponds with previous findings during the instrument comparison in Antwerp, where total number concentration measured by the size resolved monitors UFPM and SMPS were respectively 24% and 20% lower than the total number concentration measured by the EPC. The higher EPC number concentrations could be explained by a lower minimal detectable particle size (EPC 7 nm, SMPS 10 nm, UFPM 20 nm) and possibly differential diffusion losses. The difference in minimal detectable particle size will have a significant influence in environments where a nucleation mode is frequently present.

1.2.3 How do direct surface area measurements compare to estimated surface area values?

The lung deposited surface area (LDSA) measurements were taken with the NSAM monitor; this parameter was also calculated from SMPS particle size distributions for a period of one month. Calculations were carried out assuming spherical particles and according to the ICRP model for a reference worker (ICRP, publication 66, 1994). LDSA from SMPS was calculated by converting the number size distribution into a surface area size distribution and multiplying this with the size-

dependent alveolar deposition fraction as specified by the ICRP model. Namely, the size distributions recorded by SMPS were weighted by the alveolar deposition curve and integrated over different size ranges of interest (10-100, 20-100, 20-400, 100-400, and 400-1000 nm). Results show that NSAM and 20-400 nm size bins of SMPS were in good agreement within the period, with a correlation coefficient (R^2) of 0.89 and with a slope of 1.2. Overall, it can be concluded that LDSA measuring by NSAM monitor covers particles from 20 to 400 nm size range in diameter.
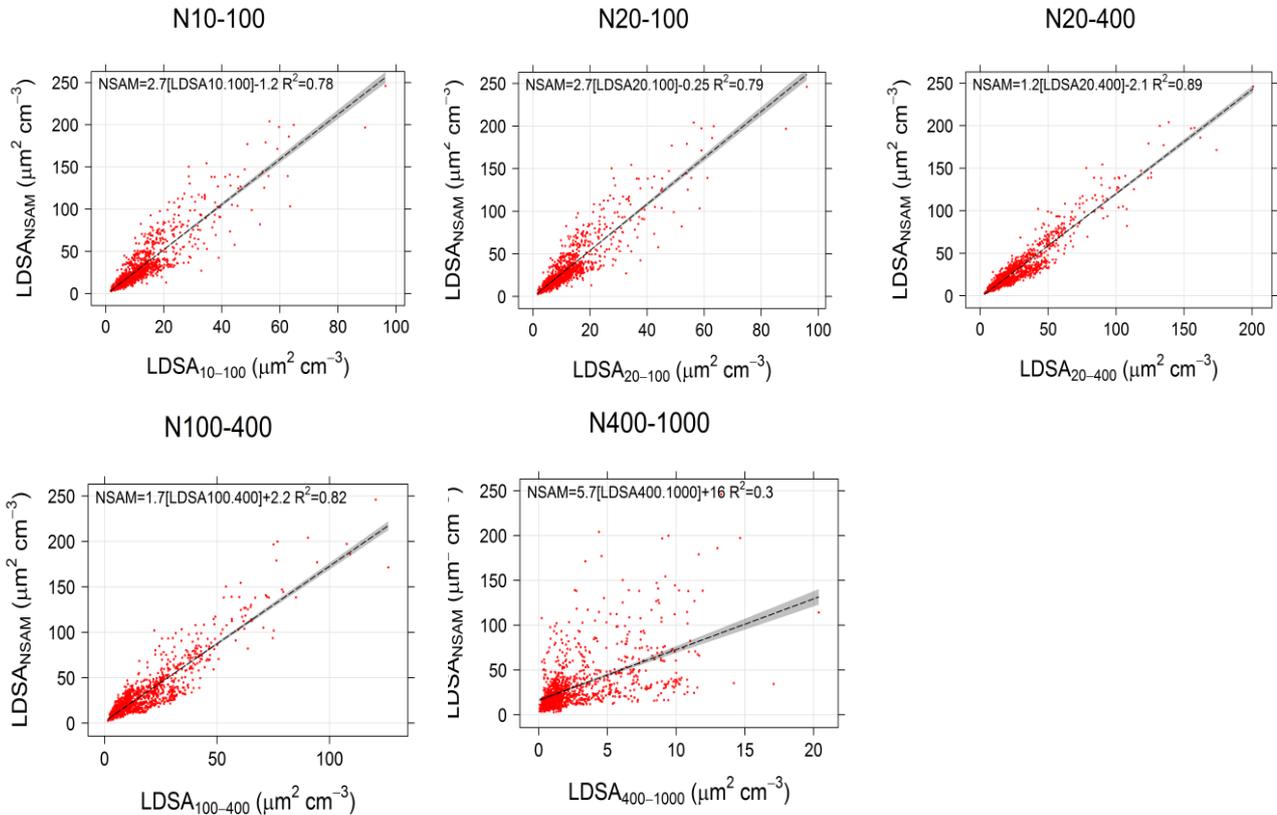

Figure 4 - LDSA comparison between NSAM and SMPS Monitors (one month data).

Overall the SMPS provides the most comprehensive data coverage over the largest range of particle sizes, however, without data treatment this amount of data can be overwhelming. This is then followed by the TSI UFPM, which appears to provide the most reliable data in the mid (30-200 nm) size range. Total particle counters, such as the TSI EPC, however, can offer a cheaper, simpler yet still reliable solution if size fractionation is not required.

1.3   What is the relationship between UFP and traffic?

1.3.1   How does particle number data compare with more commonly measured traffic-related air pollutants such as NOx and BC?

Daily-averaged NO_x concentrations (Figure 5) showed some correlation with TNC ($R^2 > 0.5$), with the highest correlation occurring in Leicester followed by Antwerp. London and Amsterdam, however, showed relatively poor correlations.
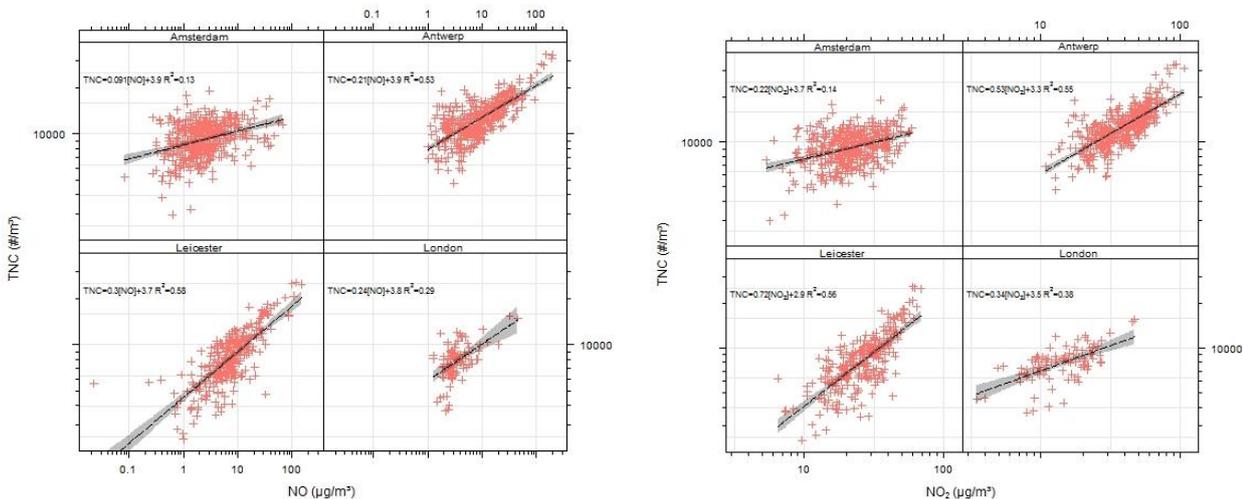

Figure 5 - Regression plots of daily-averaged total particle number concentration (# cm^{-3}) and NO and NO$_2$ (µg m^{-3}) for all considered Joaquin displayed with logarithmic scales.

