

**Intra-urban variability of ultrafine particles in  
Antwerp (February and October 2013)**

**Intrastedelijke variabiliteit van ultrafijne partikels in  
Antwerpen (februari en oktober 2013)**



## **DOCUMENT DESCRIPTION**

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### **Title**

Intra-urban variability of ultrafine particles in Antwerp (February and October 2013)

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### **Content**

This report describes two measurement campaigns on ultrafine particles in the city of Antwerp during February and October 2013. The variation in the number and size distribution of particles was studied at five locations with varying traffic intensity.

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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 and 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<sub>10</sub> and PM<sub>2,5</sub> 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.

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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), Eney 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](http://www.joaquin.eu).





# Samenvatting

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## Achtergrond

Dit rapport bevat resultaten over de ruimtelijke en temporele variabiliteit van het aantal deeltjes in Antwerpen op basis van twee meetcampagnes van een maand. De studie werd uitgevoerd in opdracht van de Vlaamse Milieumaatschappij (VMM) in het kader van het Joaquin-project (*Joint Air Quality Initiative*) gesteund door het INTERREG IVB NWE programma.

Verhoogde concentraties van zwevende stofdeeltjes (*particulate matter*, PM) in de lucht hebben een negatieve impact op de menselijke gezondheid. De huidige regelgeving over fijn stof is gericht op de massaconcentratie ( $\mu\text{g}/\text{m}^3$ ) van zwevende deeltjes. In de Europese wetgeving zijn grenswaarden opgenomen voor de massaconcentratie van PM<sub>10</sub> en PM<sub>2,5</sub>, dit zijn deeltjes met een aerodynamische diameter kleiner dan 10 respectievelijk 2,5  $\mu\text{m}$ . Er zijn echter aanwijzingen dat andere meeteenheden dan de bulk massaconcentratie ook belangrijk zijn vanuit gezondheidsstandpunt.

Daarom gaat steeds meer aandacht naar ultrafijn stof (*ultrafine particles*). Dit zijn stofdeeltjes met een aerodynamische diameter kleiner dan 0,1  $\mu\text{m}$  (PM<sub>0,1</sub> of UFP). Ultrafijne deeltjes dragen weinig bij tot de massaconcentratie van PM in de omgevingslucht, maar hebben wel hoge aantalconcentraties (aantal deeltjes/cm<sup>3</sup>). In een stedelijke omgeving is gemotoriseerd verkeer de voornaamste bron van primair UFP. De negatieve gezondheidseffecten van UFP zijn aangetoond in toxicologische studies, maar het epidemiologische bewijs is nog beperkt omwille van een gebrek aan meetstations voor UFP en aan studies op langere termijn.

## Doelstellingen

De voornaamste onderzoeksvragen van de studie waren:

- Hoe variëren UFP bij toenemende afstand tot een drukke weg;
- Is één meetstation representatief voor het meten van UFP in Antwerpen;
- Is er een verband tussen UFP en andere luchtkwaliteitsparameters;
- Is er een verband tussen UFP en de nabijheid en intensiteit van het verkeer?

## Methodiek

Er waren twee meetcampagnes van vier weken. De totale aantalconcentratie (*total number concentration*, TNC) en de grootteverdeling van deeltjes kleiner dan 1  $\mu\text{m}$  werden op vijf plaatsen gemeten in februari 2013 en op vier van deze plaatsen opnieuw in oktober 2013. De meetplaatsen bevonden zich in Antwerpen en werden geselecteerd om een grote variatie in de blootstelling aan verkeer te bekomen. Deze blootstelling hangt af van de verkeersintensiteit en de afstand van de meetplaats tot de weg. De metingen werden uitgevoerd in samenwerking met de Vlaamse Instelling voor Technologisch Onderzoek (VITO) en het *Institut Scientifique de Service Public* (ISSeP).

We voerden metingen uit op de volgende plaatsen (Tabel 1):

- een stedelijke achtergrondmeetplaats met een vast VMM-meetstation voor luchtkwaliteit;
- 2 plaatsen met een lagere verkeersblootstelling: voorstedelijke meetplaats en Stadspark;
- 2 plaatsen met een hogere verkeersblootstelling: *street canyon*<sup>1</sup> en ring.

De maximale afstand tussen twee meetplaatsen was 4,2 km. Tijdens de campagne in februari 2013 vond ook een gradiëntstudie plaats. Loodrecht op de weg bij het stedelijke achtergrondstation voerden we metingen uit op twee bijkomende plaatsen. Op die manier bestudeerden we de impact van de afstand van de weg op het aantal deeltjes.

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<sup>1</sup> Met een *street canyon* bedoelen we een vrij smalle straat met aaneengesloten hoge bebouwing.

*Tabel 1: Overzicht van de meetplaatsen, geordend volgens toenemende verkeersblootstelling*

Meetplaats	Dichtstbijzijnde adres	Afstand tot weg (m)
Voorstedelijk	Frederik Van Eedenplein	50
Stadspark	Rubenslei	45
Stedelijke achtergrond	Plantin en Moretuslei 285	30
<i>Street canyon</i>	Turnhoutsebaan 92	2
Ring	Noordersingel	10

Op elke meetplaats bepaalden we de deeltjesaantallen met twee toestellen:

- Een condensatiedeeltjesteller voor de totale aantalconcentratie (TNC, per minuut);
- Een *scanning mobility particle sizer* of een UFP-monitor voor de grootteverdeling van de deeltjes (per 10 minuten). De deeltjesaantallen werden gegroepeerd in de volgende grootteklassen: <20, 20-30, 30-50, 50-70, 70-100, 100-200 en >200 nm.

We corrigeerden de metingen voor diffusieverliezen en voor verschillen tussen de meettoestellen die vastgesteld waren tijdens voorafgaande vergelijkende metingen. In oktober bepaalden we ook de massaconcentratie van zwarte koolstof (*black carbon*, BC) en PM<sub>10</sub> (per halfuur). Stikstofdioxide (NO<sub>2</sub>) werd wekelijks gemeten op alle plaatsen. We bepaalden ook de verkeersintensiteit (aantal voertuigen per kwartier) op de weg dichtst bij elke meetplaats.

De weersomstandigheden verschilden vooral voor temperatuur en wind tussen de twee meetperiodes:

- De temperatuur en de regionale windsnelheid (gemeten op 30 m hoogte) waren lager in februari dan in oktober;
- De dominante regionale windrichting was noordnoordoost in februari en zuid tot zuidwest in oktober.

### Gradiëntstudie nabij een drukke weg

Met de gradiëntstudie bepaalden we in februari 2013 de aantalconcentratie en de grootteverdeling van de deeltjes op drie meetplaatsen op 10, 30 en 55 m van een drukke weg (2 x 2 rijstroken) bij de stedelijke achtergrondlocatie. De belangrijkste resultaten waren:

- De totale aantalconcentratie van deeltjes <1 µm (TNC) was hoger op 10 m van de weg dan op 30 en 55 m. Het aantal deeltjes op 55 m was lichtjes hoger dan op 30 m, wellicht door andere lokale bronnen, zodat er voor de totale periode van 4 weken geen duidelijke gradiënt was;
- Tijdens windafwaartse periodes was de TNC gemiddeld 17% lager op 30 m van de weg dan op 10 m en 22% lager op 55 m van de weg dan op 10 m;
- Tijdens windafwaartse periodes daalde het aantal deeltjes <25 nm bij een grotere afstand tot de weg. De grotere deeltjes verschilden minder tussen de drie meetplaatsen.

### Intrastedelijke variabiliteit van deeltjes

We onderzochten de variatie in de aantalconcentratie en de grootteverdeling van de deeltjes op vijf plaatsen in februari en op vier plaatsen in oktober 2013. We bestudeerden de gemiddelde waarden en dagverlopen per plaats en de temporele relatie en ruimtelijke divergentie tussen de meetplaatsen.

#### Gemiddelde waarden

- De verkeersintensiteit op de wegen nabij de meetplaatsen varieerde sterk (Tabel 2). Dit bevestigde dat we plaatsen selecteerden met een verschillende blootstelling aan verkeer;
- De gemiddelde TNC per meetplaats varieerde sterk (Tabel 2), met een maximum tot minimum verhouding van 4 tot 5 op weekdays en van 7 in de weekends;
- De gemiddelde TNC per meetplaats nam toe bij een hogere gemiddelde verkeersintensiteit nabij de meetplaats (Tabel 2), behalve voor de stedelijke achtergrondmeetplaats en de meetplaats in de *street canyon*. Het hoge aantal deeltjes op de *street canyon* meetplaats kan enerzijds verklaard worden door een hoge blootstelling aan verkeer door het *street canyon* effect. In smalle straten kan de verontreiniging zich minder goed verspreiden en blijft ze langer hangen. Anderzijds lag de meetplaats vlak naast de weg (Tabel 1);

- De deeltjesgrootteverdeling had algemeen een gelijkaardig verloop op alle meetplaatsen en tijdens de 2 periodes. De hoogste aantalconcentraties vonden we op weekdagen voor deeltjes <20 nm (gemiddeld 26% van de TNC). In de weekends droegen de deeltjesklassen <20 nm en 30-50 nm in gelijke mate bij tot de TNC (telkens zo'n 20%). Ultrafijne deeltjes domineerden de TNC, met een gemiddeld aandeel van 87% op weekdagen en 81% in de weekends.

Tabel 2: Gemiddelden per meetplaats van de verkeersintensiteit, aantalconcentratie van deeltjes <1 µm, concentratie van zwarte koolstof en stikstofdioxide tijdens 4 weken in februari en oktober 2013

Meetplaats	Verkeersintensiteit (10 <sup>3</sup> voertuigen/dag)	Deeltjesaantal (TNC; 10 <sup>3</sup> /cm <sup>3</sup> )	Zwarte koolstof <sup>a</sup> (µg/m <sup>3</sup> )	Stikstofdioxide (µg/m <sup>3</sup> )
Voorstedelijk	3	8	1,0	31
Stadspark	8	11	1,6	28
Stedelijke achtergrond	30	15	3,1	39
Street canyon <sup>b</sup>	14	36	-	61
Ring	255	44	3,6	82

<sup>a</sup> Enkel gemeten in oktober 2013

<sup>b</sup> Enkel gemeten in februari 2013

#### Dagverlopen

- Het gemiddelde verloop van de verkeersintensiteit doorheen de dag was voor alle plaatsen erg gelijkaardig tijdens de twee meetperiodes;
- Op weekdagen was er op alle meetplaatsen een duidelijk dagverloop in de TNC met pieken tijdens de ochtend- en avondspits. In de weekends was het aantal deeltjes gemiddeld lager en schommelde het minder doorheen de dag dan op weekdagen;
- Dit dagverloop van de TNC stemde per meetplaats visueel overeen met het dagverloop van de verkeersintensiteit.

#### Temporele correlatie

- De TNC was sterk gecorreleerd tussen de stedelijke achtergrondmeetplaats en de vier andere meetplaatsen. Het verband verschilde wel naargelang de meetperiode. In vergelijking met de stedelijke achtergrondmeetplaats was de TNC:
  - 34% (februari) en 55% (oktober) lager op de voorstedelijke meetplaats;
  - 6% (februari) en 35% (oktober) lager op de meetplaats in het Stadspark;
  - 95% (februari) hoger op de meetplaats in de *street canyon*;
  - 120% (februari) en 170% (oktober) hoger op de meetplaats langs de ring.
- Het aantal deeltjes in de beschouwde grootteklassen was matig tot sterk gecorreleerd tussen de stedelijke achtergrondmeetplaats en de vier andere plaatsen. De correlatie was lager voor deeltjes van 20-30 en 30-50 nm en nam toe voor de grotere deeltjesklassen. Dit geeft aan dat de kleinere deeltjes meer lokaal beïnvloed werden.

#### Ruimtelijke divergentie

- In vergelijking met de stedelijke achtergrondmeetplaats was de ruimtelijke divergentie van de TNC het laagst voor de meetplaats in het Stadspark en het hoogst voor de meetplaats in de *street canyon*;
- De ruimtelijke variabiliteit van het aantal deeltjes per klasse was hoger voor deeltjes <50 nm dan voor grotere deeltjes;
- De ruimtelijke variabiliteit van de deeltjesaantallen was hoger in oktober dan in februari.

#### Deeltjes in relatie tot andere luchtkwaliteitsparameters en verkeersintensiteit

We gingen het verband na tussen het aantal en de grootteverdeling van de deeltjes, drie andere luchtkwaliteitsparameters en de verkeersintensiteit. Tabel 2 vat de gemiddelde waarden samen van de gebruikte variabelen. Dit illustreert dat de gemiddelde concentratie van zwarte koolstof, NO<sub>2</sub> en het deeltjesaantal op een meetplaats algemeen toenam bij een hogere blootstelling aan verkeer.

Uit de kwantitatieve relaties bleek dat:

- De TNC per meetplaats in oktober was gecorreleerd ( $r > 0.71$ ) met de massaconcentratie van  $PM_{10}$ , op basis van dagwaarden. Het aantal dagwaarden was echter beperkt en de relatie verschilde naargelang de meetplaats. Voor halfuurwaarden was het verband tussen de TNC en  $PM_{10}$  veel zwakker. Dit komt wellicht doordat  $PM_{10}$  minder wordt beïnvloed door de uitstoot van voertuigen dan het aantal deeltjes;
- De TNC per meetplaats en periode was sterk gecorreleerd ( $r > 0.68$ ) met zwarte koolstof op halfuurniveau. Grotere deeltjes waren sterker gecorreleerd met zwarte koolstof dan kleinere, en voor alle meetplaatsen was het verband maximaal voor deeltjes in de grootteklasse van 100-200 nm ( $r > 0.76$ ). De relatie verschilde naargelang de meetplaats en periode. De helling van de regressie leek in verband te staan met de verkeersblootstelling van de meetplaats;
- De TNC op alle meetplaatsen was sterk gecorreleerd ( $r = 0.94$ ) met  $NO_2$  op weekniveau. Kleinere deeltjes waren sterker gecorreleerd met  $NO_2$  dan grotere deeltjes;
- De TNC per meetplaats en periode was gecorreleerd ( $r = 0.22$  tot  $0.60$ ) met de verkeersintensiteit op basis van halfuurwaarden. De correlatie was het sterkst voor kleine deeltjes (<20 en 20-30 nm). De relatie tussen het deeltjesaantal en de verkeersintensiteit was sterk afhankelijk van de meetplaats.

### Algemene conclusies

- De stedelijke achtergrondmeetplaats in deze studie kan als representatief beschouwd worden voor UFP in een stedelijke woongemeenschap, maar niet voor een landelijk achtergrondgebied of voor plaatsen met een hogere verkeersblootstelling;
- Het deeltjesaantal op vijf plaatsen en tijdens 2 periodes werd beïnvloed door de gemeten verkeersintensiteit, maar bleek ook afhankelijk van factoren zoals de karakteristieken van de meetplaats, de nabijheid tot lokale bronnen en weerkundige omstandigheden;
- Tijdens deze korte studie werden positieve, kwantitatieve verbanden vastgesteld tussen het deeltjesaantal in de omgevingslucht en  $PM_{10}$ , zwarte koolstof en  $NO_2$ , maar de relaties varieerden per meetplaats en periode;
- Dit suggereert dat het deeltjesaantal niet kan benaderd worden door het meten van andere, beter gekende luchtkwaliteitsparameters. Meer gegevens zijn nodig om na te gaan of en welke relaties er op langere termijn bestaan tussen deze parameters en het aantal en de grootteverdeling van deeltjes in de omgevingslucht.



## Summary

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### Background

This report presents results on the spatial and temporal variability of the number of particles in Antwerp based on two measuring campaigns of a month. The study was carried out under the authority of the Flemish Environment Agency (VMM) within the framework of the Joaquin (Joint Air Quality Initiative) project supported by the INTERREG IVB NWE programme.

Elevated concentrations of airborne particulate matter (PM) have negative impacts on human health. Current air quality legislation about PM is focused on the mass concentration ( $\mu\text{g}/\text{m}^3$ ) of airborne particles. The European legislation specifies limit values for the mass concentration of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , i.e. particles with an aerodynamic diameter smaller than 10 and 2.5  $\mu\text{m}$ , respectively. However, there are indications that also other metrics than the bulk mass concentration are relevant to human health.

Therefore there is considerable interest in ultrafine particles, which have an aerodynamic diameter smaller than 0.1  $\mu\text{m}$  ( $\text{PM}_{0.1}$  or UFP). Ultrafine particles contribute little to the PM mass concentration in ambient air, but have high number concentrations ( $\text{particles}/\text{cm}^3$ ). In urban environments, motorized traffic is the dominant source of primary UFP. The negative health impacts of UFP have been shown through toxicological studies, but epidemiological evidence is still scarce due to limited UFP monitoring sites and few long-term studies.

### Aims

The main research questions of the study were:

- How vary UFP with increasing distance to a main road;
- Is one monitoring station representative to measure UFP in the Antwerp region;
- Is there a relationship between UFP and other air quality parameters;
- Is there a relationship between UFP and the proximity to and intensity of traffic?

### Methodology

There were 2 measuring campaigns of 4 weeks. The total number concentration (TNC) and the size distribution of particles  $<1 \mu\text{m}$  were measured at 5 sites in February 2013 and at 4 of these sites again in October 2013. The sites were located in Antwerp (Belgium) and were selected to cover a wide range of exposure to traffic, depending on the traffic intensity and distance to the road. The measurements were carried out in collaboration with the Flemish Institute for Technological Research (VITO) and the *Institut Scientifique de Service Public* (ISSeP).

The measurements were carried out at the following sites (Table 1):

- An urban background site with a permanent VMM air quality monitoring station;
- 2 sites that were less traffic-exposed than the station: public park and suburban site;
- 2 sites that were more traffic-exposed than the station: street canyon<sup>2</sup> and ring road site.

*Table 1: Overview of sampling sites, ordered according to increasing traffic exposure*

Site	Nearest street address	Distance to street (m)
Suburban	Frederik Van Eedenplein	50
Public parc	Rubenslei	45
Urban background	Plantin en Moretuslei 285	30
Street canyon	Turnhoutsebaan 92	2
Ring road	Noordersingel	10

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<sup>2</sup> A street canyon is a relatively small street flanked by high buildings on both sides

The maximum distance between 2 sites was 4.2 km. During the February 2013 campaign, there was also a gradient study. Perpendicular to a main road near the urban background station 2 extra sites were included to study the impact of the distance to the road on the number of particles.