Figure 6 illustrates the correlation between different size ranges of particle number concentration and nitrogen oxides (NO_x) from data collected at Leicester. Generally, it can be observed that low correlations occur in the summer season compared with other colder periods in Leicester; in winter the R^2 for all size classes exceeded 0.5.
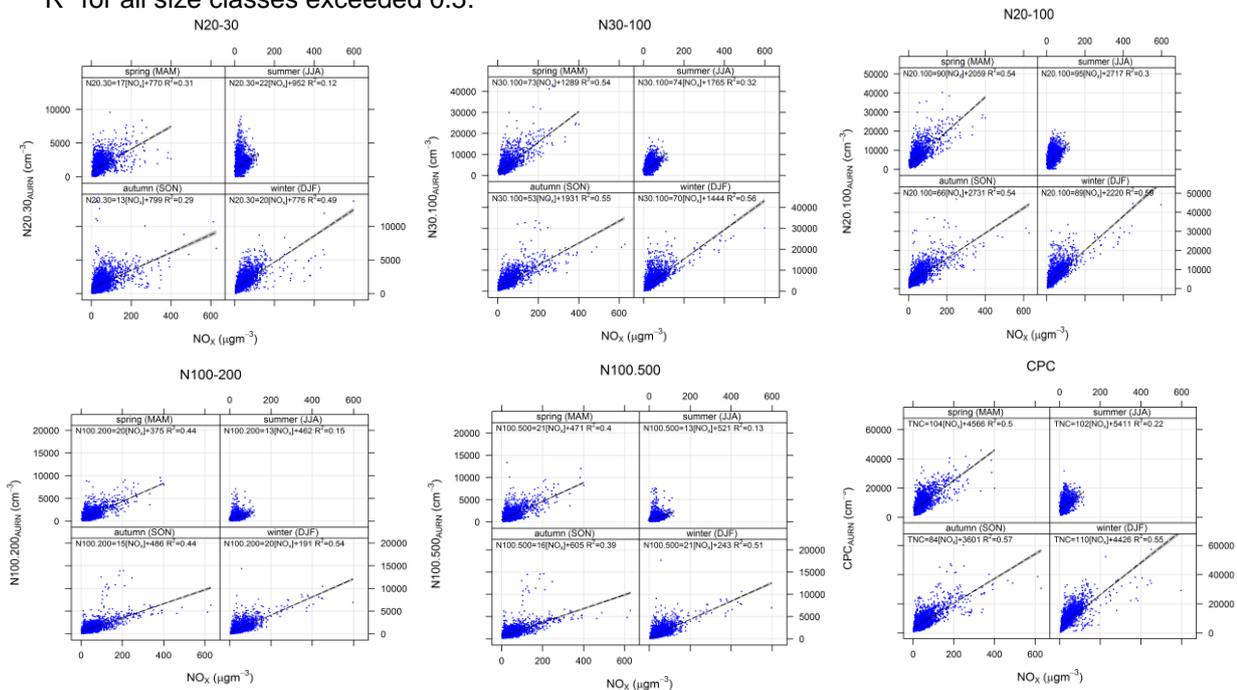

Figure 6 - Comparison between PNC measured by UFPM, and NOX at LE1S (one year data).

The strongest relations between TNC and BC at the daily interval were observed in London (R^2 = 0.50) and Antwerp (R^2 = 0.49), followed by Leicester (R^2 = 0.41). In Amsterdam, no clear relationship was observed between TNC and BC (R^2 = 0.087) (Figure 7). This is most likely due to the influence of Schiphol airport, which has proven to be an important source of 10-20 nm sized particles but emits no BC.
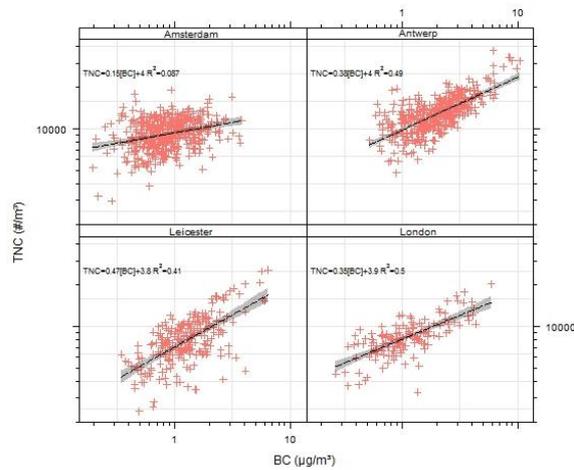

Figure 7 Correlation of daily-averaged total particle number concentration (# cm^{-3}) and BC (µg m^{-3}) for all considered Joaquin displayed with logarithmic scales.

Figure 8 shows the correlation of different size ranges of particle number concentrations with BC. It can be seen that smallest particles may originate from photochemical processes (<30 nm, R^2=0.23) and also larger particles probably (>500 nm) are very different and have low correlation; indicating that their sources are different. However, particles of size 100-500 nm show a relatively high correlation (R^2 = 0.64), and indicate that most large particles are black carbon. In addition, correlation between particles numbers with black carbon in cold seasons (R^2=0.43-0.64) are higher than warmer season (R^2=0.2-0.34), which may be related to a combination of emissions from residential wood combustion and motor vehicles.
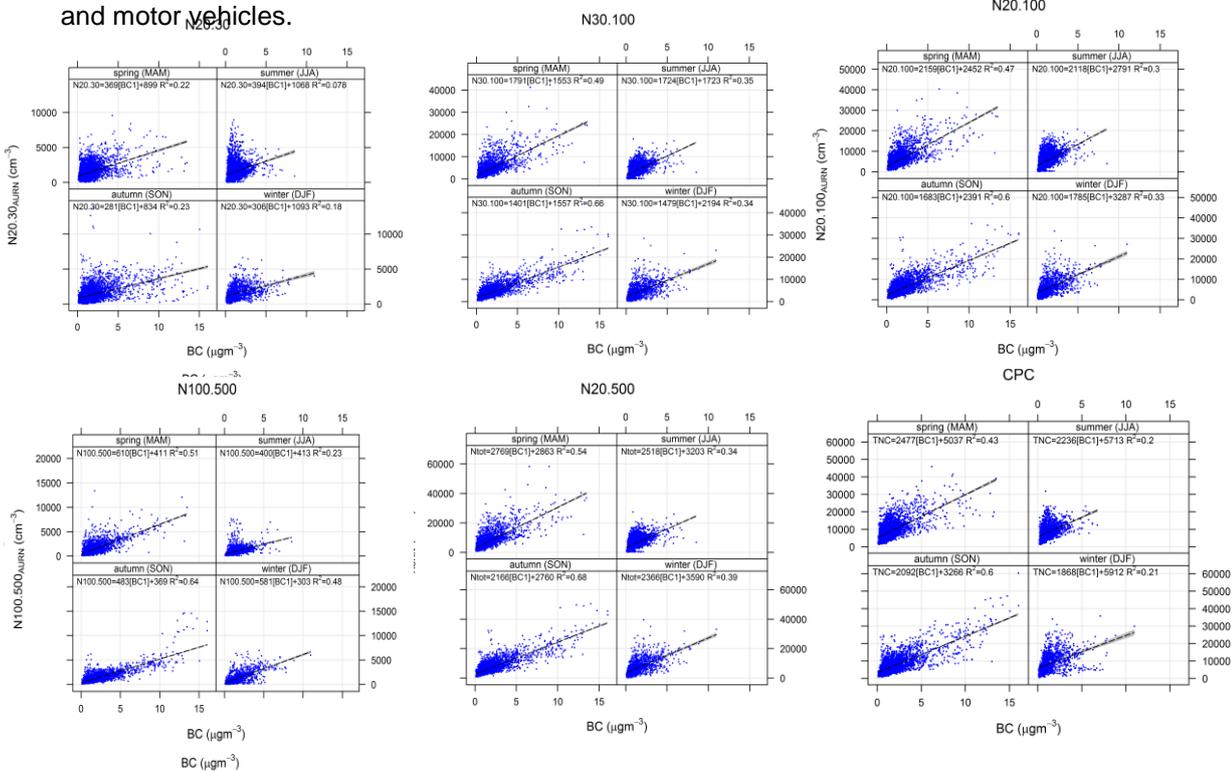
1.3.2 What is the impact of road traffic volume on particle numbers and size distribution?