At each sampling site, number concentrations were determined with two devices:

- A condensation particle counter to measure the TNC on a 1-minute basis;
- A scanning mobility particle sizer or a UFP monitor to measure the size distribution of fine particles on a 10-minute basis. The number concentrations were aggregated to the following size classes: <20, 20-30, 30-50, 50-70, 70-100, 100-200 and >200 nm.

The measurements were corrected for diffusional losses and for differences between instruments determined during preceding comparative measurements. In October we also determined the mass concentrations of black carbon (BC) and PM<sub>10</sub>. Nitrogen dioxide (NO<sub>2</sub>) was measured at all sites on a weekly basis. Traffic intensity (number of vehicles per 15 min) on the road nearest to each sampling site was determined as well.

The meteorological conditions mainly differed between the 2 campaigns for temperature and wind:

- The temperature and regional wind speed (measured at 30 m height) were lower in February than in October;
- The dominant regional wind direction was north-northeast in February and south to southwest in October.

### **Gradient study near a main road**

The gradient study in February 2013 evaluated the number concentration and the size distribution of particles at 3 sampling sites at 10, 30 and 55 m distance of a main road (2 x 2 lanes) near the urban background site. The main results were:

- The total number concentration of particles < 1 µm (TNC) was higher at 10 m from the road than at 30 and 55 m. The concentration at 55 m was slightly higher than at 30 m, probably due to other local sources, so that for the total 4-week period no clear gradient was observed;
- For downwind conditions the TNC was on average 17% lower at 30 m from the road than at 10 m and 22% lower at 55 m from the road than at 10 m;
- For downwind conditions the number of particles <25 nm decreased with increasing distance to the road. The larger particles differed less between the three distances.

### **Intra-urban variability of particle number**

The intra-urban study analysed the variability of the number concentration and the size distribution of particles at 5 sites in February 2013 and at 4 sites in October 2013. We investigated mean values and diurnal variations per site as well as temporal correlations and spatial divergence between sites.

#### Mean values

- The traffic intensity on the roads near the sites varied strongly (Table 2). This confirmed that sites with a different exposure to local traffic sources had been selected;
- The mean TNC per site varied strongly (Table 2), with a maximum to minimum ratio of 4 to 5 on week days and of 7 in the weekends;
- The mean TNC per site increased with mean increasing traffic intensity near the site (Table 2), except for the urban background site and the street canyon site. The high number of particles at the urban site can be explained by the high exposure to traffic due to the street canyon effect and the close proximity of the sampling location to the road (Table 1);
- The particle size distribution generally had a similar trend for all sites and periods. The highest number concentrations were found on week days for particles <20 nm (on the average 26% of the TNC). In the weekends the size classes <20 nm and 30-50 nm contributed equally to the TNC (about 20%). Ultrafine particles dominated the TNC, accounting on the average for 87% of the TNC on week days and 81% in the weekend.

#### Diurnal variations

- The mean variation of the traffic intensity throughout the day was for all the sites very similar during the 2 campaigns;

- On week days all sites showed a diurnal variation in the TNC with peaks during the morning and evening rush hours. In the weekends the number of particles was lower on the average and less variable through the day than on week days;
- This diurnal variation in the TNC per site was visually in line with the diurnal pattern of the traffic intensity.

*Table 2: 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	Traffic intensity (10 <sup>3</sup> vehicles/day)	Particle number (10 <sup>3</sup> particles/cm <sup>3</sup> )	Black carbon <sup>a</sup> (µg/m <sup>3</sup> )	Nitrogen dioxide (µg/m <sup>3</sup> )
Suburban	3	8	1.0	31
Public parc	8	11	1.6	28
Urban background	30	15	3.1	39
Street canyon <sup>b</sup>	14	36	-	61
Ring road	255	44	3.6	82

<sup>a</sup> Only measured in October 2013

<sup>b</sup> Only measured in February 2013

#### Temporal correlation

- The TNC was strongly correlated between the urban background site and the 4 other sites. However, this relationship differed between the February and October campaigns. Compared to the urban background site the TNC was:
  - 34% (February) and 55% (October) lower at the suburban site;
  - 6% (February) and 35% (October) lower at the public park site;
  - 95% (February) higher at the street canyon site;
  - 120% (February) and 170% (October) higher at the ring road site.
- The particle number in the distinguished size classes was moderately to strongly correlated between the monitoring station and the 4 other sites. The correlation was lower for particles of 20-30 and 30-50 nm and increased for the larger size classes. This indicates that the smaller particles were more locally influenced.

#### Spatial divergence

- In comparison with the urban background site, the spatial divergence in the TNC was lowest at the public park site and highest at the street canyon site;
- The spatial variability in the number concentration per size class was generally higher for particles <50 nm than for larger particles;
- The spatial variability in particle numbers was higher in October than in February.

#### Particle number in relation to other air quality variables and traffic intensity

We studied relationships between the number concentration and the size distribution of (ultra)fine particles, 3 air quality parameters and the traffic intensity. Table 2 summarizes the mean values of the variables used. It illustrates that the mean concentration of BC, NO<sub>2</sub> and the particle number at a site increased with increasing traffic exposure of the site.

The quantitative relationships revealed that:

- The TNC per site in October was correlated ( $r > 0.71$ ) with the mass concentration of PM<sub>10</sub>, based on daily means. However, the daily data set was limited and the relationship varied per site. At the half-hourly level the relationship between TNC and PM<sub>10</sub> was much weaker. This is most likely due to the fact that PM<sub>10</sub> is less affected by vehicle exhaust emissions than the number of particles;
- The TNC per site and period was strongly correlated ( $r > 0.68$ ) with BC at the half-hourly level. Larger particles were more strongly correlated to BC than smaller particles, and for all sites a maximum correlation was found for particles of 100-200 nm ( $r > 0.76$ ). The relationship varied per site and period, and the slope of the regression appeared to depend on the traffic exposure of the site;

- The TNC at all sites was strongly correlated ( $r = 0.94$ ) with  $\text{NO}_2$  at the weekly level. Smaller particles were more strongly correlated with  $\text{NO}_2$  than bigger particles.
- 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  $20\text{-}30$  nm). The relationship between the particle number and traffic intensity varied clearly between the sites.

### General conclusions

- The urban background station in this study can be considered as a community-representative monitoring site for UFP, but it is less representative for rural background areas and more traffic-exposed sites;
- The particle number at 5 sites and during 2 periods was influenced by traffic intensity, but also depended on other factors such as the site characteristics, the proximity to local sources and meteorological conditions;
- During this short-term study positive relationships were found between UFP and  $\text{PM}_{10}$ , BC and  $\text{NO}_2$ , but the relationships varied by site and period;
- This suggests that particle numbers cannot be approximated by measurements of other more well-known air quality parameters. More data are needed to examine whether and which relationships exist on the long-term between these parameters and the number and size distribution of ambient particles.

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# 1 Introduction

## 1.1 Background

Joaquin (Joint Air Quality Initiative, <http://www.joaquin.eu>) is an EU cooperation project supported by the INTERREG IVB North-West Europe programme (<http://www.nweurope.eu>). The general aim of the project is to support health-oriented air quality policies in Europe. To achieve this, the project will provide policy makers with the necessary evidence on the current local and/or regional situation (e.g. measurements of emerging health relevant parameters), provide them with best-practice measures that can be taken and motivate them to adapt and strengthen their current air quality policies.

Elevated airborne particulate matter (PM) has significant adverse health impacts. Studies indicate that the lifetime exposure to the current concentrations of fine dust leads to a reduction in our average life expectancy by one year. Current EU air quality legislation concerning PM is focused on monitoring, limiting and reducing the mass concentration ( $\mu\text{g}/\text{m}^3$ ) of airborne particles. However, toxicological and epidemiological research argues that other particle metrics may constitute better links to health endpoints than the bulk mass concentration.

For instance, there is evidence that particles from combustion sources (black carbon, BC) are more relevant to human health than particles from other sources. A recent WHO report<sup>3</sup> concluded that for short-term health, BC is a better indicator of harmful particulate substances from combustion sources (especially traffic) than undifferentiated PM mass. However, the evidence from long-term studies for the relative strength of association is still inconclusive. While BC may not be a major directly toxic component of PM, it may operate as a universal carrier of a wide variety of chemicals of varying toxicity to the lungs, the body's major defense cells and possibly the systemic blood circulation.

There is also considerable interest in ultrafine particles (UFP), which is particulate matter with a diameter smaller than  $<0.1 \mu\text{m}$  (100 nm). Ultrafine particles contribute only little to the ambient  $\text{PM}_{10}$  mass and the number of ambient particles usually increases sharply for diameters decreasing into the UFP range<sup>4</sup>. Therefore, for UFP the number concentration, i.e. number of particles per volume of air ( $\text{particles}/\text{cm}^3$ ), is a more appropriate measurement metric than the mass concentration.

UFP are able to penetrate deeply into the respiratory system and deposit efficiently in the lower human lung. Exposure to UFP has been linked to pro-inflammatory response at the cellular level and to increased incidence of e.g. asthma<sup>5</sup>. The negative health impacts of UFP have been shown through toxicological studies, but epidemiological evidence is still scarce due to limited monitoring sites and few long-term studies<sup>6</sup>.

In urban environments, motor vehicle traffic is the dominant source of primary UFP, although other sources such as food cooking and wood burning can contribute as well. Secondary atmospheric production of new UFP (new particle formation) can also be important in some conditions. Studies have shown that concentrations of UFP are elevated near roads but then decrease to near urban background concentrations within several hundred meters, primarily as a result of dilution and partly by coagulation. The factors that impact the magnitude and extent of these gradients include traffic conditions, temperature, relative humidity, wind direction and speed, atmospheric stability, and mixing height.

Because measurements of UFP are not included in policy-oriented air quality monitoring programmes, one of the aims of the Joaquin project is to carry out continuous long-term UFP measurements in different cities in the North-West European region. Furthermore, short-term measurements at several sites in a single city are of interest.

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<sup>3</sup> Janssen N.A.H., Gerlofs-Nijland M.E., Lanki T., Salonen R.O., Cassee F., Hoek G., Fischer P., Brunekreef B., Kryzanowski M. (2012). Health effects of black carbon. WHO Regional Office for Europe.

<sup>4</sup> Hinds W.C. (1999). Aerosol technology: properties, behaviour and measurement of airborne particles. John Wiley & Sons.

<sup>5</sup> Moore K.F., Krudysz M.A., Pakbin P., Hudda N., Sioutas C. (2009) Intra-community variability in total particle number concentrations in the San Pedro Harbor area (Los Angeles, California). *Aerosol Sci. Technol.* 43, 587-603.

<sup>6</sup> Bigazzi A.Y., Figliozzi M.A. (2012). Impacts of freeway traffic conditions on in-vehicle exposure to ultrafine particulate matter. *Atmospheric Environment* 60, 495-503.

- As a first step, the instrumental approaches used for UFP monitoring were assessed. From literature review and a laboratory test, an evaluation was made of commercially available UFP devices in order to choose appropriate instrumentation and methodologies to measure the particle number concentration and size distribution under monitoring network conditions.
- In 2012, based on this evaluation three different types of monitors were purchased. Before installing these instruments in monitoring stations across the NWE region, the instruments were compared at an urban background site in Antwerp (Wilrijk; Dec 2012 - Jan 2013). The results of this comparison are described in report 2013/MRG/R/172<sup>7</sup>.
- In 2013, two measurement campaigns on UFP were conducted in Antwerp under the authority of VMM. Different types of air quality locations were selected to find out (1) how the UFP concentration and distribution varies with increasing distance to a main road and (2) if one central monitoring station is representative for UFP concentrations in the Antwerp region.

The results of the first measuring campaign, carried out in February 2013, have already been reported (2013/MRG/R/173)<sup>8</sup>. The second campaign of October 2013 has partly been described in report 2014/MRG/R/59<sup>9</sup>, but this included the measurements at only three of the four sampling sites.

## 1.2 Aims

Measurements of UFP number concentration at a single monitoring station may not be indicative of actual human exposure in the communities surrounding the station. Due to their short atmospheric lifetimes and strong dependence on very local sources, particle number concentrations may vary significantly on short spatial and temporal scales.

Therefore, we were interested in gaining a better understanding of the variance of UFP levels at the local and urban scale. In February 2013, short-term monitoring of UFP concentrations was conducted simultaneously at four locations with varying traffic intensity and exposure and at three sampling points at increasing distance to a busy road. To assess the temporal stability of the observations, the measurements were repeated at a subset of the locations in October 2013.

The present study analyses the spatial-temporal variability of UFP number size distributions and particle number concentrations using one month of measurements at seven (February 2013) and four (October 2013) locations in Antwerp. Next to UFP, nitrogen dioxide (NO<sub>2</sub>) was sampled at all sites. In October, black carbon (BC) and PM<sub>10</sub> were measured as well.

This report combines the main findings of the above-mentioned studies<sup>8,9</sup>. In addition, it includes the following topics that have not been reported before:

- measurements of the public parc location during the October campaign;
- measurements of traffic intensity and relationships between UFP and traffic;
- comparison of the results of the February and October campaigns.

The main questions of the study are:

- how varies the UFP concentration and distribution with increasing distance to a main road;
- is one monitoring station representative for assessing exposure to UFP in the Antwerp region;
- is there a relationship between UFP and other traffic related parameters (NO<sub>2</sub> and BC);
- is there a relationship between UFP and the distance to and intensity of traffic sources?

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<sup>7</sup> Frijns E., Van Laer J., Aerts W., Brabers B., Berghmans P. (2013). UFP instrument comparison at an urban background location in Antwerp. Study accomplished under the authority of VMM. VITO report 2013/MRG/R/172. Flemish Institute for Technological Research, Mol, Belgium. Available at <http://www.joaquin.eu>.

<sup>8</sup> Frijns E., Van Laer J., Berghmans P. (2013). Short-term intra-urban variability of UFP number concentration and size distribution. Study accomplished under the authority of VMM. VITO report 2013/MRG/R/173. Flemish Institute for Technological Research, Mol, Belgium. Available at <http://www.joaquin.eu>.

<sup>9</sup> Frijns E., Van Laer J., Berghmans P., Bergmans B., Lenartz F. (2014). Short-term intra-urban variability of UFP number concentration and size distribution - October 2013 campaign. Study accomplished under the authority of VMM. VITO report 2014/MRG/R/59. Flemish Institute for Technological Research, Mol, Belgium. Available at <http://www.joaquin.eu>.

## 2 Materials and methods

### 2.1 Sampling sites

In February 2013, measurements of ultrafine particles were carried out at seven sites in Antwerp. Table 3 and Figure 1 give an overview of the sites. All sites were within the major ring road around Antwerp, except for the suburban site. In October 2013, measurements occurred at four of the seven sites.

Table 3: Overview of sampling sites

Site	Nearest street address	Lambert coordinates (m)		Distance to street (m)	Sampling in	
		X	Y		Feb	Oct
Gradient - street	Plantin en Moretuslei 285	154402	211058	10	x	
Monitoring station	Plantin en Moretuslei 285	154407	211080	30	x	x
Gradient - parking lot	Plantin en Moretuslei 285	154416	211098	55	x	
Public parc	Rubenslei	153168	211474	45	x	x
Suburban	Frederik Van Eedenplein	151473	212594	50	x	x
Urban	Turnhoutsebaan 92	154418	211732	2	x	
Ring road	Noordersingel	155518	211569	10	x	x



Figure 1: Location of the seven measurement sites in Antwerp. The gradient street site and gradient parking lot site are near the monitoring station



### 2.1.1 Monitoring station at urban background site and gradient study (Plantin en Moretuslei)

The VMM air quality monitoring station Antwerp-Borgerhout (42R801) is located 30 m from a main street, the Plantin en Moretuslei. This is a busy access road that connects the inner city with a major highway eastwards (E313). The road is east-west orientated and has four traffic lanes, two in each direction. A bus stop is situated in the vicinity of the station. As the distance to the road exceeds 10 m, the station is categorised as an urban background site.

The monitoring station is set on a terrain of a primary school, next to the playground. The buildings near the station are one floor high (school) and six floors high (office building). The only other road close by is a local road (Montensstraat) which runs north-south.

In the station the following air pollutants are routinely measured at the half-hourly level: black carbon, NO, NO<sub>2</sub>, SO<sub>2</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, O<sub>3</sub>, ethylbenzene, toluene, mp-xylene, o-xylene and benzene. From February 2013 on, also the total number concentration and size distribution of (ultra)fine particles were measured within the framework of the Joaquin project.

In February 2013 a gradient study was conducted perpendicular to the Plantin en Moretuslei. The gradient consisted of three locations at 10, 30 and 55 m from the nearest lane (Figure 2):

- 10 m: street site;
- 30 m: monitoring station;
- 55 m: parking lot of an office building.



Figure 2: Three measuring locations next to the Plantin en Moretuslei, used for the gradient study

### 2.1.2 Public parc site (Stadspark)

The public parc site is situated in a small city park with a surface area of about 11 ha. The land use consists of mature and mainly deciduous trees, lawns and water bodies. The closest road to the site is the Rubenslei at a distance of 45 m, which is a two-lane local street. Parallel to the Rubenslei a busier road (Frankrijklei; 2 x 3 lanes) is located.

The public parc site is situated approximately 1300 m from the monitoring station in a straight line. The instruments were situated directly next to a local government office. In February 2013 a trailer of ISSeP was used, in October 2013 a trailer of ECN (Figure 3).

### 2.1.3 Suburban site (Frederik Van Eedenplein)

The suburban site is the only site that is situated on the other bank of the river Scheldt. The river lays about 110 m in easterly direction from the suburban site (Figure 4 right). The closest road, the Thonetlaan, is about 45 m from the site in westerly direction. This road has two wide lanes with a large verge in between (total of 30 m). The site itself is characterized by grass, shrubs and small trees. A few meters from the location is a community centre for elderly that was not in use during the UFP measurement campaigns.

The suburban site is situated approximately 3300 m from the monitoring station in a straight line.



Figure 3: Public parc site in February (left) and October 2013 (right)



Figure 4: Suburban site in February (left, view to the Thonetlaan) and October 2013 (right, view to the river Scheldt)

#### 2.1.4 Urban site (Turnhoutsebaan)

The urban measuring site is situated at the Turnhoutsebaan 92. The Turnhoutsebaan runs generally east-west through the city parallel to the Plantin en Moretuslei. The street consists of two traffic lanes and two tram/bus lanes situated in between. On both sides of the road buildings with two to seven floors are present.

The urban site is located approximately 650 m from the monitoring station in a straight line. The instruments were positioned directly next to the traffic lane (Figure 5).

#### 2.1.5 Ring road site (Noordersingel)

The ring road site is situated next to the highway R1, which consists of 11 lanes (6 lanes southerly, 5 lanes northerly) in four different carriageways. The site is located close to the junction Antwerp-East where the E313 connects with the R1. As a result another three traffic lanes were located near the sampling location. The sampling site was located next to the traffic lanes in the Brussels direction. A pumping station was situated further away from the ring road, behind the monitoring infrastructure.

The ring road is located approximately 1200 m from the monitoring station in a straight line. The instruments were positioned about 10 m from the right lane. In February 2013 a weather-proof box was used, in October 2013 a trailer of ISSeP (Figure 6).





Figure 5: Urban site



Figure 6: Ring road site next to the Antwerp-East junction in February (left) and October 2013 (right)

## 2.2 Sampling periods, setup and instruments

The measuring periods were as following:

- February campaign: from 04/02/2013 to 04/03/2013;
- October campaign: from 08/10/2013 to 04/11/2013.

### 2.2.1 Ultrafine particles (UFP)

During the February campaign the UFP instruments were set up in trailers (gradient - street site, public parc site, suburban site), a stationary monitoring station (urban background site), a truck (gradient - parking lot site) or weather proof housings (urban site and ring road site) (Table 4). During the October campaign only trailers (public parc, suburban, ring road site) and a stationary monitoring station were used (Table 5).

The measuring cabins were all equipped with air conditioning.

The total particle number concentration (TNC) was measured with an Environmental Particle Counter (EPC, TSI 3783). Only at the suburban site in October 2013, the TNC at the suburban location was measured with a Condensation Particle Counter (CPC, TSI 3786) (Table 5).

Table 4: Overview of housings and instrumentation per sampling site in February 2013

Site	Housing	TNC	Size distr.	NO <sub>2</sub>	BC	PM <sub>10</sub>
Gradient - street	Trailer A	EPC	Grimm SMPS	Sampler	MAAP	-
	Station	EPC	Grimm SMPS	Sampler + 42i	MAAP	ESM
Gradient - parking lot	Truck	EPC	Grimm SMPS	Sampler	-	-
Public parc	Trailer B	EPC <sup>a</sup>	lft cust. SMPS	Sampler	-	-
Suburban	Trailer C	EPC <sup>a</sup>	lft cust. SMPS	Sampler	-	-
Urban	Box	EPC <sup>b</sup>	UFP monitor	Sampler	-	-
Ring road	Box	EPC <sup>b</sup>	UFP monitor	Sampler	-	-

<sup>a</sup> From 18/02 to 04/03/2013 only; sampling from high volume inlet together with lft customized SMPS

<sup>b</sup> From 04/02 to 18/02/2013 only; sampling from TSI 3031200 together with UFP monitor

Table 5: Overview of housings and instrumentation per sampling site in October 2013

Site	Housing	TNC	Size distr.	NO <sub>2</sub>	BC	PM <sub>10</sub>
Gradient - street	-	-	-	-	-	-
Monitoring station	Station	EPC	Grimm SMPS	42i	MAAP	ESM
Gradient - parking lot	-	-	-	-	-	-
Public parc	Trailer A	EPC	Grimm SMPS	42i	MAAP	Gravimetric
Suburban	Trailer C	CPC	lft cust. SMPS	Sampler	AE33	Grimm PDM
Urban	-	-	-	-	-	-
Ring road	Trailer B	EPC	lft cust. SMPS	Sampler	AE22	Grimm PDM

The particle size distribution was measured with three different types of instruments:

- Grimm SMPS: scanning mobility particle sizer consisting of an <sup>85</sup>Kr bipolar charger (185 MBq), a long differential mobility analyser (DMA, Grimm) and a butanol-based CPC (Grimm 5420).
- lft customized SMPS: scanning mobility particle sizer consisting of a <sup>63</sup>Ni bipolar charger (100 MBq), a long DMA (lft) and a butanol-based CPC (TSI 3772).
- UFP monitor: ultrafine particle monitor (TSI 3031) consisting of a unipolar charger (corona needle), a long DMA (TSI) and an electrometer.

The following sampling systems were used:

- TSI environmental sampling system (TSI 3031200) with PM<sub>10</sub> head, PM<sub>1</sub> cyclone and Nafion dryer and sampled 3 L/min (EPC) and 5 L/min (UFP monitor), respectively.
- Grimm sampling system with TSP head and Nafion dryer and sampled 0.3 L/min.
- High flow sampling system with PM<sub>10</sub> head, Nafion dryer and a long sampling tube with a high flow rate (2.4 m<sup>3</sup>/h or 40 L/min).

The UFP monitors were connected to a TSI sampling system, the Grimm SMPSs were connected to a Grimm sampling system and the lft customized SMPSs were connected to the high flow sampling system. The EPCs were connected to a TSI sampling system or the customized sampling system. During the February campaign, five EPCs were installed at seven sites, resulting in four weeks of consecutive measurements at three sites (gradient study at the Plantin en Moretuslei) and two weeks of consecutive measurements at the four other sites (Table 4):

- When the EPCs were positioned at the urban site and ring road site (04/02 to 18/02/2013), they were connected to the TSI environmental sampling system together with a UFP monitor.
- When the EPCs were positioned at the public parc and suburban site (18/02 to 04/03/2013), they were connected to the high flow system together with the lft customized SMPSs.
- In trailer A, the EPC and UFP monitor were always connected to a single TSI sampling system.

Regarding the characteristics of the particle counters, the CPC 3786 has a lower size limit ( $D_{50}$  down to 2.5 nm) than the EPC 3783 (7 nm) (Table 6). This difference is taken into account by a correction factor, as described below (2.3.2). Both instruments use water as condensing fluid.

The Grimm and IFT customized SMPS both determine the particle size distribution for a large number of size classes in a similar range. The higher upper particle size of the Grimm SMPS (Table 6) is of little relevance because the relatively small number of particles in the larger size classes.

In contrast to the two types of SMPS, the UFP monitor only measures six size classes: 20-30, 30-50, 50-70, 70-100, 100-200 and 200-800 nm. This monitor consequently gives no information on the number of particles <20 nm. Furthermore, the instrument comparison indicated that particles >200 nm are not reliably determined by the UFP monitor.

*Table 6: Overview of instrument characteristics and settings used*

	EPC	CPC	UFP monitor	Grimm SMPS	IFT cust.
	TSI 3783	TSI 3786	TSI 3031	5420 L-DMA	SMPS
Lower particle size (nm)	7	2.5	20	10	8.5
Upper particle size (nm)	1000	1000	200 (800 <sup>a</sup> )	1118	850
Size classes	1	1	6	45	64
Measurement time (min)	1	1	10	10	5
Aerosol flow rate (L/min)	0.12	0.3	4.0	0.3	1.0
Working fluid	Water	Water	-	Butanol	Butanol

<sup>a</sup>The instrument comparison indicated that the particle number in size class 200-800 nm could not reliably be measured by the UFP monitors

The particle counters logged the average TNC per minute. The UFP monitor and Grimm SMPS had a scan time of 10 min and the IFT customized SMPS had a scan time of 5 min (Table 6).

More information on the UFP instruments and sampling systems can be found in the report about the instrument comparison.

### 2.2.2 Particulate matter (PM<sub>10</sub>)

At the monitoring station PM<sub>10</sub> was determined in two ways:

- Gravimetrically: reference method according to EN12341, in which PM<sub>10</sub> is collected for 24 h on a filter (Pall Tissuquartz 2500 QAT-UP) by a sequential sampler (Leckel SEQ 47/50; 2.3 m<sup>3</sup>/h). By weighing the filter before and after exposure, the PM<sub>10</sub> concentration is determined.
- ESM: the ESM FH 62 I-R monitor uses the technique of  $\beta$ -attenuation; the attenuation of  $\beta$ -rays is directly related to the mass on the filter. To avoid condensation of water the inlet is heated. By this process semi-volatile compounds such as volatile organic carbon compounds and ammonium compounds can volatilize from the filter. To account for this underestimation, an experimentally determined calibration factor (x 1.25) is applied to the ESM measurements so that these PM<sub>10</sub> data can be considered to be equivalent with the gravimetric reference method. The ESM PM<sub>10</sub> data were available at the 30-min level.

During the October campaign, PM<sub>10</sub> was also determined at the other sites:

- Gravimetrically (public parc);
- Grimm PDM (suburban and ring road): the Particle Dust Monitor (Grimm 1.109) is an optical particle counter which measures the particle number concentration in 31 size channels from 0.25 to 32  $\mu$ m. The PM mass was derived from the particle number per size class based on a calibration with gravimetric measurements (see Annex 2). The Grimm PDM data were available at the 5-min level.

The VMM obtained accreditation for the gravimetric and ESM measurements of PM<sub>10</sub> (Annex 1).



### 2.2.3 Black carbon (BC)

Half-hourly black carbon (BC) concentrations were measured at the two of the three gradient sites in February 2013 (Table 4) and at all sites in October 2013 (Table 5).

Three types of instruments were used:

- MAAP (gradient street site, monitoring station, public parc site in October): Multi-Angle Absorption Photometer (Thermo Scientific MAAP 5012). The MAAP determines the BC concentration by simultaneous measurement of optical absorption and scattering of light (670 nm) by particles collected on a glass-fibre filter tape.
- AE22 (ring road site): dual wavelength aethalometer (Magee Scientific AE22). The AE22 measures light absorption by particles at two wavelengths: 880 nm (IR), quantitative for the mass of "black" or elemental carbon, and 370 nm (UV), indicating the presence of aromatic organic compounds as found in smoke from burning wood, biomass and tobacco.
- AE33 (suburban site): 7-wavelength aethalometer (Magee Scientific AE33), measuring light absorption by particles at seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm).

For the aethalometers (AE22 and AE33) the measurements at 880 nm were used for BC. Simultaneous measurements at a single site were used to determine instrumental correction factors (see 2.3.2).

### 2.2.4 Nitrogen dioxide (NO<sub>2</sub>)

During the February campaign, weekly average NO<sub>2</sub> concentrations were measured at all sites with diffusive samplers (IVL) outside of the instrument housings. In October, diffusive samplers for NO<sub>2</sub> were used at the ring road and suburban site only.

At the urban background site (monitoring station), the half-hourly NO<sub>2</sub> concentration was measured continuously with a chemiluminescence NO-NO<sub>2</sub>-NO<sub>x</sub> analyser (Thermo Scientific 42i). During the October campaign, a similar monitor was used in trailer A at the public parc site. Annex 1 gives more information on these accredited measurements.

### 2.2.5 Meteorological conditions

The meteorological conditions (temperature, relative humidity, wind speed and wind direction at 30 m height, half-hourly data) were measured at the VMM monitoring station Antwerp-Luchtbal (42M802; Havanastraat; Lambert X = 153884 m, Y = 216970 m) and assumed to be representative for all sites.

For the gradient study during the February campaign, the wind conditions measured at the traffic site were used (Vaisalla instrumentation at 4 m height; trailer A).

### 2.2.6 Traffic intensity

Information on traffic near the sampling sites was obtained from "Agentschap Wegen en Verkeer" and "Verkeerscentrum Vlaanderen" (ring road site).

At the ring road site (junction Antwerp-East), Verkeerscentrum Vlaanderen has permanent counting loops in the road. We used traffic intensities (number of vehicles per time unit) on the 14 individual lanes that pass in the direct neighbourhood of the air quality sampling location.

At the other sites Agentschap Wegen en Verkeer installed temporary counting loops on the road next to the sampling site, which generally functioned well throughout the two UFP campaigns:

- Plantin-Moretuslei: 4 lanes (monitoring station);
- Turnhoutsebaan: 2 lanes (urban site);
- Rubenslei: 2 lanes (public parc);
- Thonetlaan: 2 lanes (suburban site).

During the February campaign, traffic intensities at the public parc site and suburban site were only measured from 22/02/2013 onwards.

The traffic intensities were available at 15-min intervals. For all sites we used the sum of the traffic intensity on the individual lanes. For the ring road site and public parc site more detailed information on the traffic intensity per vehicle type was available, but these were not used in the present study.

## 2.3 Data analysis

All data were screened for irregularities. Data collected during instrument errors or maintenance was removed from the analysis.

### 2.3.1 Diffusion correction

For ultrafine particles, particle losses to the surface of the sampling system and the measuring device can occur via diffusion. Therefore sampling pipes are kept as short as possible and laminar flow conditions are aimed for. Nevertheless, meaningful diffusional losses during sampling and measurement occur for particles <100 µm, so that diffusion correction factors should be applied.

The diffusion correction factors used were:

- manufacturer factors integrated into the instruments algorithms (Grimm SMPS, UFP monitor);
- factors based on the formula for cylindrical pipes by Gormley and Kennedy<sup>10</sup> (IfT customized SMPS);
- factors based on the approximation formula by Hinds (Grimm SMPS).

The difference between the results of the formula by Gormley and Kennedy<sup>11</sup> and by Hinds is limited.

The TNC determined by particle counters can only be corrected for diffusion losses when a size distribution is available. Since size distribution monitors were present at all sites, 10-min averaged EPC and CPC data were corrected for diffusion losses based on the raw size distribution data (from 20 nm onwards) and Hinds correction factors.

More information on the diffusion loss correction can be found in the reports about the UFP measuring campaigns of February and October 2013.

### 2.3.2 Instrument correction

After diffusion loss correction the data were also corrected for instrumental differences. Information on the measurement campaigns involved is given in Annex 2.

### 2.3.3 Data processing and statistical analysis

All data were aggregated to the half-hourly level for further analysis. The size distribution data were aggregated to the following size classes: <20, 20-30, 30-50, 50-70, 70-100, 100-200 and >200 nm. The data were aggregated to the hourly level to calculate diurnal patterns on week days and weekend days. These hourly means were also used to calculate mean values per site and period.

Single linear regression plots and Pearson correlation coefficients were used to compare sites and periods. We used regressions including an intercept as well as regressions forced through the origin, i.e. with a zero intercept. We considered the regressions through the origin as most representative to evaluate differences in temporal variability between sites or periods. The values of the coefficient of determination ( $R^2$ ) of the regressions were calculated using R software<sup>12</sup> since Microsoft Excel does not correctly calculate  $R^2$  values for regressions forced through the origin.

The Pearson correlation coefficient ( $r$ ) is a standard method used to evaluate the (linear) relationship between paired data points. The correlation can vary from 0 (no correlation, independent data points) to  $\pm 1$ , indicating perfect positive or negative correlation. The correlation helps to determine what

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<sup>10</sup> Gormley P.G., Kennedy M. (1949). Diffusion from a stream following through a cylindrical tube. Proceedings of Royal Irish Academy 52, 163-169.

<sup>11</sup> Gormley P.G., Kennedy M. (1949). Diffusion from a stream following through a cylindrical tube. Proceedings of Royal Irish Academy 52, 163-169.

<sup>12</sup> R Development Core Team (2012). R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. ISBN 3-900051-07-0, <http://www.R-project.org>

fraction of the number concentrations at any particular site can be explained by the concentrations simultaneously measured at the other sites.

A limitation of Pearson correlations, however, is that perfect correlation can be observed between two sites where the concentrations vary by a consistent factor. In other words high correlations between paired sites would only imply uniform temporal variation. Therefore, calculating  $r$  alone does not necessarily provide sufficient information to characterize the variability between sites.

Another useful method to characterize the spatial variability between site pairs is using the so-called coefficient of divergence (COD)<sup>13</sup>, which is defined as<sup>14</sup>:

$$COD_{jk} = \sqrt{\frac{1}{n} \sum_{i=1}^n \left( \frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}} \right)^2}$$

where  $j$  and  $k$  are two sites,  $x_{ij}$  and  $x_{ik}$  represent the  $i^{\text{th}}$  concentration for a given sampling period at  $j$  and  $k$ , respectively, and  $n$  is the number of simultaneous observations. The COD for a given pair of sites can vary from 0, when concentrations are identical at both sites, to 1, when concentrations are highly different. So a low COD indicates a high level of homogeneity in concentrations between site pairs, and a high COD the opposite. The COD therefore specifically addresses the limitation to the correlation described above. A COD value larger than 0.2 can be considered heterogeneous.

In the present study CODs were calculated at the hourly level to determine the variability between the monitoring station and the other sites.

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<sup>13</sup> Moore K.F, Krudysz M.A., Pakbin P., Hudda N., Sioutas C. (2009) Intra-community variability in total particle number concentrations in the San Pedro Harbor area (Los Angeles, California). *Aerosol Sci. Technol.* 43, 587-603.

<sup>14</sup> Wilson J.G., Kingham S., Pearce J., Sturman A. (2005). A review of intra urban variations in particulate air pollution: implications for epidemiological research. *Atmos. Environ.* 39, 6444-6462.

### 3 Results and discussion

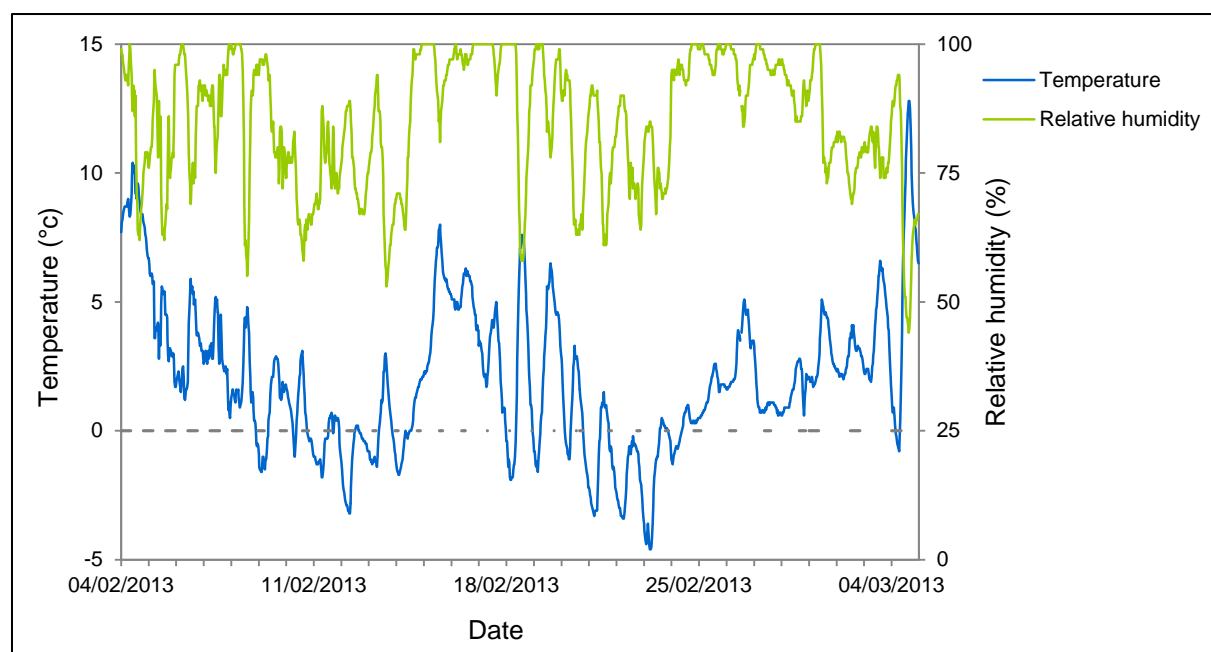
#### 3.1 Meteorological conditions

The temperature of the ambient air at the VMM monitoring station Antwerp-Luchtbal was lower during the measurement campaign in February (2.0 °C) than during the October campaign (12.6 °C) (Table 7). In February it was frequently freezing (Figure 7), resulting in five days with a mean temperature below zero. The temperature (30-min data) varied from -4 to 12°C during the February campaign (Figure 7) and from 4 to 23°C during the October campaign (Figure 8).