Figure 9 shows the relationship of N20-30, N30-100 with traffic volume at LE1S during workdays and weekends and diurnal variations of UFP with traffic related-pollutants concentrations. High UFP concentration was observed starting at 6:00-7:00, peaking at 8:00, and then decreased gradually during the day, probably as a consequence of the increasing boundary layer height and the increase of wind speed that produces the dispersion and dilution of fine atmospheric pollutants. The increased UFP at 8:00 is consistent with NO_2, and BC, suggesting that those particles are produced from morning and evening rush-hour traffic. Particle number and other related traffic parameters levels during weekends were reduced as a consequence of the lower road traffic intensity, especially in the smaller size fraction. Overall, these results confirm that traffic emission is the major source of particle number concentration in urban environments. In Amsterdam, the lack of diurnal particle number variation in the 10-20 nm size class and weak association with traffic-related pollutants, suggests the presence of non-traffic-related UFP sources.
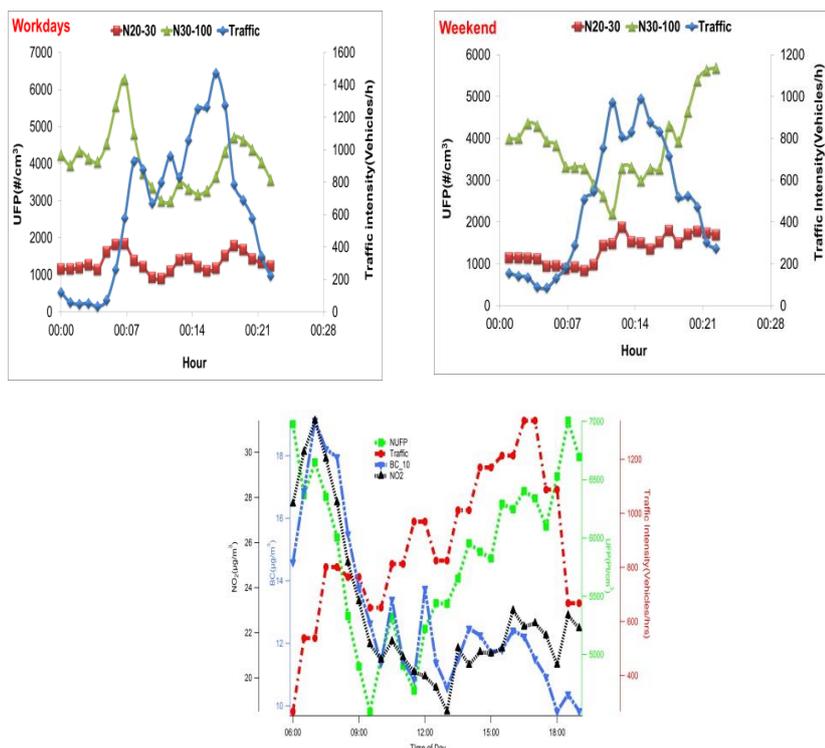

Figure 9 - Diurnal Variation of particle number and Traffic Intensity at LE1S in Leicester (one month data).

Analysis was also carried out in Antwerp at four sites across the city in order to further assess particle and traffic trends. As can be seen in Table 3 the mean TNC per site increased with mean increasing traffic intensity near the site except for at the urban background site and the street canyon site. The high number of particles at the street canyon site can be explained by the high exposure to traffic due to the street canyon effect and the proximity of the sampling location to the road.

The TNC per site and period was correlated (r = 0.22 to 0.60) with the traffic intensity at the half-hourly level. The highest correlations were found for small particles (<20 nm and 20-30 nm) (Table 4). The relationship between the particle number and traffic intensity varied clearly between the sites.

Table 3 - Mean per site of traffic intensity, total number concentration of particles < 1 μm, concentration of black carbon and nitrogen dioxide during 4 weeks in February and October 2013

Site in Antwerp	Traffic intensity (10^3 vehicles/day)	Particle number (10^3 particles/cm^3)	Black carbon[a] (μg/m^3)	Nitrogen dioxide (μg/m^3)
Suburban	3	8	1.0	31
Public park	8	11	1.6	28
Urban background	30	15	3.1	39
Street canyon[b]	14	36	-	61
Ring road	255	44	3.6	82

[a] Only measured in October 2013
[b] Only measured in February2013

Table 4 - Pearson correlation coefficient between half-hourly traffic intensity and particle number concentration in different size classes per site and measuring period

Site	Period	<20 nm	20-30 nm	30-50 nm	50-70 nm	70-100 nm	100-200 nm	>200 nm	TNC[a]	R^2_{max}[b]
Mon. station	Feb	**0.42**	0.42	0.21	0.09	0.01	-0.05	-0.07	0.22	0.18
	Oct	**0.51**	0.41	0.33	0.30	0.28	0.26	0.20	0.41	0.26
Public parc	Feb	**0.66**	0.62	0.55	0.46	0.39	0.29	-0.08	0.60	0.43
	Oct	**0.51**	0.30	0.17	0.13	0.11	0.10	0.07	0.30	0.26
Suburban	Feb	**0.44**	0.37	0.30	0.21	0.15	0.11	-0.14	0.34	0.20
	Oct	**0.40**	0.25	0.14	0.07	0.04	0.02	0.01	0.29	0.16
Urban	Feb	-	**0.33**	0.31	0.28	0.25	0.16	0.00	0.31	0.11
Ring	Feb	-	**0.43**	0.34	0.23	0.17	0.06	0.06	0.32	0.19
	Oct	0.39	**0.40**	0.42	0.46	0.47	0.47	0.39	0.44	0.16

[a] TNC is the total number concentration according to SMPS measurements
[b] The maximum correlation is given in bold and expressed as R^2_{max} in the last column

Overall, the degree of correlation with other traffic indicators and diurnal cycling show that traffic is a significant, but not exclusive, source of ultrafine particles at all the sites investigated, particularly around the <20-30 nm range.

1.4 What is the Intra-urban spatial variation of particle number data - (based on the 2 Antwerp campaigns)?

To evaluate the intra-urban variability of the monitored pollutants, UFP measurements of the fixed monitoring sites (AD1S, AP1S and LE1S) were compared to simultaneous measurements at the second trailer locations (AD2T, AP2T and LE2T) for Amsterdam, Antwerp and Leicester (Table 5). In London, no measurements at a second trailer location were performed. For more information on the setup and different locations of the trailer measurements, see Methods section.

Table 5 - Applied fixed (S) and trailer (T) measurements to evaluate the intra-urban pollutant variability.