The average relative humidity was similar in both measuring campaigns (Table 7). The 30-min relative humidity varied from approximately 50 to 100% (Figure 7, Figure 8). Precipitation during the February campaign (22 mm) was lower than in October (107 mm). There were twice as much as rainy days (> 1 mm precipitation) in October (n = 17) than in February (n = 8).

*Table 7: Daily average, minimum and maximum of meteorological variables during the two campaigns*

Variable	February campaign			October campaign		
	Mean	Min	Max	Mean	Min	Max
Temperature (°C)	2.0	-1.9	8.7	12.6	8.0	17.9
Relative humidity (%)	85	70	99	87	75	97
Precipitation (mm/day)	0.8	0.0	5.0	3.7	0.0	31.4
Wind speed (m/s)	3.8	1.4	6.5	4.8	1.5	10.5



*Figure 7: Time profile of temperature and relative humidity (30 min) during the February campaign*

The wind rose (Figure 9 left) shows that north-northeast wind directions occurred most frequently during the February campaign. In October the main wind direction was south to southwest. The daily mean wind speed at 30 m height was higher in October (4.8 m/s) than in February (3.8 m/s) (Table 7).

During the February campaign the EPC measurements at four sites occurred only during the first two weeks or last two weeks. The right wind rose in Figure 9 indicates that in week 1-2 the regional wind direction at Antwerp-Luchtbal was north-northeast, while it was variable in week 3-4.

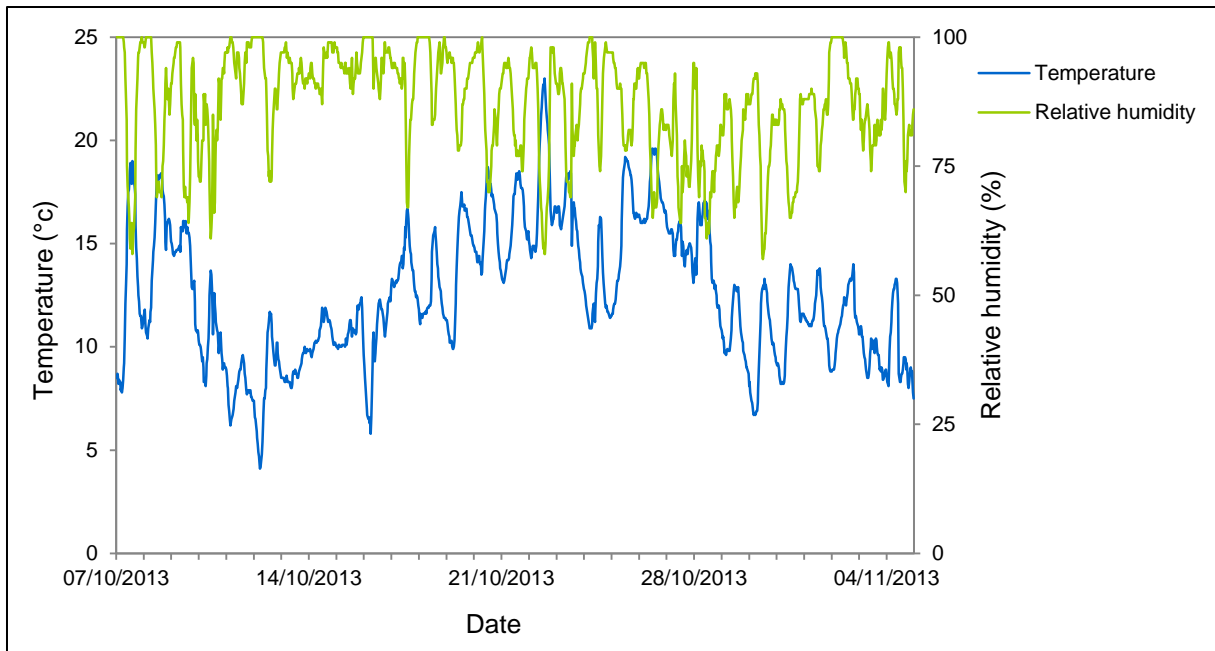


Figure 8: Time profile of temperature and relative humidity (30 min) during the October campaign

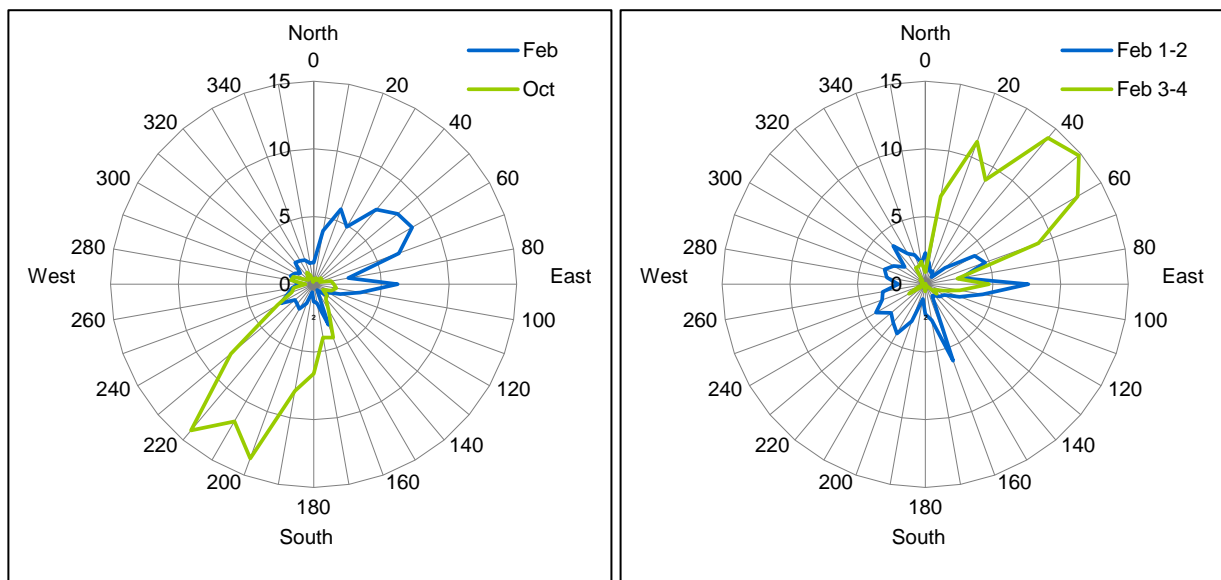


Figure 9: Wind rose (frequency (%) of wind directions) during the February and October campaigns (left) and during week 1-2 and week 3-4 of the February campaign (right)

For the gradient study in February, downwind data had to be selected. On a map of the study area a line was drawn perpendicular to the Plantin en Moretuslei, with a wedge starting at the location at 55 m from the street. The downwind range was determined between 150 to 235 degrees, using the wind data measured at the 10 m street site. More details can be found in the report about the February campaign.

## 3.2 Traffic intensity

### 3.2.1 Average traffic intensity

Traffic intensities varied strongly between the sites but were remarkably similar during the February and October campaigns. For both measuring campaigns, on week days and in the weekend, the mean daily traffic intensity on the main road near the sites increased in the following order: suburban < public parc < urban < monitoring station < ring road (Table 8).

*Table 8: Mean traffic intensity (vehicles/day) near the sites on week days and in the weekend during the February and October campaigns. The relative standard deviation (%) is given between brackets*

Site	Week		Weekend		Daily mean
	February	October	February	October	Feb + Oct
Monitoring station	32 004 (6)	32 210 (10)	22 961 (17)	24 478 (15)	29 707
Public parc	8 483 (6)	8 299 (11)	6 037 (13)	6 107 (12)	7 723
Suburban	3 277 (10)	3 197 (14)	2 567 (19)	2 603 (15)	3 045
Urban	13 457 (6)	-	13 577 (10)	-	13 507
Ring road	270 276 (4)	275 373 (6)	197 020 (11)	221 820 (9)	254 687

On the main road near the station there were on average about 32 000 vehicles on week days and 23 000 (February) to 24 500 (October) vehicles in the weekend. This corresponds well to reported values for February 2010, based on video counting: 37 000 vehicles on week days and 25 000 vehicles in the weekend<sup>15</sup>.

The relative standard deviation (standard deviation / mean x 100) on the daily mean traffic volumes was larger in the weekend than on week days. This is caused by combining Saturdays, with relatively more traffic, and Sundays, with less traffic. For example, in February near the monitoring station there were on average about 26 500 vehicles on Saturday and 19 500 vehicles on Sunday.

The daily traffic volume on the road nearest to the public parc site (Rubenslei) was about 25% of the traffic near the monitoring station, both on week days and on weekend days. It should be remarked, however, that parallel to the Rubenslei there is a busier road (2 x 3 lanes) for which no traffic intensity information was available

At the suburban site, the daily traffic volume was only 10% of the traffic near the station. We have no data on the number of ships passing by on the nearby river Scheldt. At the urban site, there were only measurements in February. Expressed as a percentage of the traffic at the monitoring station, the daily traffic volume was 42% on week days and 59% in the weekend.

The highest traffic intensity was recorded on the highway roads near the ring site. Relative to the monitoring station, the daily traffic volume was 8.5 times higher on week days and 8.5 (February) to 9 (October) times higher in the weekend.

### 3.2.2 Diurnal variation in traffic intensity

Figure 10 shows the diurnal variation in the hourly traffic intensity near the sampling sites during week days, except for the ring road site. The traffic peaked during the morning rush hour from 8:00 to 9:00 and generally had another less pronounced peak during the evening rush hour. At the urban site the evening peak was meaningfully higher than the morning peak.

The diurnal patterns were almost identically in the two campaigns, so that for most sites, the lines for February and October almost overlap each other. For the monitoring station, standard deviations are shown in Figure 10, illustrating the low spread on the average hourly traffic intensity and the statistical similarity of the patterns in February and October.

As shown in Figure 11 the traffic intensity during weekend days was generally highest from the late morning until the evening (11:00 to 19:00). At the urban site there was relatively more traffic during the weekend than at the other sites, and with the highest values from 14:00 to 19:00. In the later evening and around midnight, the traffic intensity was higher on weekend days than on week days. Standard deviations in the weekend were larger than during week days because of the combination of Saturdays and Sundays.

<sup>15</sup> Mishra V.K., Kumar P., Van Poppel M., Bleux N., Frijns E., Reggente M., Berghmans P., Panis L.I., Samson R. (2012). Wintertime spatio-temporal variation of ultrafine particles in a Belgian city. *Science of the Total Environment* 431, 307-313.

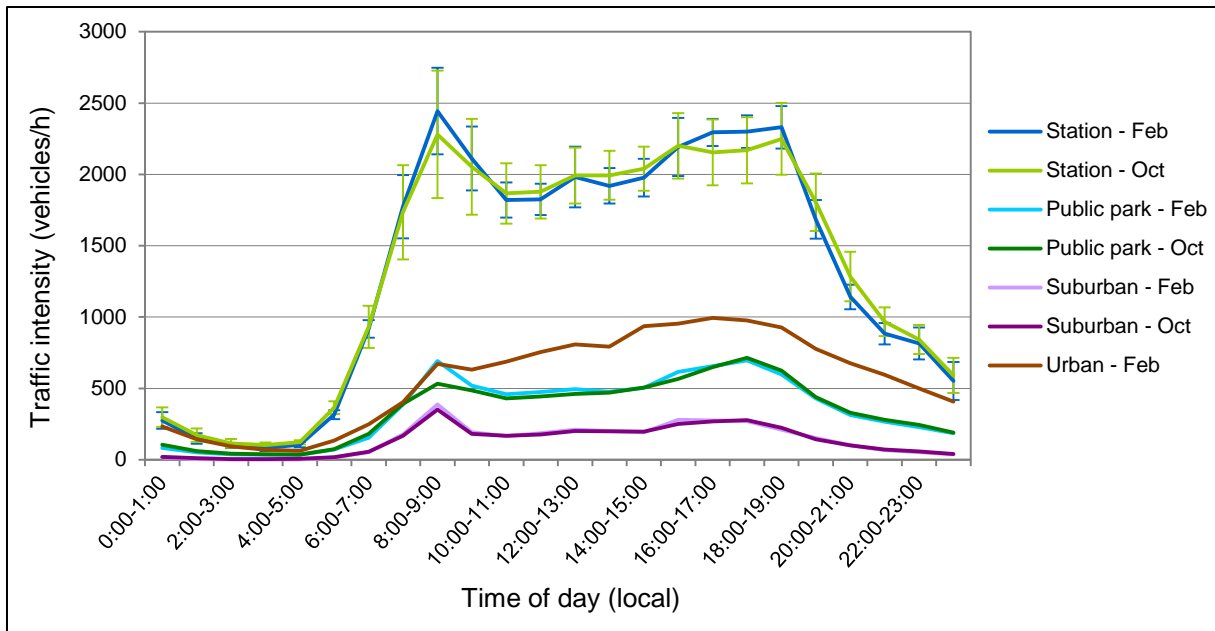


Figure 10: Hourly average traffic intensity on week days near all sites except for the ring road. Standard deviations are shown for the monitoring station only

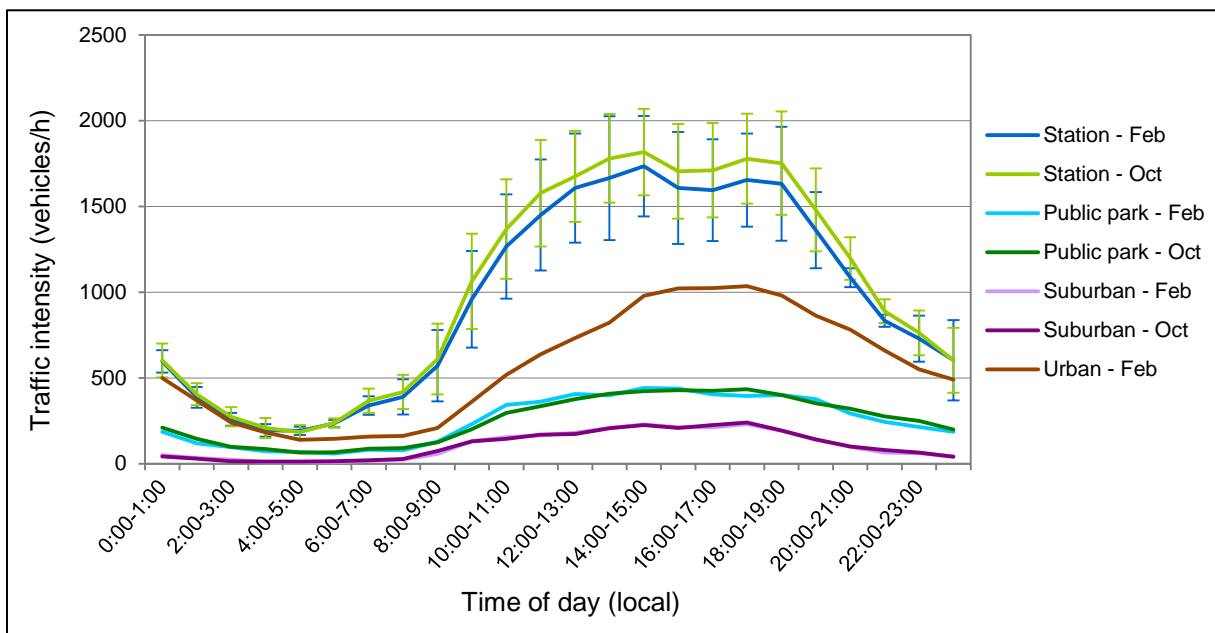


Figure 11: Hourly average traffic intensity in the weekend near all sites except for the ring road. Standard deviations are shown for the monitoring station only

Figure 12 and Figure 13 show the hourly traffic intensity on the highways near the ring road site on week days and weekend days, respectively. For the ease of comparison the diurnal patterns at the monitoring station (also shown in Figure 10 and Figure 11) are given as well.

At the ring road site, traffic on week days peaked earlier in the morning than at the other sites (7:00 to 8:00) and the evening peak was higher and more pronounced. During the weekend, the highest traffic intensity was observed from 14:00 to 19:00. At night, from 23:00 to 4:00, the traffic intensity near the ring road site was higher during the weekend than on week days. For example, during the February campaign, from 0:00 to 1:00 there were on average about 4 500 vehicles in the weekend and about 2 000 vehicles on week days.