Site	Location	Type	Monitoring period	Pollutants
AD1S	Vondelpark, Amsterdam	Fixed	14/5/2013 – 30/5/2013	BC, TNC, SMPS
AD2T	Nieuwendammerdijk, Amsterdam	Trailer	14/5/2013 – 30/5/2013	BC, TNC, UFP, SMPS
AP1S	Borgerhout, Antwerp	Fixed	7/10/2013 – 4/11/2013	BC, TNC, SMPS
AP2T	Stadspark, Borgerhout	Trailer	7/10/2013 – 4/11/2013	BC, TNC, UFP, SMPS
LE1S	Leicester University, Leicester	Fixed	5/4/2014 – 29/5/2014	BC, TNC, UFP
LE2T	Brookfield, Leicester	Trailer	5/4/2014 – 29/5/2014	BC, TNC, UFP, SMPS

The raw half-hourly UFP data of the trailer were averaged to hourly- and daily concentrations and plotted against the UFP concentrations obtained from the fixed monitoring sites in Amsterdam, Antwerp and Leicester (Figure 10).
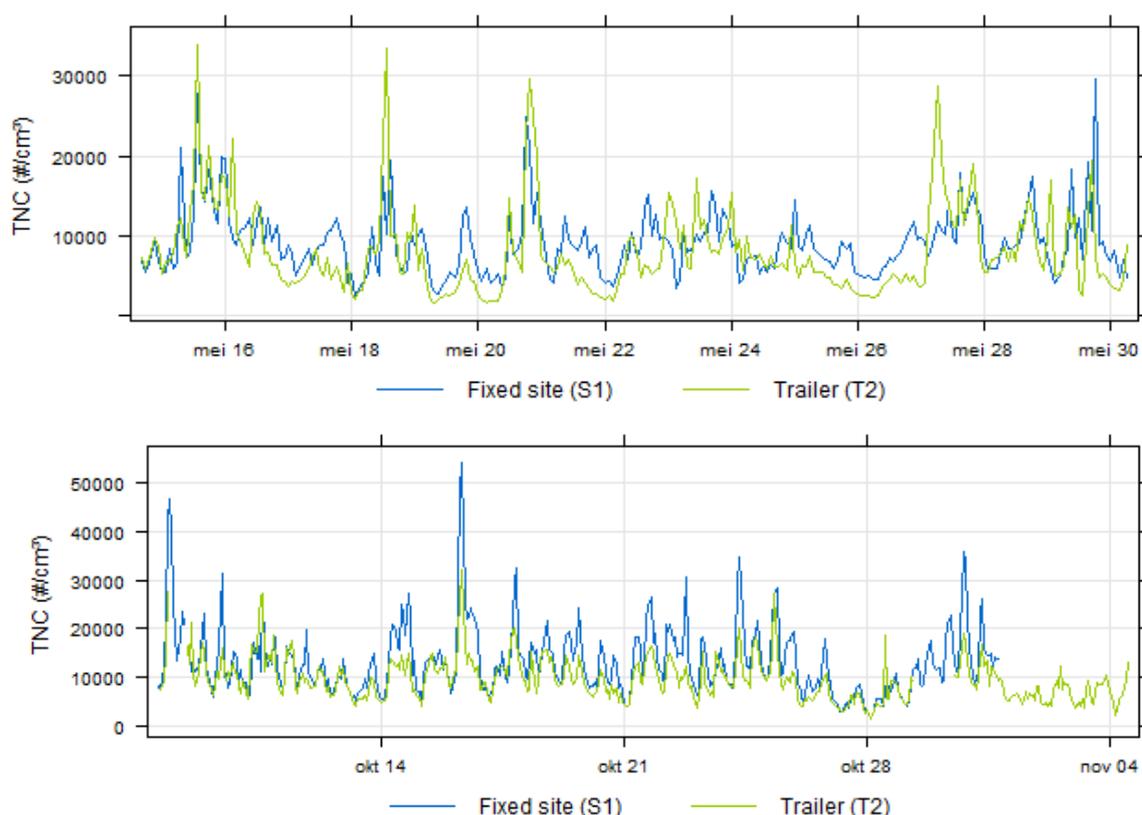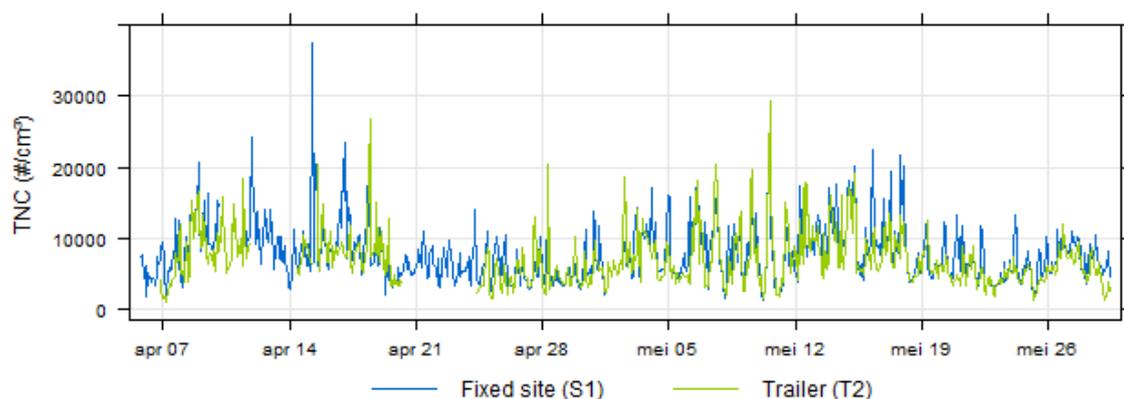
Figure 10 - Temporal variation of the total particle number (TNC) concentration at the respective fixed (S1) and trailer (T2) locations in Amsterdam (top), Antwerp (middle) and Leicester (bottom).

From the temporal variation plots, it becomes clear that the UFP concentrations at both considered locations (fixed and trailer location) covariate in time. Especially for Antwerp and Leicester, the covariance in pollutant concentrations between both locations seems very good, while for Amsterdam some deviations between both monitoring locations can be observed.

Although the intra-urban UFP concentrations covariate in time, some deviations (e.g. Amsterdam) and differences in the order of magnitude can be observed within the individual cities. For Amsterdam, both locations (fixed and trailer location) are located in relatively green areas. Nevertheless, the trailer location has no important local traffic sources and is located further from Schiphol airport. This might explain the predominant lower UFP concentration at the trailer location. The UFP concentration in Antwerp is clearly lower at the trailer location (T2) which is not surprising as the trailer was located inside an urban green area (Stadspark) while the fixed monitoring site is located near a busy access road of Antwerp. For Leicester, no clear deviations between the fixed and trailer site can be observed. Both locations are relatively quiet, and at a comparable distances from a main road, respectively Welford road (22 500 vehicles/day in 2013) and London Road (20 500 vehicles/day in 2013) as documented in the methods section of WP1.

To evaluate the intra-urban variability of total UFP concentration (TNC), coefficients of divergence (COD) and correlation coefficients (r) were calculated for each pollutant in each city (Table 6). As already suggested by the temporal variation plots, weakest association (highest COD and lowest r) is obtained for the Amsterdam sites and best correlations are obtained for the Antwerp locations. Nevertheless, the differences in UFP concentrations between both sites is largest for Antwerp, which is not surprising as the fixed site is located near a busy access road while the trailer was located within an urban green area (Stadspark).

Table 6 - Coefficient of divergence (COD) and Spearman Rank correlation coefficients (r) for UFP total number concentration (TNC) between the fixed and trailer locations within Amsterdam, Antwerp and Leicester..

	TNC	
	COD	Spearman Rank (r)
Amsterdam	0.25	0.59
Antwerp	0.16	0.85
Leicester	0.18	0.77

To evaluate potential intra-urban differences in UFP size distribution, the simultaneous average size distributions for both monitoring sites were plotted for Amsterdam, Antwerp and Leicester (Figure 11). Based on the UFP size distributions, we can conclude that large proportional differences in number concentration can be observed, depending on the considered particle size class. On average, largest intra-urban variation in total particle number concentration was observed for Antwerp (38%), followed by Amsterdam (24%) and Leicester (20%). For Amsterdam, the 10-20 nm particle number concentration was 48% lower at the trailer location (Nieuwerdammerdijk), compared to the fixed monitoring location (Vondelpark). For Antwerp, the largest difference was observed in the largest

14

particle size range (100-200 nm), with a 49% lower particle number concentration at the trailer location (Stadspark), compared to the fixed monitoring location (Borgerhout). In Leicester, the largest difference was observed in the 70-100 nm size range, with 30% lower particle number concentrations at the trailer location (Brookfield), compared to the fixed monitoring site (University AURN).
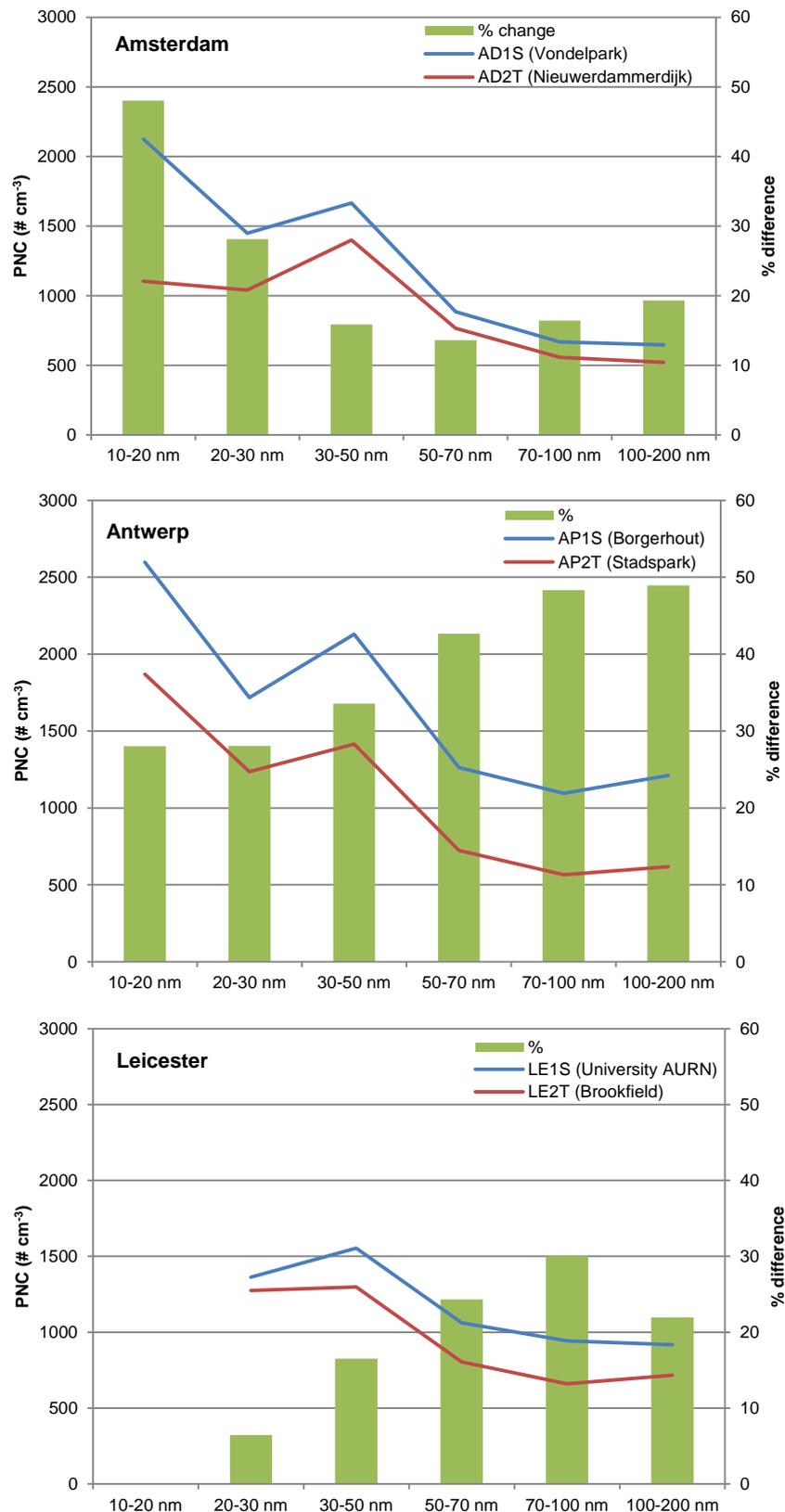

Figure 11 - Average UFP size distribution and percentage difference between the fixed (S) and trailer (T) locations in Amsterdam, Antwerp and Leicester.

Overall UFPs covariate fairly well in time at different locations within the city. Nevertheless, proportional differences in particle number concentration are obtained between the individual intra-urban sites, influenced by their proximity to urban UFP sources. This implies that the location of the UFP monitoring station is of primordial importance when evaluating the citizen's exposure to UFPs in urban environments.

2 Chemical Characterisation of PM₁₀ Filters

In order to attempt to improve air quality the sources of contributing to air pollution must be identified. Although PM₁₀ (particulate matter <10mm) is routinely measured, its composition is rarely determined. To assess PM₁₀ composition daily PM₁₀ filters were collected across the monitoring site (Wijk aan Zee – WZ1S, Amsterdam – AD1S, Antwerp – AP1S, Leicester – LE1S and Lille – LL1S) and from these every sixth filter was analysed for a range of ions, elements, elemental/organic carbon, reactive oxygen species and monosaccharide anhydride markers of wood burning. By using a mathematical model and the chemical characterization dataset source profiles were extracted that contributed to the PM₁₀ mass measured. Using known profiles from real-life sources, the calculated profiles to real sources were translated to estimate the contributions on the PM₁₀ mass in a process called Positive Matrix Factorization (PMF).

2.1 How important is chemical characterisation and what has it shown?

PMF analysis showed that identified thirteen calculated source contributions for PM₁₀: nitrate and sulphate-rich secondary aerosol, fresh and aged sea spray, steel industry, biomass burning, traffic, two sources related to metal industry, residual oil combustion, two sources related to crustal material and one unknown source related to calcium.

Profile name	WZ1S	AD1S	AP1S	LL1S	LE1S
SIA (Nitrate)	8.96	9.18	9.99	10.95	5.84
SIA (Sulphate)	3.66	2.93	2.96	2.71	2.59
Cement-like profile (Ca)	3.59	1.69	4.46	5.29	1.71
Aged sea spray	2.69	2.54	2.44	1.87	2.38
Sea spray	3.44	1.73	1.42	1.48	1.92
Crustal material with OC and K	0.99	1.48	1.74	1.36	1.88
Traffic	0.56	0.65	1.99	1.45	1.15
Biomass burning	0.73	0.74	1.24	2.20	0.70
Crustal material (primarily resuspended)	0.32	0.75	1.48	1.54	0.74
Metal industry/chemical processing (Cr)	1.05	1.06	1.12	0.30	0.34
Steel industry (Fe)	1.58	0.21	0.47	0.22	0.18
Residual oil combustion	0.99	0.46	0.38	0.42	0.19
Metal industry (As, Cd, Pb)	0.01	0.01	0.03	0.02	0.01
Average PM₁₀ contribution (sum sources)	**28.58**	**23.42**	**29.71**	**29.81**	**19.63**

Table 7 - Overview of the thirteen calculated source profiles and their average contribution in mg/m3 on the PM₁₀ mass – the names of the sources are derived from the comparison with real life sources.

It can be seen that secondary inorganic aerosol (NO_3^{2-}) is the largest contributor at all locations, whereas other more minor sources varied more between sites e.g. increased contribution of steel industry (Fe) and sea spray at the coastal industrial city of Wijk aan Zee, a marked increase in contribution by wood burning in Lille and the highest contribution of traffic at the Antwerp site owing to its location next to a major road.