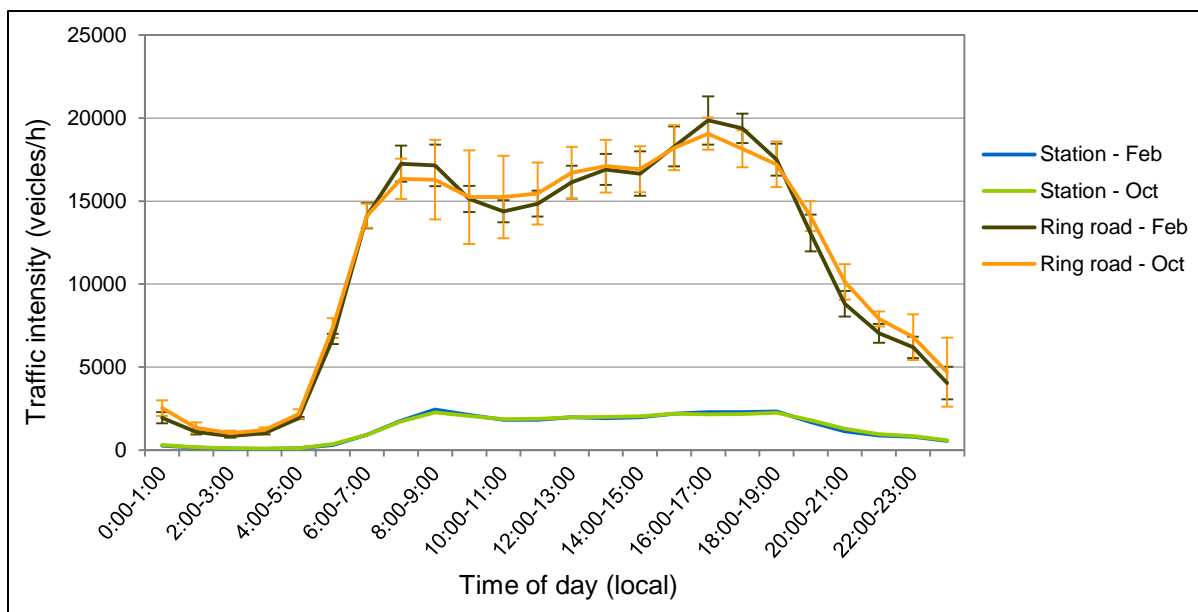


Figure 12: Hourly average traffic intensity on week days near the station and ring road site. Standard deviations are shown for the ring road only

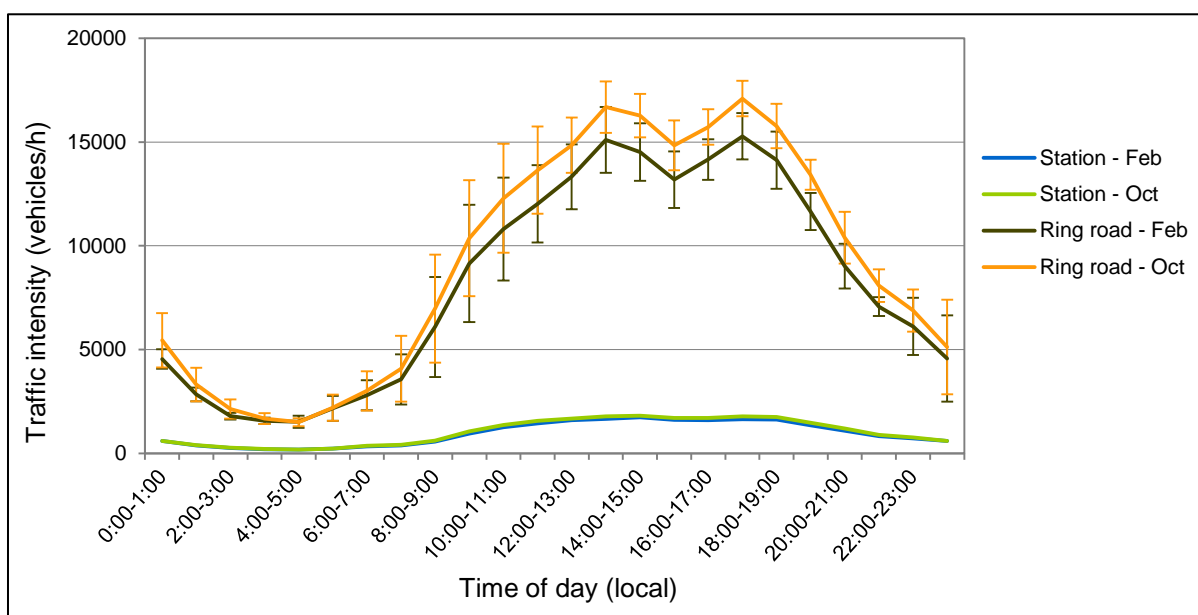


Figure 13: Hourly average traffic intensity in the weekend near the station and ring road site. Standard deviations are shown for the ring road only

### 3.3 Gradient study at monitoring station

The results of the gradient study at the Plantin en Moretuslei in February 2013 are given in detail in the report about the February campaign. The correlations presented here are based on half-hourly data, while the report about the February campaign used 10-min data. The correlation coefficients were generally higher for the half-hourly data than for the 10-min data.

The diurnal variability in the total number concentration of particles  $<1 \mu\text{m}$  (TNC) at the monitoring station is given in section 3.4. For the three sites at the Plantin en Moretuslei (10, 30 and 55 m from the road) a similar diurnal pattern was observed.

Figure 14 shows TNC at 30 m (monitoring station) and 55 m (parking lot) from the road as a function of TNC at 10 m (street site). The regressions shown in Figure 14 were forced through the origin. These are considered to be most representative to evaluate differences in temporal variability between



sites and allow an easy interpretation of the slopes. We also calculated linear regressions including an intercept. Table 9 summarizes the results of both types of regressions.

Figure 14 illustrates that there was a strong correlation between TNC at the three sites. Irrespective of the wind direction, the slope of the regressions through the origin was below 1, indicating a lower TNC at 30 and 55 m than at 10 m. Based on the slopes there were on average 15% less particles at 30 m than at 10 m and 11% less particles at 55 m than at 10 m. The higher TNC at 55 m than at 30 m could be caused by other local sources such as the road in easterly direction from the 55 m site and the parking lot.

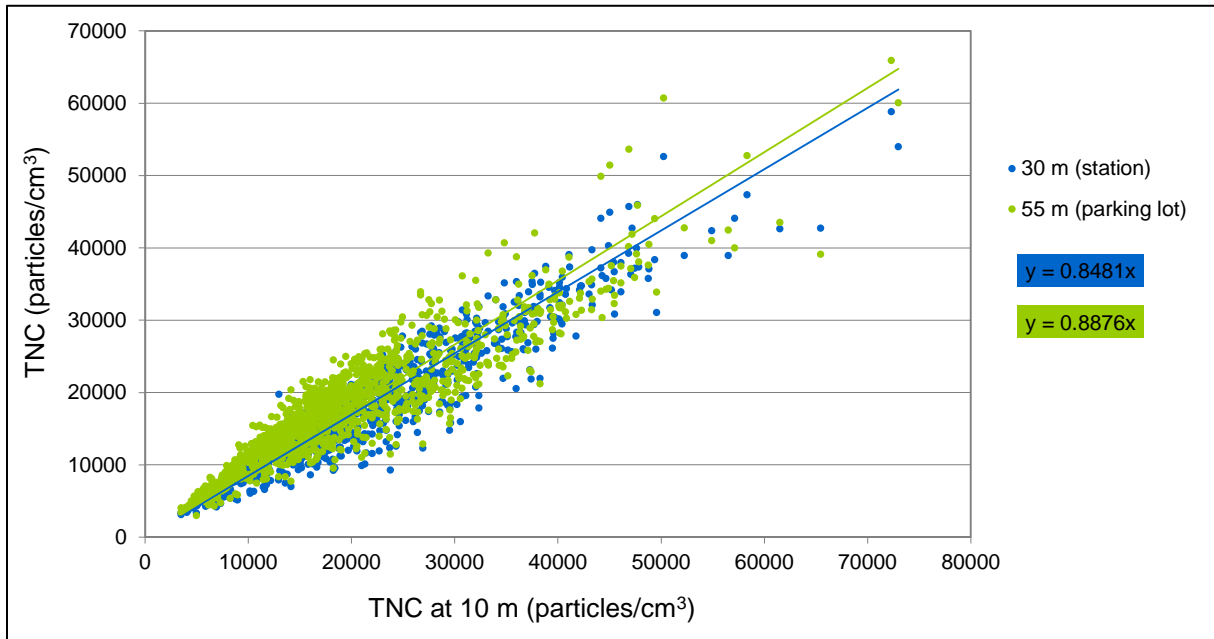


Figure 14: Total particle number concentration (TNC) at 30 and 55 m from the street as a function of TNC at 10 m, irrespective of wind conditions

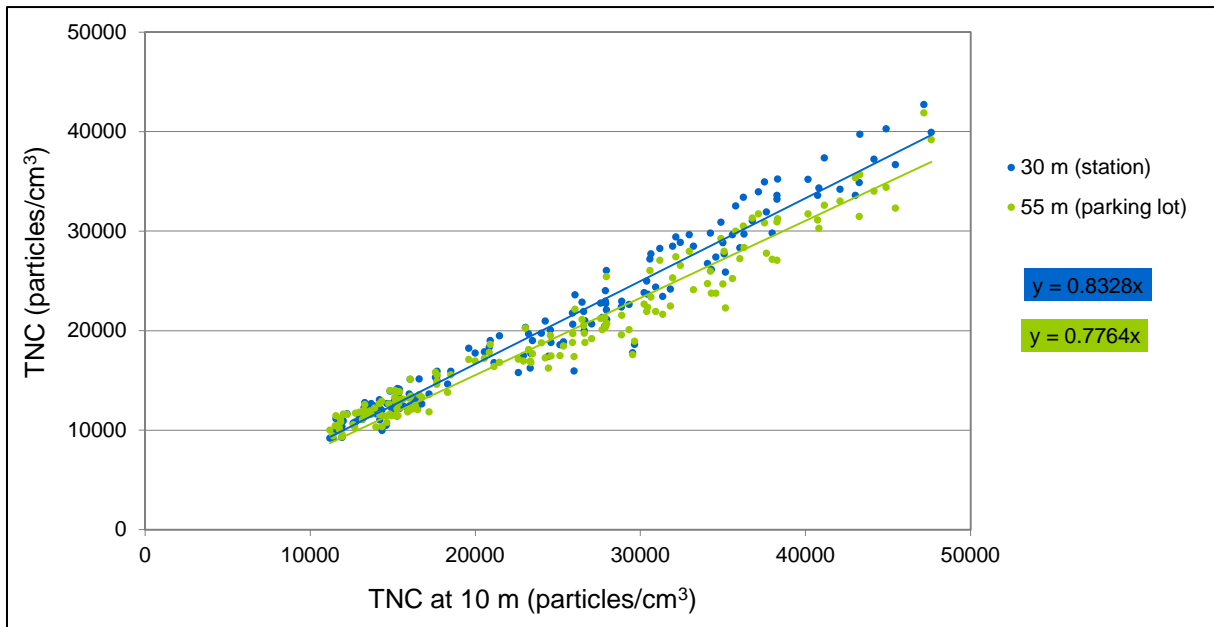


Figure 15: Total particle number concentration (TNC) at 30 and 55 m from the street as a function of TNC at 10 m during downwind conditions

Table 9: Slope ( $a$ ), intercept ( $b$ ) and coefficient of determination ( $R^2$ ) of single linear regressions ( $y = ax + b$ ) and regressions forced through the origin ( $y = ax$ ) between total particle number concentration (TNC) at 10 m and TNC at 30 and 55 m for all data and for downwind conditions. The Pearson correlation coefficient ( $r$ ) is mentioned for the regressions through the origin

Site	Data set	$y = ax + b$				$y = ax$	
		$a$	$b$	$R^2$	$r$	$a$	$R^2$
30 m - station	All (February)	0.768	1927	0.89	0.95	0.848	0.85
	Downwind	0.838	-152	0.96	0.98	0.833	0.99
	Downwind week days	0.842	170	0.95	0.97	0.847	0.99
	Downwind weekend	0.768	788	0.97	0.99	0.802	0.99
55 m - parking	All (February)	0.749	3319	0.84	0.92	0.888	0.97
	Downwind	0.732	1253	0.95	0.97	0.776	0.22
	Downwind week days	0.738	1478	0.94	0.97	0.785	0.99
	Downwind weekend	0.661	2234	0.95	0.98	0.758	0.99

We also restricted the data set to periods when the wind was blowing perpendicular to the road along the three sampling locations. Figure 15 shows the regression plots for these downwind conditions. For this period the slopes of the regressions through the origin were 0.83 for the 30 m site and 0.78 for the 55 m site (Table 9). So compared to the street site at 10 m from the road, during downwind conditions there were on average 17% less particles at 30 m and 22% less particles at 55 m. It should be noted that there were only 80 hours or about 3 days of downwind conditions during the 4-week period.

We also examined the downwind periods on week days and in the weekend separately. For downwind conditions on week days (13/02/2013 13:00 until 15/02/2013 07:00) there were on average 15% less particles at 30 m and 21% less particles at 55 m than at 10 m (Table 9). During the weekend (09/02/2013 11:00 until 11/02/2013 00:00) there were on average 20% less particles at 30 m and 24% less particles at 55 m than at 10 m (Table 9).

The particle size distribution at the three gradient sites had a similar shape for all the data of the 4-week February campaign. The report about this campaign gives more information on the particle size distribution on week days and in the weekend, for downwind and wind-independent conditions. In summary, on week days the biggest difference in the downwind average size distributions between the 10, 30 and 55 m location was found for particles from 10 to about 30 nm. A single mode (unimodal distribution) was present for all three locations. For particles from 30 nm on no difference between the size distribution peaks could be observed between the sites. In contrast to the expectations, the peak particle diameter decreased in size with increasing distance from the road. Evaporation may explain the decrease in particle size between the 10 and 30 m location. The wind-independent mean particle size distributions were smoother than the downwind distributions due to the larger data set and were probably more influenced by stable surrounding sources than by traffic from the main street.

To determine whether ambient particles with different size varied differently with increasing distance to the road, the particle number in four size classes (10-25, 25-50, 50-100, 100-200 nm) was normalized by dividing the number concentration per size range by the TNC at 10 m. For downwind conditions on week days, the normalized particle number concentrations for different size ranges showed a linear decrease in 10-25 nm particles with increasing distance to the road. For downwind conditions during the weekend, the decrease of 10-25 nm particles appeared to be more exponential. The normalized particle concentrations for larger particles were more comparable at the three sites along the gradient.

### 3.4 Intra-urban variability in particle concentration

#### 3.4.1 Average total particle number concentration

Table 10 summarizes the mean total particle number concentration (TNC) on week days and weekend days for the two measuring campaigns. The overall mean is also given.

It should be noted that the EPC data for the February campaign did not cover the same period for the five sites. Three EPCs were available and one of these had to measure continuously at the monitoring station. Therefore the other two EPCs measured the first two weeks at the urban and ring road sites and during the last two weeks at the suburban and public parc sites.

Table 10: Mean total particle number concentration (particles/cm<sup>3</sup>, hourly EPC data) on week days and weekend days during the February and October campaigns

Site	Week		Weekend		Mean
	February	October	February	October	Feb + Oct
Monitoring station	15 188	16 710	14 644	11 318	15 101
Public parc	12 417 <sup>a</sup>	11 673	9 564	7 810	11 086
Suburban	9 426 <sup>a</sup>	8 842	5 898	5 898	8 210
Urban	35 169 <sup>b</sup>	-	37 148	-	35 735
Ring road	39 664 <sup>b</sup>	50 451	41 175	42 599	44 152
Max / min ratio	4.2	5.7	7.0	7.2	5.4

<sup>a</sup> From 18/02 to 04/03/2013 only. The mean TNC at the monitoring station then was 11 937 particles/cm<sup>3</sup>

<sup>b</sup> From 04/02 to 18/02/2013 only. The mean TNC at the monitoring station then was 17 566 particles/cm<sup>3</sup>

The mean daily TNC values differed clearly between the five sites. For both campaigns, on week days and in the weekend, the mean daily TNC per site increased in the following order: suburban < public parc < monitoring station < urban < ring road (Table 10). For example, on week days in February, the mean daily TNC varied by a factor of 4.2, ranging from about 9 500 particles/cm<sup>3</sup> at the suburban site to about 40 000 particles/cm<sup>3</sup> at the ring road site.

The order of sites for the mean TNC was the same as for the traffic intensity (section 3.2.1), except for the urban site and the monitoring station. The traffic intensity at the urban site (13 000 vehicles/day) was less than half than at the monitoring station (30 000 vehicles/day), while the mean particle number concentration was more than twice as high at the urban site. This is most likely related to the site characteristics and the position of the monitor. The urban site is characterised as a street canyon and the monitor was positioned directly next to the road. The monitoring station is an urban background site where sampling occurred 30 m from the main road.

The maximum to minimum ratio on week days was higher in October (5.7) than in February (4.2) and higher in the weekend (7.0-7.2) than on week days. The mean daily TNC values were generally lower in the weekends than on week days (ratio of 0.63-0.96), except for the urban and ring road sites in February (Table 10).

### 3.4.2 Diurnal variation in total particle number concentration

Figure 16 shows the hourly average diurnal variability in TNC (EPC) on week days at the monitoring station. For the February campaign an additional distinction was made between the first two weeks and last two weeks in order to compare with the EPC data of the other sites. The error bars represent the standard deviations of the hourly means.

On week days a clear diurnal pattern in TNC at the monitoring station was observed, with peaks in the morning (07:00 - 10:00) and evening (17:00 - 20:00). The concentrations were higher in October than in February, but the large standard deviations show that there is a meaningful day-to-day variation as well. During the February campaign, the concentrations were higher in week 1-2 than in week 3-4.

Figure 17 compares the diurnal variation in TNC (EPC) between the monitoring station, the public parc and the suburban site. For the February campaign only week 3-4 are shown. TNC at the three sites followed the same trend. Morning and evening rush hours occurred at the same times at the sites. In February, the public parc site and monitoring station showed similar concentration levels during the day, and the lowest concentrations were measured at the suburban site. In October, TNC at the monitoring station was higher than in February, and the rush hour peaks were also more pronounced, with highest values of about 25 000 particles/cm<sup>3</sup> from 08:00-09:00. For the public parc and also for the suburban site, similar TNC levels and diurnal patterns were observed during the two campaigns.

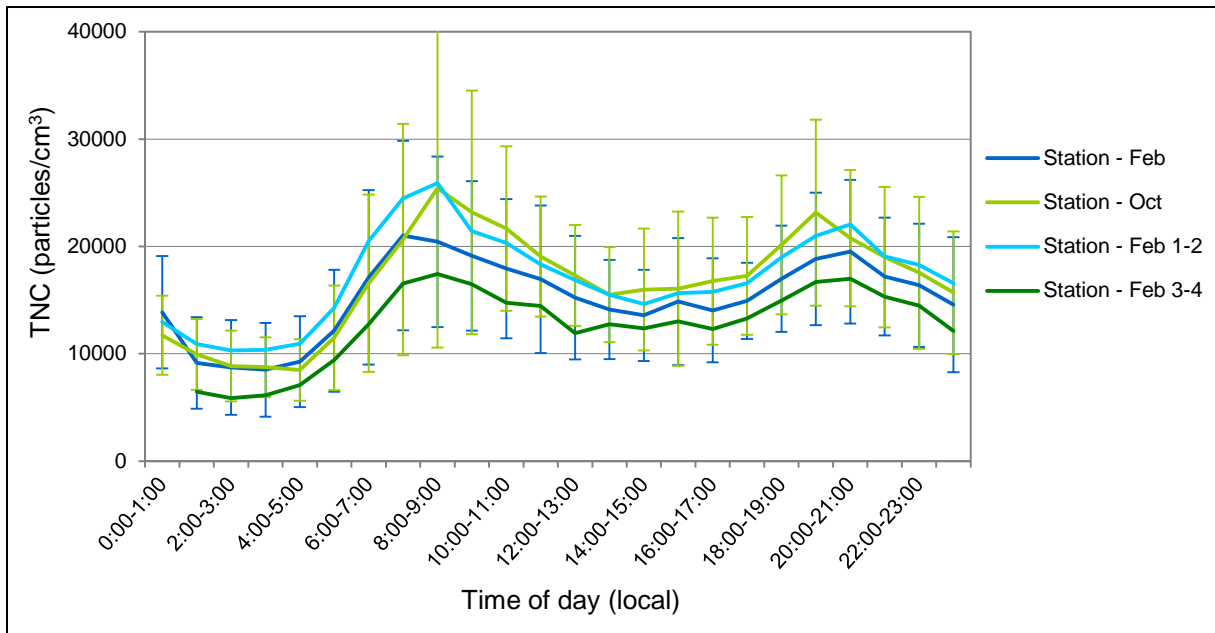


Figure 16: Hourly average total number concentration (TNC) at the monitoring station on week days of the February and October campaigns and during week 1-2 and week 3-4 of the February campaign. Standard deviations are given for the February and October campaigns

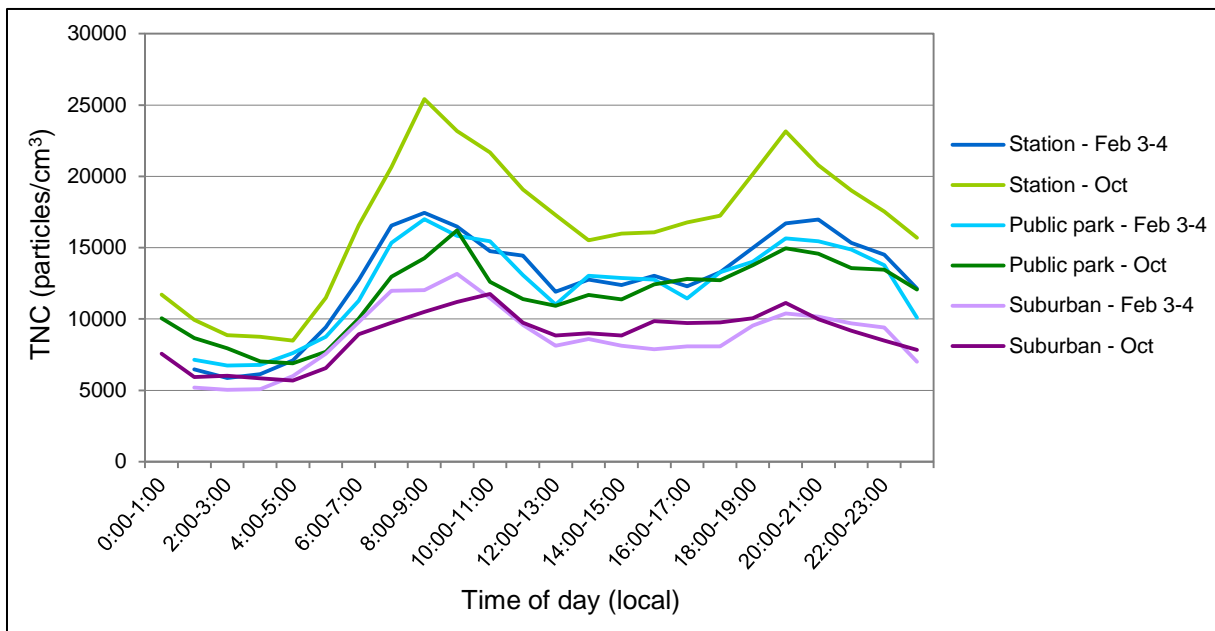


Figure 17: Hourly average total number concentration (TNC) at the monitoring station, public parc and suburban site on week days of the February and October campaigns

Figure 18 compares the diurnal variation in TNC (EPC) between the monitoring station, the urban site and the ring road site. For the February campaign only week 1-2 are shown. The figure shows that the morning rush hour started earlier at the ring road site than at the urban site and monitoring station. The highest mean hourly TNC values of about 53 000 (February) and 62 000 particles/cm<sup>3</sup> (October) were measured in the morning at the ring road site.

The urban site (February only) had also high number concentrations during morning rush hours on week days, but in contrast to the other sites the peak was higher in the evening than in the morning. This is in line with the diurnal pattern for the traffic intensity, as the evening traffic peak at the urban site was meaningfully higher than the morning peak (Figure 11).

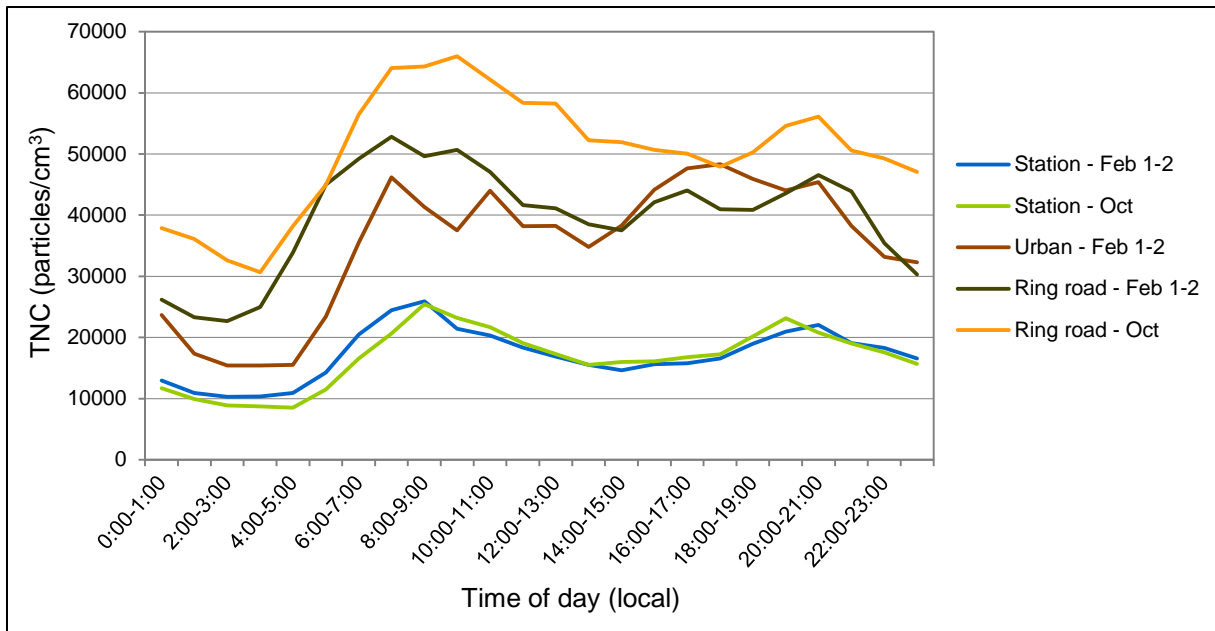


Figure 18: Hourly average total number concentration (TNC) at the monitoring station, urban site and ring road site on week days of the February and October campaigns

Figure 19 to Figure 21 show the hourly diurnal patterns in TNC during the weekends. In comparison with the week days, the TNC was generally lower and less variable through the day in the weekends. Furthermore, the morning peaks in the weekends appeared in general later than on week days. It should be noted that the data sets for the weekends were rather small (only 4 to 8 values per hour).

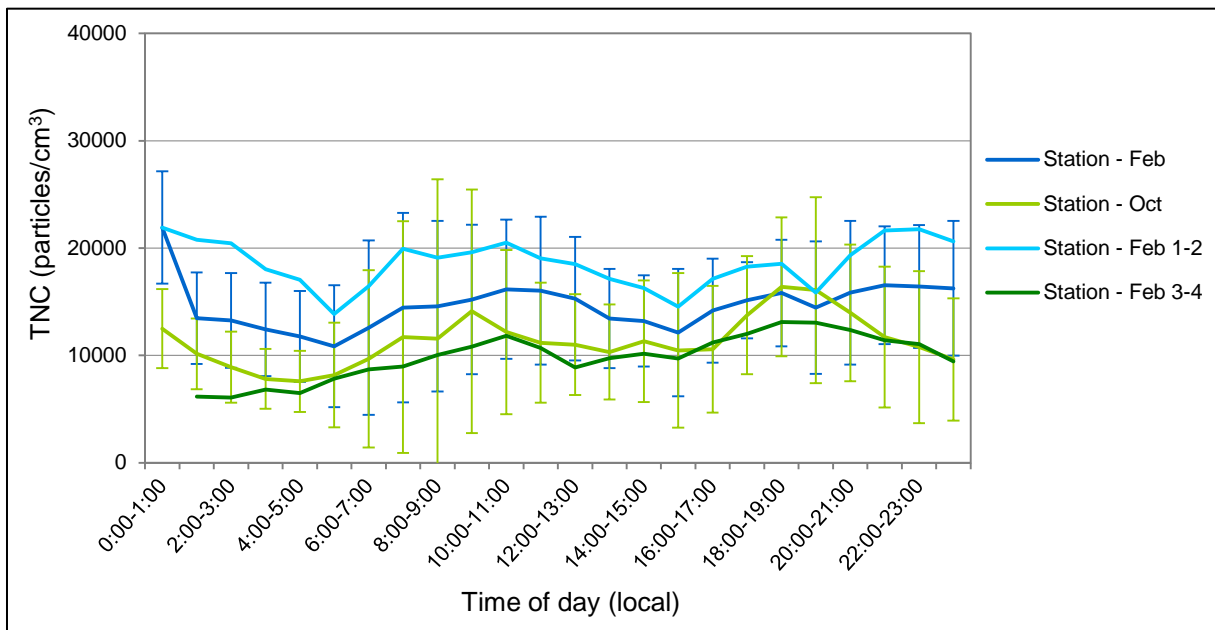


Figure 19: Hourly average total number concentration (TNC) at the monitoring station in the weekends of the February and October campaigns and week 1-2 and week 3-4 of the February campaign. Standard deviations are given for the February and October campaigns

Figure 19 compares the different periods for the monitoring station. In the weekends, TNC at the monitoring station was lower during the October campaign than in February, in contrast to the week days (Figure 16, see also Table 10). The particle concentration in the weekends of week 1-2 and 3-4 of the February campaign were higher and lower, respectively than for the total February campaign, which is in line with the observations for week days. The hourly standard deviations in the weekends were larger than on week days, probably due to combining Saturdays and Sundays.

Figure 20 compares the diurnal variation in TNC in the weekend between the monitoring station, the public parc and the suburban site. The relative order of the sites and periods was similar on weekend days (Figure 20) as on week days (Figure 17). In the weekends, the particle number concentrations at these sites reached the highest values in the afternoon and evening.

Figure 21 compares the diurnal variation in TNC in the weekend between the monitoring station, the urban site and the ring road site. Throughout the day, particle number concentrations at the urban site were similar to the concentrations at the ring road and much higher than at the monitoring station. At the urban site, high values were observed from noon until night. At the ring road site the TNC peaked at noon in February, while in October there were peaks in the late morning and in the evening.

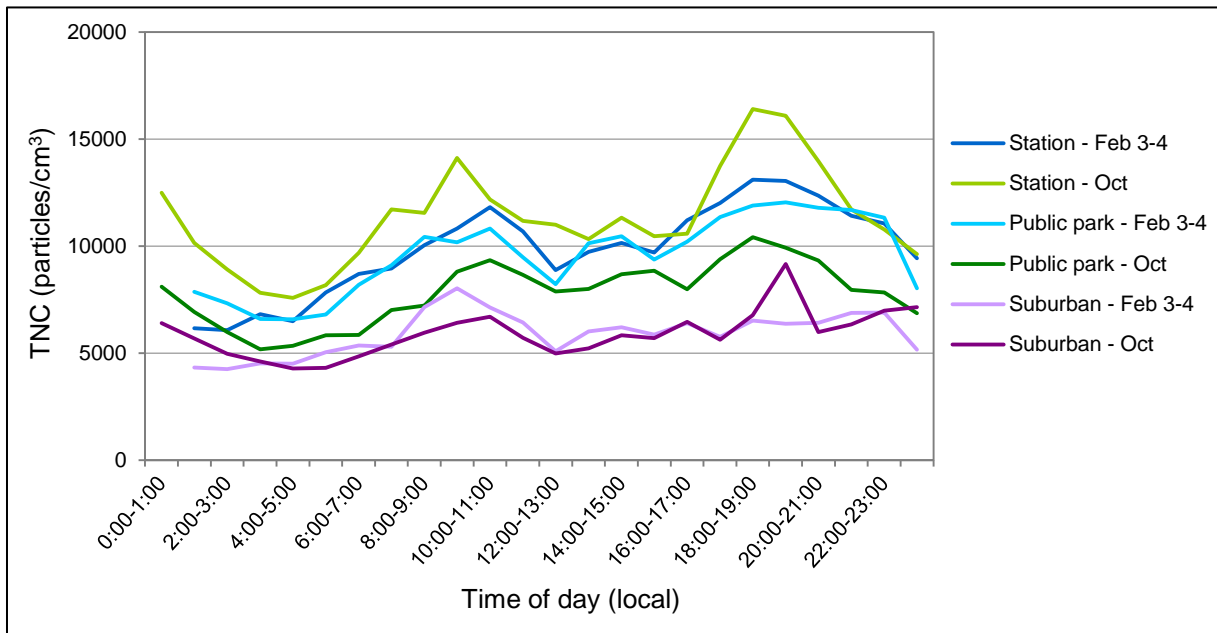


Figure 20: Hourly average total number concentration (TNC) at the monitoring station, public parc and suburban site in the weekends of the February and October campaigns

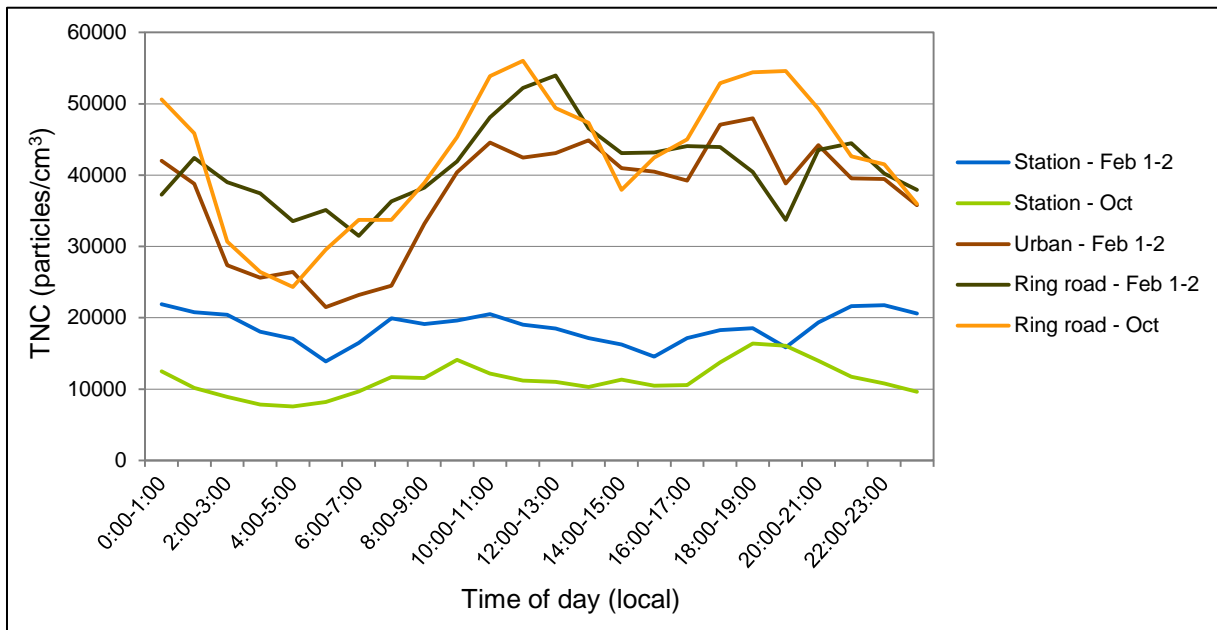


Figure 21: Hourly average total number concentration (TNC) at the monitoring station, urban site and ring road site in the weekends of the February and October campaigns

### 3.4.3 Correlation of total particle number concentration between sites

In this section we investigate the relation between the total number concentration of particles  $<1 \mu\text{m}$  (TNC) at a given site with the concentration at the monitoring station. As explained in section 3.3, two types of regressions were calculated. All results are summarized in Table 11. It should be noted that the  $R^2$  value of a regression through the origin ( $y = ax$ ) is by default higher than the  $R^2$  value of a normal regression ( $y = ax + b$ ), but cannot be interpreted in the usual way.

Table 11: Slope ( $a$ ), intercept ( $b$ ) and coefficient of determination ( $R^2$ ) of single linear regressions ( $y = ax + b$ ) and regressions forced through the origin ( $y = ax$ ) between total particle number concentration (TNC) at the monitoring station and the other sites during the February and October campaigns. The Pearson correlation coefficient ( $r$ ) is mentioned for the regressions through the origin

Site	Period	$y = ax + b$				$y = ax$	
		$a$	$b$	$R^2$	$r$	$a$	$R^2$
Public parc	February 3-4	0.780	2281	0.69	0.83	0.943	0.95
	October	0.470	3809	0.71	0.84	0.651	0.93
Suburban	February 3-4	0.653	116	0.72	0.85	0.661	0.95
	October	0.316	2785	0.50	0.70	0.449	0.89
Urban	February 1-2	1.462	10099	0.50	0.71	1.944	0.91
Ring	February 1-2	1.688	10487	0.41	0.64	2.188	0.87
Ring	October	1.412	27324	0.41	0.64	2.752	0.86

Figure 22 to Figure 25 only show the lines of the regressions forced through the origin because these are considered to be most representative to evaluate differences in temporal variability between the sites and periods.