Although expensive and quite time consuming, chemical characterisation gives not only valuable information on the breakdown of PM₁₀ sources but also the ability to enhance the capacity to control PM₁₀ levels and prevent breaches of legislative limits; which is of particular interest to regulatory and industry controlling authorities.

2.2 Evaluating specific risks to PM_{10} levels – what is the impact of wood burning on air quality and its contribution to PM_{10}?

The contribution of wood burning, as quantified using the wood burning marker levoglucosan, to PM_{10} is negligible in summer months but begins to rise in autumn, peaks in winter before reducing in spring (Figure 12). Baseline contributions to PM_{10} were similar across sites in summer of 1-2%. In autumn and winter larger discrepancies were seen between sites, with Lille consistently showing the highest levels of burning, averaging at around a 12% contribution in winter with several days in excess of 20%.
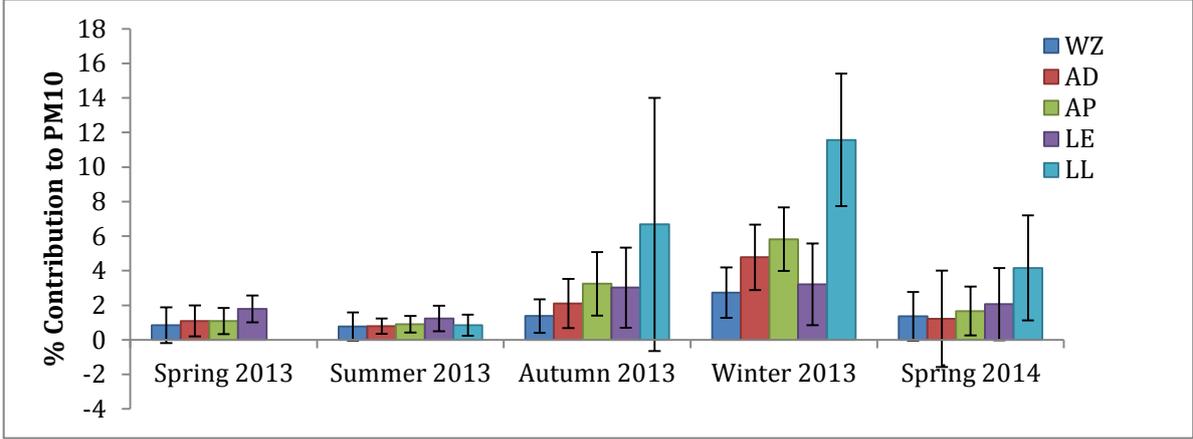

Figure 12 – Contribution of wood burning to total PM10 across five sites in NW Europe, data shows mean contribution ± standard deviation.

Wood burning is also subject to changing fashions and aesthetic influences. This is demonstrated by the fact that even in winter there is no correlation between wood burning and mean daily temperature (

Figure 13) as would be expected if wood burning was being used solely as a heating source.
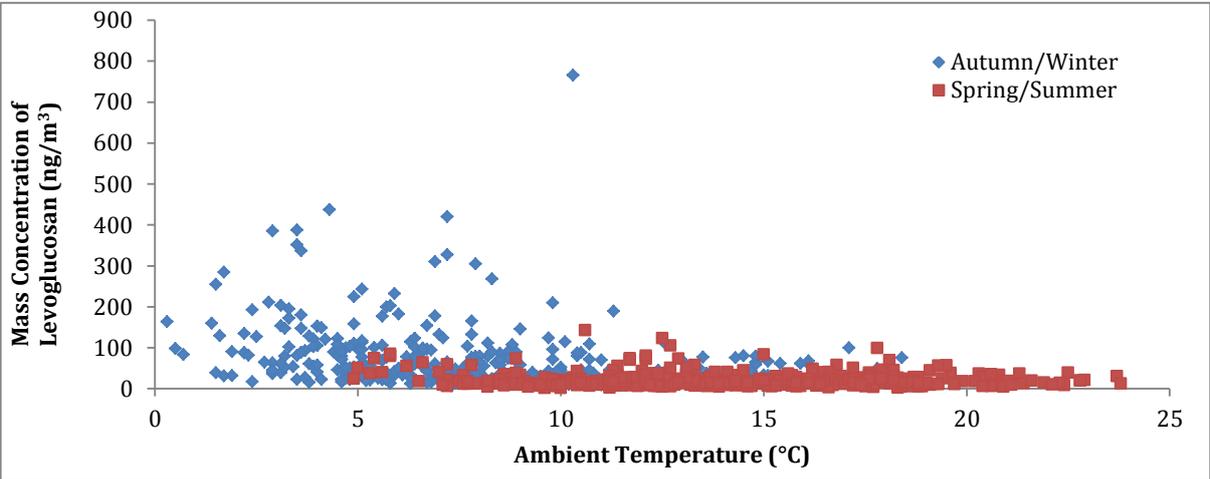

Figure 13 – Correlation of mean daily temperature levels and levoglucosan levels at Leicester over a 2 year period.

Whilst not being the highest contributor to PM_{10}, wood burning already represents enough of a contribution to PM_{10} to cause concern in some locations. Contributions are likely to increase in coming years owing to a variety of reasons including increasing costs of conventional heating fuels and the emergence of various government schemes which encourage renewable energy usage.

2.3 How does the toxicity of PM$_{10}$ as determined by biological assays (cell cultures) compare to the oxidative potential and chemical composition of PM$_{10}$ in Antwerp?

Particulate matter samples (PM$_{10}$ dust) collected in the urban area of Antwerp (Borgerhout, station AP1) were toxicologically characterised using a battery of tests and compared with results for PM$_{10}$ that was simultaneously sampled at a background location in Flanders (Houtem). Different cellular responses (cytotoxicity, pro-inflammatory changes, DNA damage) of airway epithelial cells to PM$_{10}$, the mutagenic and oxidative potential of PM$_{10}$ and the presence of endotoxins were evaluated in samples from Antwerp. PM-mediated cellular responses (cytotoxicity, pro-inflammatory changes, DNA damage) of airway epithelial cells to PM$_{10}$ and endocrine disruptive, mutagenic and oxidative potential of PM$_{10}$ were assessed in samples from Houtem.

- The studies have shown that the air quality for a given location can be characterised toxicologically.
- The PM$_{10}$ fraction induced a concentration-dependent decrease in cell viability and an increase in inflammatory cytokine induction (IL-8).
- Compared to the background location a significant increase in inflammatory mutagenic and oxidative potential of PM$_{10}$ collected in Antwerp was seen.
- Using the Androgen Responsive Element (ARE) assay, it was shown for the second time that substances with estrogenic activity are present in the Flemish ambient air. The results of the ARE test showed no agonistic androgenic effects in the PM$_{10}$ extracts, but antagonistic androgenic substances might be present in the extracts.

The biological *in vitro* measurements can help to identify which characteristics of air pollution in addition to PM mass, such as particle size, chemical composition, oxidative potential, etc. are most related to health effects due to exposure to air pollution. Toxicological characterisation of PM$_{10}$ using a battery of *in vitro* tests aims to give more insight into the unhealthy properties of the mixture of substances present in ambient air. Biological assays respond to the mixture of all substances together and directly take into account the bioavailability of the compounds.

To determine which physical and chemical determinants provide a possible explanation for the observed *in vitro* effects, the toxicological results were compared to either the physical or chemical characteristics of PM$_{10}$ or the meteorological conditions.

The results of this study agree with other studies which have shown that both PM mass as well as the chemical composition of particulate matter determine the observed *in vitro* toxicity.
- The survival of the airway cells decreased with increasing concentrations of black carbon, elemental carbon, ions (Cl$^-$, Na$^+$, NH$_4^+$ and K$^+$), the metals cadmium, lead, and antimony and the presence of products of wood combustion in the ambient air.
- The observed inflammatory response was correlated with the presence of metals (copper, manganese and zinc).
- DNA strand breaks due to oxidative damage were increased with increasing concentrations of cadmium in PM$_{10}$.
- The measured mutagenic activity (direct and indirect) of the filter extracts could be related to the total PAH content and to the sum of the carcinogenic and non-carcinogenic PAHs, the oxidative ability of PM$_{10}$, the amount of black carbon, organic and elemental carbon and the markers for wood combustion in ambient air.
- A relationship was seen between the measured estrogenic activity in the extracts and black carbon and the sum of non-carcinogenic PAHs.
- Oxidative potential of PM$_{10}$ on Teflon filters was associated with various metals in ambient air (lead, zinc, copper, arsenic, manganese, cadmium, chromium, iron, molybdenum, aluminum, barium), elemental carbon and black carbon in the ambient air.
- No chemical component was predictive for all studied health endpoints. The results of this study gave no evidence that neither the PM characteristics nor the oxidative potential were a better predictor for the harmful health effect of PM compared to particle mass concentration.

Variation of biological responses was evaluated using physico-chemical measurements as explanatory variables in multi-variate linear regression analyses.

- The harmful nature of levoglucosan on the viability of the bronchial epithelial cells was confirmed in the multiple model. Levoglucosan explained 29% of the variance in cytotoxicity of air samples.
- The presence of Cl⁻ ions in the ambient air and levoglucosan showed a negative correlation with the IL-8 induction in Beas-2B.
- The positive association between DNA breaks resulting from oxidative damage, and ambient Cd concentration was confirmed in the multiple regression model.
- A significant positive relationship was found between the direct (-S9) and indirect (+S9) mutagenic activity and ambient levoglucosan. The mutagenic activity also depended on the PM_{10} mass concentrations.
- The presence of non-carcinogenic PAHs explained 16% of the variance in the estrogenic activity of the samples.
- The presence of chromium, lead and black carbon in the ambient air explained 74% the variance in the oxidative potential of the samples.
- A significant interaction was found between PM_{10} and temperature for the effect on the mutagenicity. No significant interactions between wind speed and PM characteristics were found.

Overall, no chemical component was predictive for all studied health endpoints. The results of this study gave no evidence that either the PM characteristics or the oxidative potential were a better predictor for the harmful health effect of PM compared to particle mass concentration. The harmful nature of levoglucosan on the viability of the bronchial epithelial cells was confirmed in the multiple model. Levoglucosan explained 29% of the variance in cytotoxicity of air samples.

3 Deployment of Mobile Monitoring Station

In order to assure the quality and comparativeness of data acquired a mobile van equipped with identical instrumentation was deployed across the monitoring sites to act as a reference site. The mobile station was also deployed at an alternative site to examine the representativeness of the fixed sites. Finally it was used as an educational tool in a series of "Public Events" where member of the public had the opportunity to look at the equipment and talk to air quality experts.

3.1   How effective was the mobile campaign?

3.1.1   As a validation-harmonization tool?

The mobile station was an excellent tool for data harmonisation, and generally confirmed that the instruments were working correctly and that there was good correlation between the fixed site and mobile station instrumentation as shown by the example in

Figure 14a. Checks were made for instrumental and inlet flows, general operation of the instruments. Also sizing correctness was verified with challenging the instruments with PSL monodisperse aerosol.

However, several problems were also flagged throughout the assessment period; including anomalously low readings taken by the Eltham site UFPM (Figure 14b) which allowed the post processing correction using SMPS measurements to ensure accurate readings for the site.
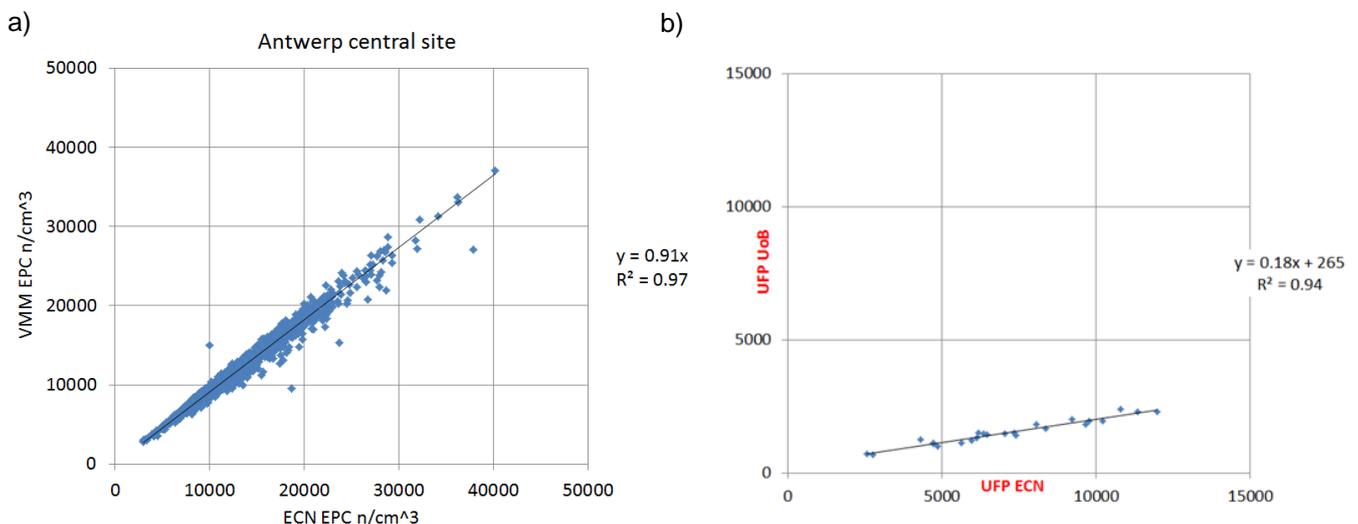
a)

$$y = 0.91x$$
$$R^2 = 0.97$$

b)

$$y = 0.18x + 265$$
$$R^2 = 0.94$$

Figure 14 – a) Correlation between Antwerp EPC and mobile station EPC, b - Correlation between Eltham UFP and mobile station UFP.

Other problems that were detected and resolved from the deployment included: the detection of interference from the slider vane pumps on particle number and BC measurements at the fixed site in Amsterdam (solved by placing filters in exhaust lines of pumps), deterioration of SMPS performance at the Antwerp site and flow anomalies detected and corrected for the MAAP and ESS inlets at Leicester.

3.1.2   As a public awareness tool?

The mobile station was transported to a variety of locations in various Member States across the project area in order to assist with raising the importance of air quality with the general public. During these public events which involved various activities and information displays, it acted as a central focal point. It had much interest from visitors and allowed members of the public to visualise what an air quality station and the equipment used to monitor the air looked like. In particular, it proved a major attraction at the 'Big Bang' science festival held in the South East of England aimed at children aged 10-17.

Overall the mobile station was an extremely valuable addition to the project providing valuable data harmonisation and validation as well as a focal point for educational purposes.

4 Air Pollution Observatory

Air quality networks produce large amounts of data which is of importance to the public, as well as experts and policy makers. In order to produce a central repository for this data, which is widely accessible and understandable, an online integrated observatory was created for visualisation of near real-time data produced by the Joaquin project and other European networks.

4.1 What was the impact of the geographic information system (GIS) on the general public/scientist/policy makers?

General Public: the GIS gives the general public the opportunity to easily access and understand the evolving state of their local air quality themselves rather than solely relying upon regulation and policy. It raises awareness regarding emerging health relevant pollutants and enables individuals to see how such pollutants evolve throughout the day. This tool will give people the information they need to reduce their exposure, allowing them to judge for themselves when best to perform certain activities outdoors, e.g. going for a walk or run. Particular sectors of the general public who will receive the largest impact from using the GIS are school children (learning about pollution) and vulnerable groups (e.g. the elderly, asthmatics, COP suffers etc).