Figure 22 is a scatter plot of the half-hourly TNC at the public parc in relation to the monitoring station. There was a strong temporal correlation between the two sites ( $r > 0.83$  for the regression with an intercept). During week 3-4 of the February campaign, the slope of the regression through the origin was 0.94, so that the TNC at the public parc site was on average 6% lower than at the monitoring station. In October, TNC at the parc site was about 35% lower than at the station. This indicates that the relationship in TNC between the two sites varied over time.

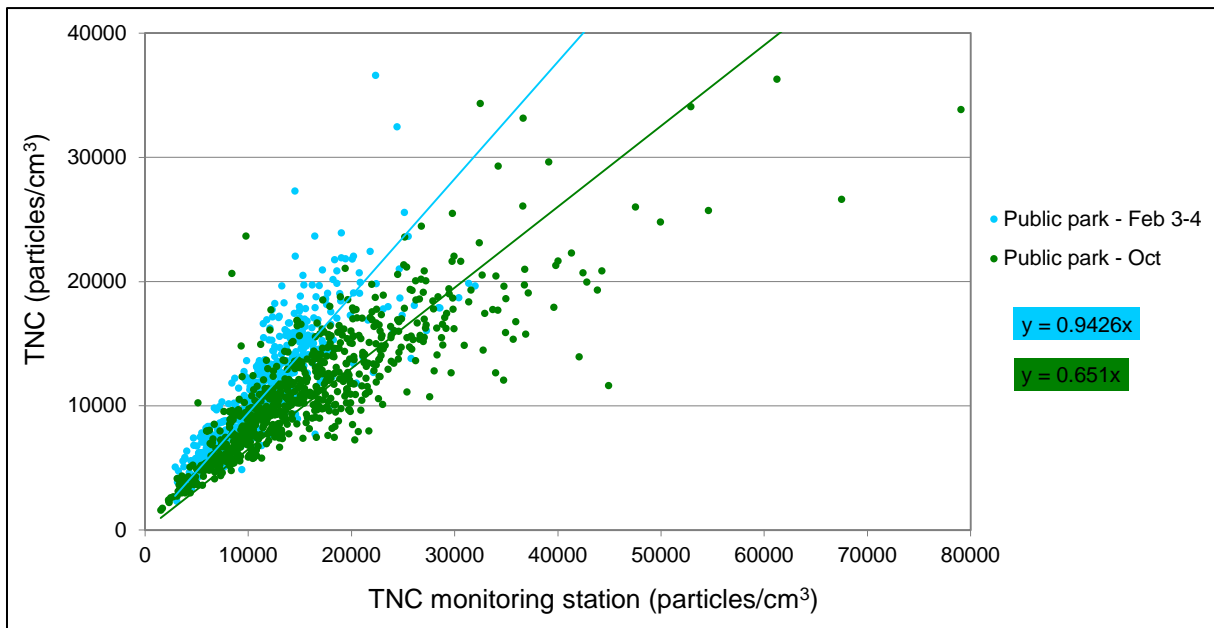


Figure 22: Half-hourly total number concentration (TNC) at the public parc site in relation to TNC at the monitoring station during the February and October campaigns

It is relevant to repeat that the average TNC at the parc site was similar in both measuring campaigns, (11 602 particles/cm<sup>3</sup> in week 3-4 of the February campaign and 10 569 particles/cm<sup>3</sup> in October). At the monitoring station, in contrast, the TNC in October (15 170 particles/cm<sup>3</sup>) was higher than in week 3-4 of the February campaign (11 937 particles/cm<sup>3</sup>) (see also Table 10 and Figure 17).

Figure 23 relates the TNC at the suburban site to the monitoring station. Compared to the monitoring station, TNC at the suburban site was about two thirds in February (slope  $a = 0.66$ ) and less than half in October ( $a = 0.45$ ). In February, the intercept of the normal regression was small (Table 11) and not significantly different from zero ( $P = 0.62$ ). The temporal correlation for TNC between the public parc site and the monitoring station was lower in October ( $r = 0.70$ ) than in February ( $r = 0.85$ ).

At the urban site (Figure 24), the particle number concentration was almost twice as high compared to the monitoring station ( $a = 1.94$ ) in February week 1-2. The temporal correlation was moderate ( $r = 0.71$ ).

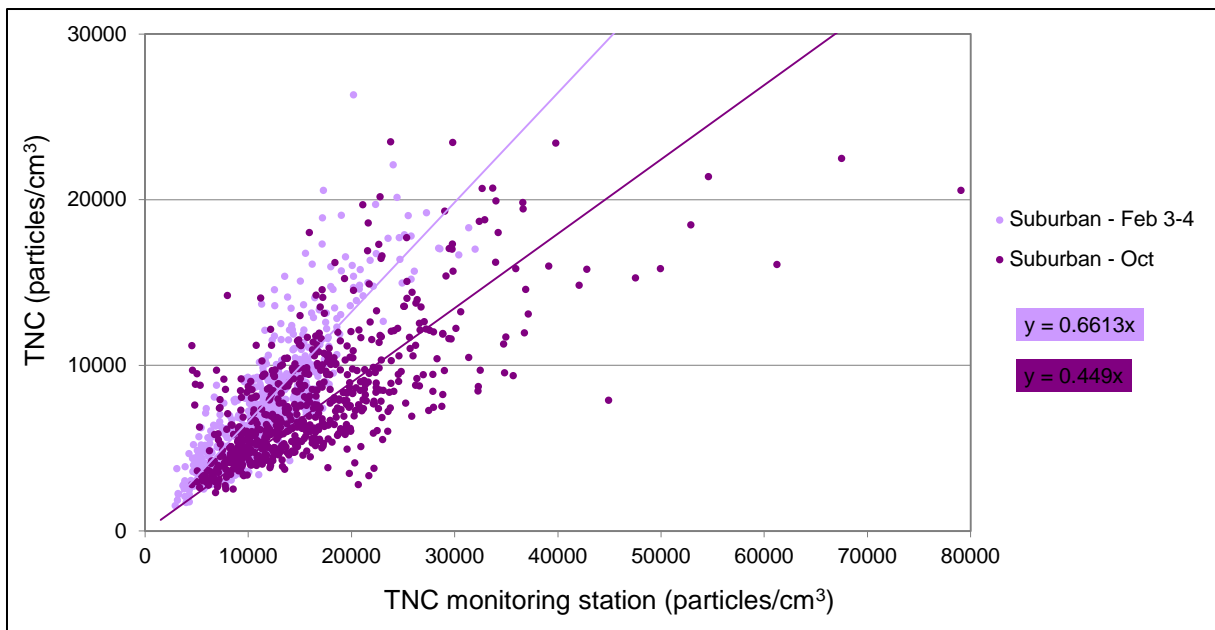


Figure 23: Half-hourly total number concentration (TNC) at the suburban site in relation to TNC at the monitoring station during the February and October campaigns

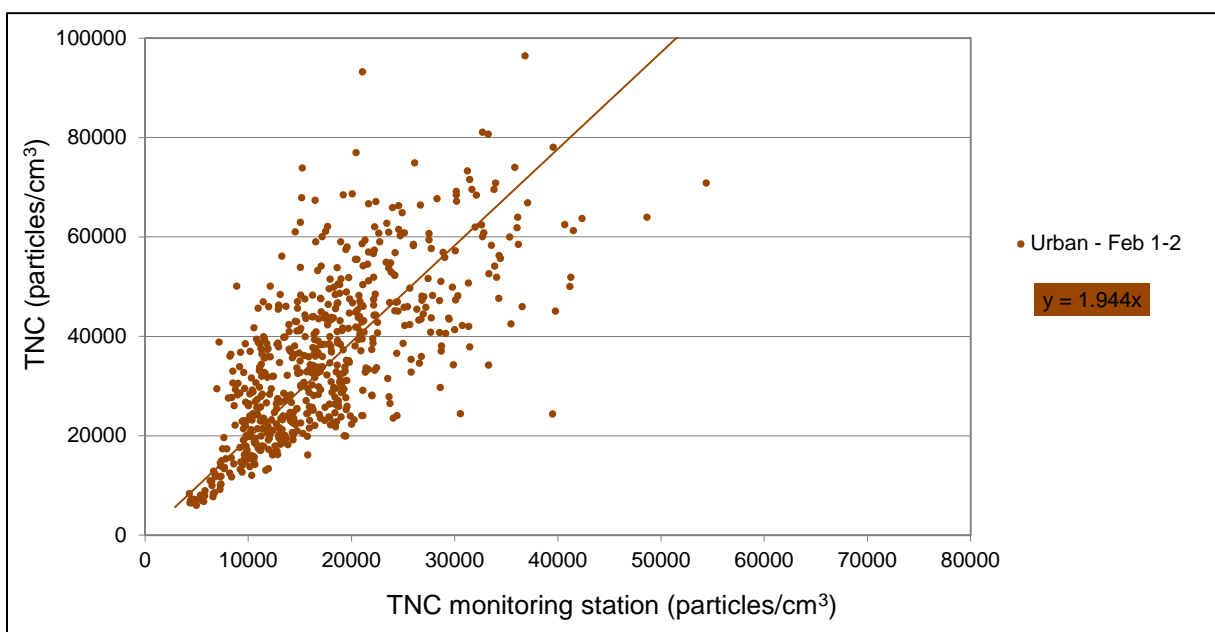


Figure 24: Half-hourly total number concentration (TNC) at the urban site in relation to TNC at the monitoring station during the February campaign



At the ring road site (Figure 25), the number concentration was on average two to three times higher than at the monitoring station, as the regression slopes were 2.2 in February and 2.7 in October. The temporal correlation with the monitoring station ( $r = 0.64$ ) was lower than for the other sites ( $r > 0.70$ ).

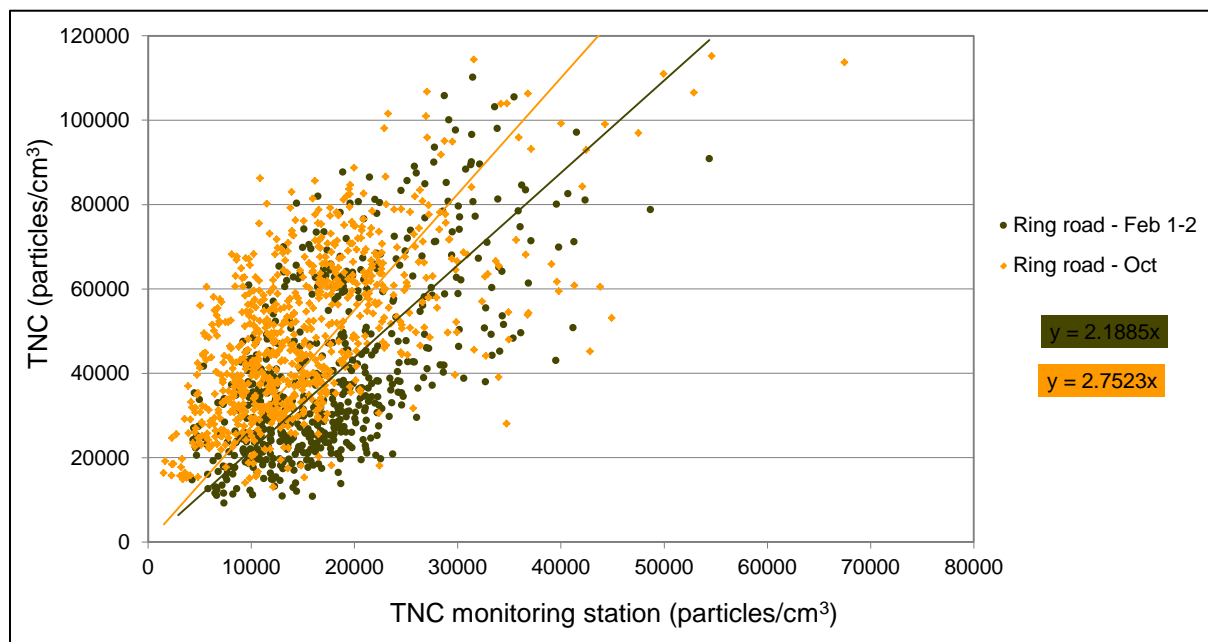


Figure 25: Half-hourly total number concentration (TNC) at the ring road site in relation to TNC at the monitoring station during the February and October campaigns

Until now we used Pearson correlation coefficients to study the temporal correlation of the total particle number concentration between two sites. To characterize the spatial variability between site pairs the coefficient of divergence (COD) was used. A COD value  $> 0.2$  was considered as heterogeneous.

The results of this COD analysis were that for the TNC in February the pair of sites public parc vs. monitoring station was most homogeneous, with an overall COD value of 0.09 (Table 12), followed by the pair monitoring station vs. suburban site. The pair urban site vs. monitoring station was moderately spatially heterogeneous and the pair monitoring station vs. ring road was most heterogeneous.

In October the COD values for the TNC were higher than in February for all sites (Table 12), indicating a higher spatial divergence in October.

Table 12: Coefficient of divergence (COD) between the hourly total particle number concentration (TNC) at the monitoring station and the other sites during the February and October campaigns

Site pair	February	October
Public parc vs. monitoring station	0.09 <sup>a</sup>	0.19
Suburban vs. monitoring station	0.24 <sup>a</sup>	0.37
Urban vs. monitoring station	0.35 <sup>b</sup>	-
Ring vs. monitoring station	0.40 <sup>b</sup>	0.55

<sup>a</sup> From 18/02 to 04/03/2013 only

<sup>b</sup> From 04/02 to 18/02/2013 only

### 3.4.4 Particle size distribution

In February at two of the five sites (urban site, ring road site) UFP monitors were used to measure the particle size distribution. This monitor gives no information on the number of particles  $< 20$  nm. To be able to compare the size distribution between all sites and periods the SMPS data were recalculated to the size ranges of the UFP Monitor.

Figure 26 shows the mean size distribution at all five sites on week days. The mean values per size class were based on the mean hourly data per class.

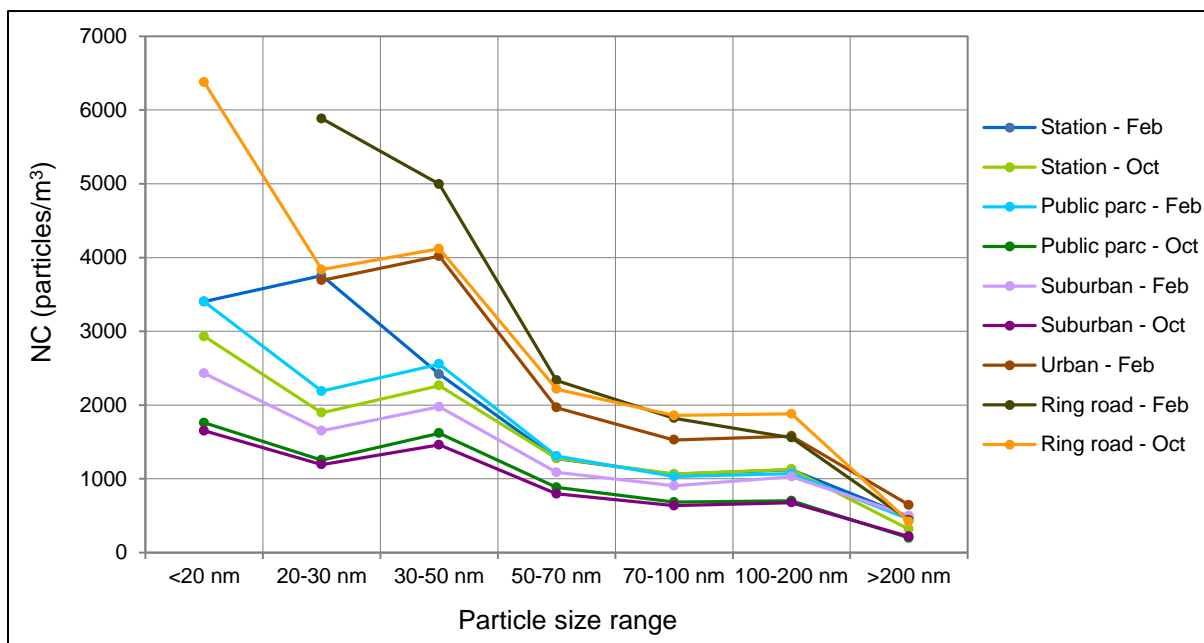


Figure 26: Mean particle number concentration (NC) per size class and site on week days of the February and October campaigns

In general the mean size distribution had a similar trend for all sites and periods: the highest particle number concentrations were measured in the smallest size range (<20 nm), followed by a relative decrease for 20-30 nm particles, a second peak in the 30-50 nm size range and a decrease in number concentration from 50 to 200 nm. In the >200 nm size class only low number concentrations occurred.

The similarity in the size distribution between the sites was also clear when we expressed the relative fraction of particles for each size class (for sites with an SMPS). On week days, on average 26% of the TNC consisted of particles <20 nm. The size classes 20-30 and 30-50 nm each accounted for 20-21%. The classes 50-70, 70-100 and 100-200 nm contributed 9-11%, and only 4% of the particles were in the 200-1000 nm range. Consequently, on week days UFP accounted for 86% of the TNC.

There were two exceptions to this general size distribution trend. First, for the monitoring station in February, the highest number concentrations were found in the 20-30 nm size range instead of in the <20 nm range, and the concentrations decreased from 20 to 200 nm. Second, also for the ring road site in February the highest number concentrations were found in the 20-30 nm size range, but for this site and period we have no information on the <20 nm size class.

In February particles <20 nm were not measured at the ring road and urban site but based on the site characteristics and the other measurements, it cannot be ruled out that in February also at these sites high concentrations occurred in the <20 nm range. High particle number concentrations in the small size ranges are attributed to fresh particle supply from nearby roads. When sampling occurs further away from the traffic or in a street canyon more 'aged', coagulated and bigger particles are expected. In this respect, highest concentrations in the smallest size range seems more likely at the ring road site (cf. the results for October) and less likely at the urban site.

Figure 27 shows the mean size distribution during the weekends. The main difference with the size distribution on week days was the relatively lower concentration of 20-30 nm particles compared to 30-50 nm particles at the ring road and urban site in February. Furthermore, during the weekends the concentration of particles <20 nm was generally relatively lower than on week days: concentrations of <20 nm and 30-50 nm particles were of similar magnitude, except for the monitoring station in February and for the ring road site in October (no information on <20 nm available for February). This finding suggests that relatively less fresh (small) particles and relatively more aged (larger) particles

were present during the weekend than on week days. It is unknown if this is caused by less (heavy) traffic in the weekend or by another reason.