Scientists: there is a distinct lack of long-term information available regarding the abundance and spatial distribution of health relevant pollutants, such as UFP number concentration and particle composition. The continued operation of the Joaquin network and GIS will allow the accumulation of such vital data, giving scientists the resources they require to better understand air pollution and its impacts on health and the environment.

Policy makers: when scientists have the opportunity to study emerging health relevant pollutant parameters in detail over long time scales and significant spatial distances, they will be able to better understand how to tackle the issue of poor air quality and its multitudinous array of impacts. Consequently, they will be able to present policy makers with the information they require to form the most effective policies to reduce pollution and exposure and ultimately to protect health and the environment.

4.2 Is the Sensor Observation Service (SOS) useful for this sort of project from a server and user point of view?

The flexibility of the SOS package makes it a very useful package for use as a server of data and as a tool for the end point user. Once a base framework has been constructed, the SOS can be configured to incorporate data from multiple different instruments and sites without the purchase of additional hardware; this also makes it a very cost effective option for such applications. Without a "price barrier to entry" it will be easier for the Joaquin partnership to encourage other users to join the project and provide data for the GIS. Furthermore, the adaptability and simplicity of the end user interface means that the system can be used as an effective communications tool for a wide range of uses, from the general public to scientists and policy makers, for a wide range of applications.

4.3 What impact can GIS-based urban planning have on mitigating traffic emissions related health impacts?

The method developed in Joaquin to assess the resilience of urban planning to outdoor air pollution enabled to air pollution to be studied from different point of view. Indeed, the concept of urban resilience, defined as the ability of an urban system to absorb a perturbation and maintain its functions, offers a new paradigm. The method was based on the calculation of three capacities (the capacity of urban planning to decrease emissions, to decrease concentrations and to decrease exposure). The GIS-based method has been tested in Greater Paris, with results showing how urban planning influences air quality in this agglomeration. This method could be useful to urban planners and policy makers in order to prioritize actions for adapting the city to improve outdoor air quality.

Overall, it is only the combined effects of public support and the enforcement of effective policy that can bring about an improvement in air quality; the Joaquin GIS offers an easy to use centralised data portal that can be used to promote success in both of these areas.

5 Mitigating Air Pollution Related Health Impacts

There is a need to implement new schemes in urban areas in order to help translate current air quality research in to practical measures to improve air quality. In Joaquin several measures were assessed as potential solutions to poor air quality in urban agglomerations.

5.1 How can the best mitigation strategies be communicated to decision makers?

5.1.1 What is the Joaquin Decision Support Tool?

This tool has been developed to support decision makers and their assistants, such as civil servants, in choosing the best fit measures to design urban air quality traffic policies. It provides the information on air pollution reduction measures in a web tool (www.joaquin.eu) with factsheets for every measure. There are two ways to make a selection of potential measures, either by ranking the available measures or by using categories or keywords. The tool lists the measures meeting the search-criteria according to their Joaquin score, which is a combination of the potential to improve air quality and the strength of the evidence supporting that.

The factsheets, in essence condensed reviews made by a board of international experienced experts in the JOAQUIN project-team, are designed as one page leaflets, providing a brief description of the measure, the JOAQUIN view in a few lines and the score of the measure by a green, yellow or red stamp.
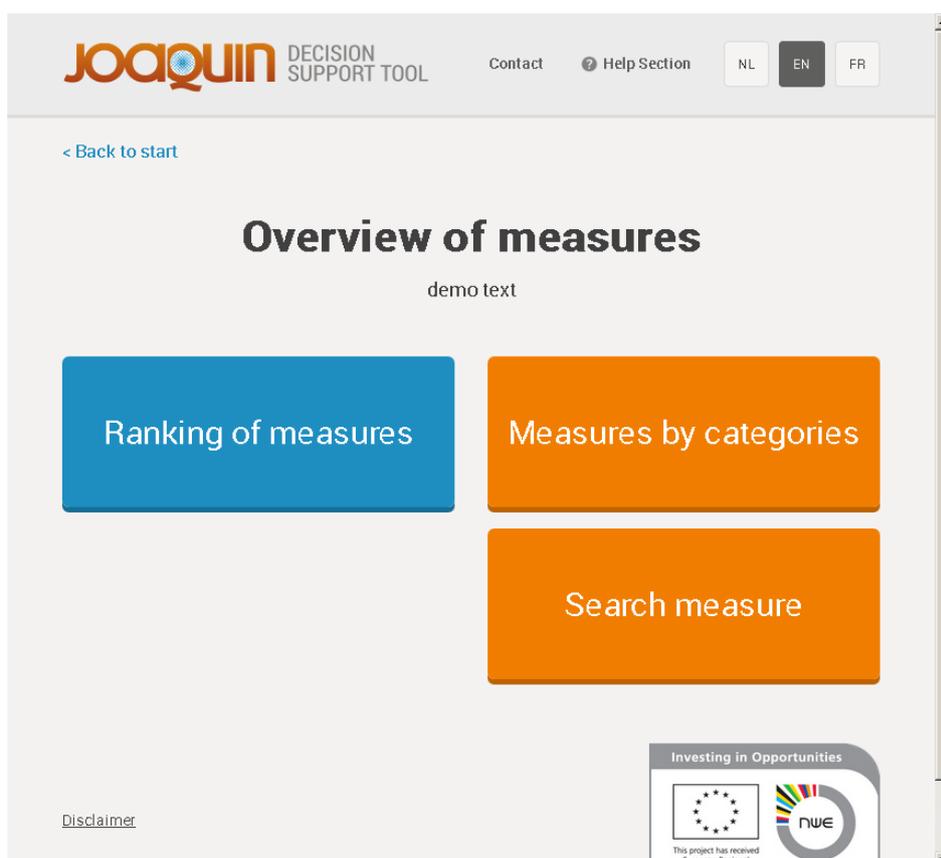
5.1.2 How is the Joaquin Decision Support tool used?

Picking the right measure does not necessarily mean it can be implemented straight away. The following list, with tips, may contribute to improved air quality policies. Needless to say it is always necessary to adapt your approach to the local situation.

Step 1: Know your local situation:
Who has valuable information?
What is the local air quality?
What is the most dominant source of air pollution in my city?

Step 2: Formulate your aim and ambition
What drives your need for improved air quality?
Legislation: Meet EU (or national) Air Quality Guidelines?
Create a healthier living environment for the public?

Step 3: Pick a (set of) measure(s)
Use the Decision Support Tool for inspiration.

Step 4: Make sure your information and ambition match
Define the air quality effectiveness of your proposed measure(s). Depending on your aim and ambition this may be a rough indication, or a component-specific and precise (model) estimation may be necessary.
Get the right specialists involved. For inspiration on organisations, have a look at the JOAQUIN partnership.

Step 5: Make sure your ambitions and proposed measures have support
Measures are often influencing habits or investments by society, businesses, and/or governments. You may therefore face opposition when you propose (a set of) measures.

5.1.3   Will the tool be useful for the user?

The scientists in Joaquin have studied emerging health relevant pollutant parameters in detail, and understand the issue of poor air quality and its various impacts. Consequently, they are able to select and summarize the information relevant for policies aiming to reduce pollution and exposure in cities, and consequently to protect health and the urban environment. The tool can furthermore be used in policy making and advising policy-makers, roles fulfilled by many of the JOAQUIN partners.

Overall, an improvement of air quality depends on effective policy with the right measures taken. The Joaquin Decision Support Tool offers relevant and accessible information on air pollution reduction measures in a user friendly web tool (www.joaquin.eu). The information in the factsheets support decision makers and others, in choosing the best fit measures that will improve the local air quality.