For the sites with an SMPS, in the weekends on the average 22% of the TNC consisted of particles <20 nm and 20% of particles in the 30-50 nm size range. Particles of 20-30 nm accounted for 16%. The classes 50-70, 70-100 and 100-200 nm contributed 11-14%, and only 5% of the particles were in the 200-1000 nm size range. Consequently, during the weekends UFP contributed 81% of the TNC.

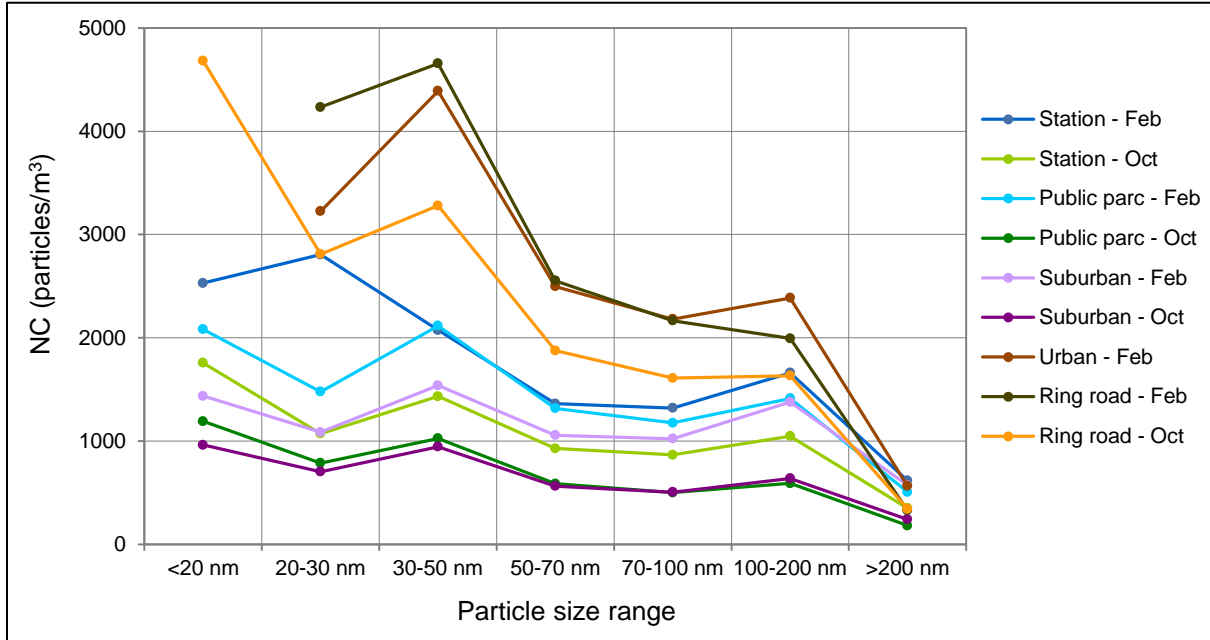


Figure 27: Mean particle number concentration (NC) per size class and site in the weekends of the February and October campaigns

To check how the particle number concentrations per size range were related between the monitoring station and the other sampling sites, Pearson correlation coefficients were calculated for each size range (30-min data), as shown in Figure 28.

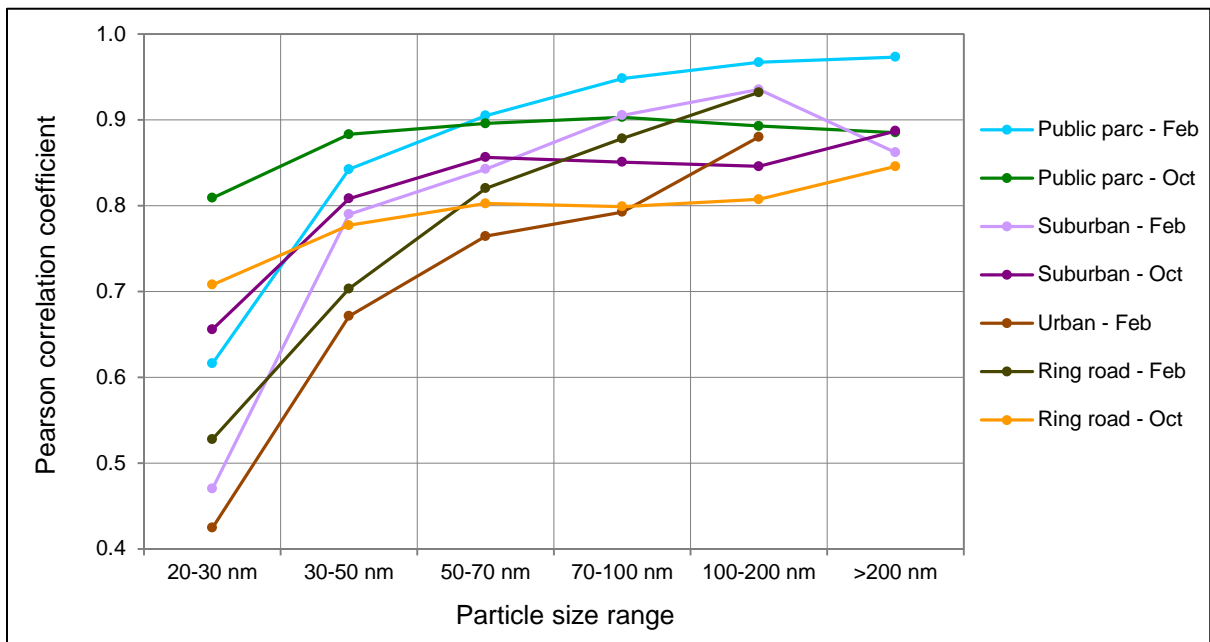


Figure 28: Pearson correlation coefficients between the half-hourly particle number concentrations at the monitoring station and at the other sites during the February and October campaigns. Note that the y-axis starts at a value of 0.4

Strong temporal correlations between the monitoring station and the four other sites were found for the number concentrations in the size classes from 50-70 nm to 100-200 nm ( $r = 0.76 - 0.86$  in February and  $r = 0.80 - 0.85$  in October). The correlations between the number concentrations in the 20-30 nm and 30-50 nm size classes were lower, suggesting that these particles were more locally influenced, so that the temporal variability for these fractions was higher. The correlation values for the 20-30 nm and 30-50 nm ranges were higher in October than in February for all the considered site pairs (public parc vs. monitoring station; suburban site vs. monitoring station; ring road site vs. monitoring station). This was particularly pronounced for the 20-30 nm size range.

In February very low particle numbers were found at the ring road and urban site for particles larger than 200 nm. This was caused by the problems of the UFP monitors to reliably measure these bigger particles, as was shown by the instrument comparison. Therefore two correlation values for the >200 nm class have been omitted from Figure 28 ( $r = 0.49$  for the urban site in February,  $r = 0.52$  for the ring road site in February).

To determine the spatial homogeneity between the VMM monitoring station and the other sites, coefficient of divergence (COD) values were also calculated for each of the six particle size ranges, as shown in Figure 29.

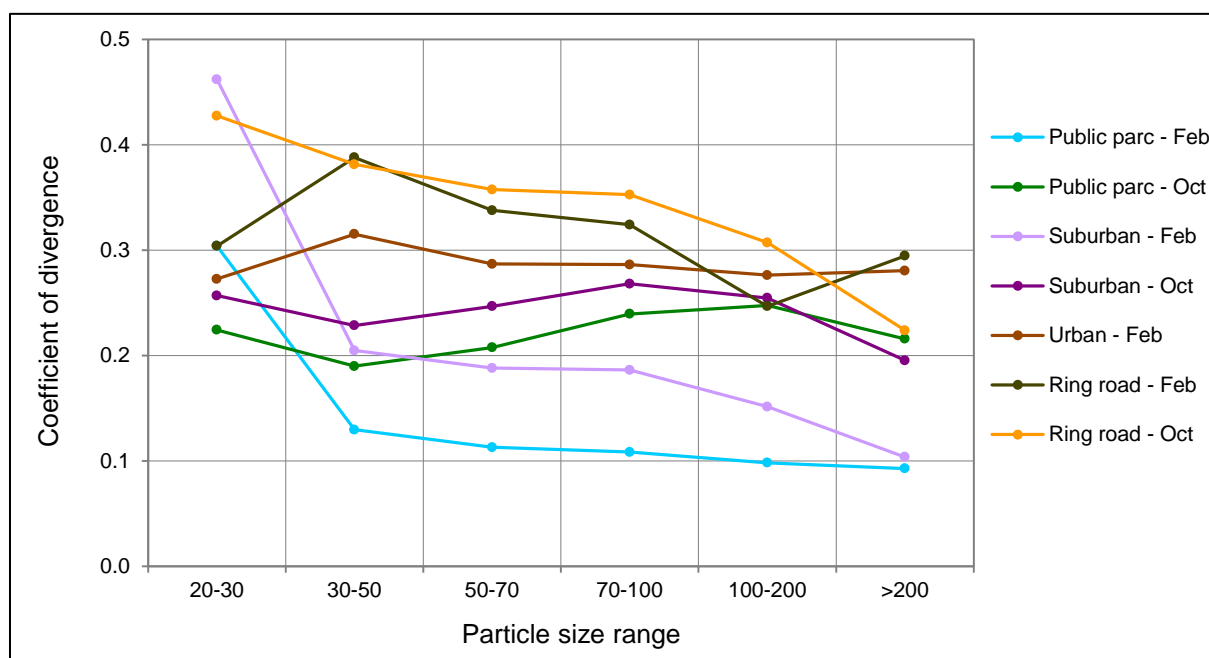


Figure 29: Coefficient of divergence (COD) between the hourly particle number concentrations at the monitoring station and at the other sites during the February and October campaigns

In February a large spatial variability was found for particles in the 20-30 nm size range for all pairs of sites (COD values > 0.2). Comparing the monitoring station with the public parc or suburban site revealed lower spatial variability for particles >30 nm, with COD values smaller than 0.2. On the other hand a moderate heterogeneity was found comparing the urban or ring road site with the monitoring station, with COD values larger than 0.2 for all ranges. So in general a higher spatial divergence was found for smaller particles and an increasing spatial homogeneity for bigger particles.

In October the COD values for all size ranges were larger than in February. Comparing the monitoring station with the public parc or suburban site showed no great changes in COD for increasing particle sizes, in contrast to February. For the ring road site, the highest spatial divergence was found for 30-50 nm particles and an increasing uniform spatial homogeneity for bigger particles.

### 3.5 Particle concentration in relation to other air quality variables

#### 3.5.1 Particulate matter (PM<sub>10</sub>)

PM<sub>10</sub> concentrations with high time resolution (30 min or less) were only measured at the monitoring station during both campaigns and at the ring road site and suburban site in October. At the public parc site and monitoring station, daily gravimetric data were available in October. The PM<sub>10</sub> data for the ring road location were limited and were therefore not taken into account for further analyses. Information about the relationship between the daily PM<sub>10</sub> concentrations with the reference method and the continuous Grimm monitors is given in the report about the October campaign.

The October report presented correlations between PM<sub>10</sub>, which is the mass concentration of particles <10 µm, and the total number concentration of particles <1 µm (TNC, obtained by SMPS) at the half-hourly level. The correlation between 30-min PM<sub>10</sub> and TNC was moderate for the monitoring station and lower for the suburban site (Table 13). Here we also show the relationship between PM<sub>10</sub> and TNC at the daily level. The Pearson correlation coefficients for daily averaged concentrations were clearly higher than for the half-hourly data, with correlations of 0.66 (suburban) to 0.84 (public parc) (Table 13).

Table 13: Pearson correlation ( $r$ ) and coefficient of determination ( $R^2$ ) between half-hourly/daily PM<sub>10</sub> concentration and total particle number concentration during the October campaign

Site	Half-hourly		Daily	
	$r$	$R^2$	$r$	$R^2$
Monitoring station	0.42	0.18	0.71	0.50
Public parc	-	-	0.84	0.71
Suburban	0.25	0.06	0.66	0.43

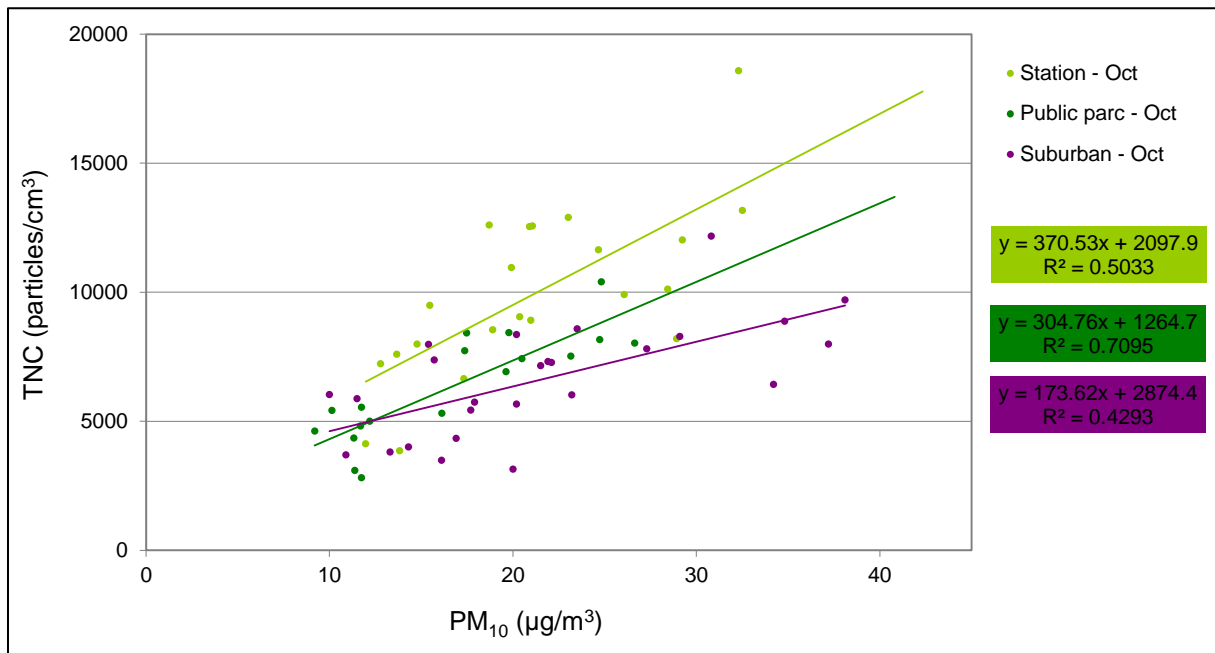


Figure 30: Daily total number concentration in relation to the PM<sub>10</sub> concentration for three sites during the October campaign

Figure 30 shows the corresponding single linear regressions between daily TNC and PM<sub>10</sub>. It should be noted that these results were obtained for a small data set (18 to 26 pairs of daily values, depending on the site). Nevertheless, the results indicate that in October 2013 for these sites there was a positive but site-specific relation between PM<sub>10</sub> and TNC.

The regression slope was similar for the monitoring station and the parc site, and lower for the suburban site. In other words, a given increase in PM<sub>10</sub> was accompanied by a lower increase in TNC at the suburban site than at the more traffic-exposed monitoring station and public parc site. The intercept of the regression was higher for the monitoring station than for the parc site. This implies that for a given PM<sub>10</sub> level the TNC was higher at the station than at the parc site.

### 3.5.2 Black carbon

The report about the October campaign showed the diurnal variation in hourly BC concentrations on week days and in the weekend. On week days there was a diurnal variation at all sites, with higher values during the day and the rush hours in particular. BC concentrations were highest at the ring road site and lowest at the suburban site. At the monitoring station BC decreased significantly between the morning and evening rush hour, which was not the case for the ring road site. During the weekends the diurnal variation was less pronounced and the highest hourly concentrations were found during the evening hours.

The BC concentration was correlated between the four sites of the October campaign. The correlation coefficient (30-min data) with the monitoring station was 0.81 for the public parc, 0.44 for the suburban site and 0.50 for the ring road site.

The report about the October campaign showed relationships between the BC concentration and the total number concentration (TNC). Here we also investigated the relationship between BC and the different particle size classes and included the available BC measurements of the February campaign.

Table 14 shows that TNC correlated was moderately strong (suburban site,  $r = 0.73$ ) to strongly correlated (monitoring station in October,  $r = 0.87$ ) to the BC concentration. This confirms previous studies where particle number concentrations tended to correlate well with traffic by-products such as BC. Also the number of particles per size class was correlated with BC, and it was clear that the correlation coefficient increases for larger particle classes. For all the sites, the maximum correlation value between BC and the particle size classes used was found for 100-200 nm particles.

Table 14: Pearson correlation between half-hourly black carbon concentration 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 <sup>a</sup>	R <sup>2</sup> <sub>max</sub> <sup>b</sup>
Street site 10 m	Feb	0.46	0.55	0.67	0.77	0.86	<b>0.90</b>	0.85	0.78	0.81
Mon. station	Feb	0.37	0.37	0.61	0.75	0.84	<b>0.88</b>	0.84	0.71	0.78
	Oct	0.69	0.77	0.82	0.89	0.92	<b>0.93</b>	0.81	0.87	0.87
Public parc	Oct	0.28	0.51	0.68	0.79	0.85	<b>0.84</b>	0.77	0.68	0.71
Suburban	Oct	0.27	0.43	0.55	0.65	0.69	<b>0.76</b>	0.74	0.73	0.54
Ring	Oct	0.72	0.68	0.70	0.81	0.84	<b>0.86</b>	0.79	0.78	0.74

<sup>a</sup> TNC is the total number concentration according to EPP/CPC measurements

<sup>b</sup> The maximum correlation per row is given in bold and expressed as R<sup>2</sup><sub>max</sub> in the last column

Figure 31 shows single linear regressions between BC and the number concentration of 100-200 nm particles (NC100-200). In February, a similar relationship between these variables was found for the two sites with BC data near the Plantin en Moretuslei: the monitoring station at 30 m from the road and the street site at 10 m from the road. However, in October, the relationship between BC and NC100-200 for the monitoring station had a lower slope than in February. In other words, at this site an increase in BC in October was accompanied by a lower increase in NC100-200 than in February.

Figure 32 shows single linear regressions between BC and NC100-200 per sampling site in October. The regression line of the monitoring station, shown in Figure 31, was repeated for the ease of comparison. Figure 32 shows that the relationship between BC and NC100-200 was site-specific. Interestingly, the slope of the regression per site appeared to increase with increasing exposure to traffic at the site: suburban site < public parc < monitoring station < ring road site.

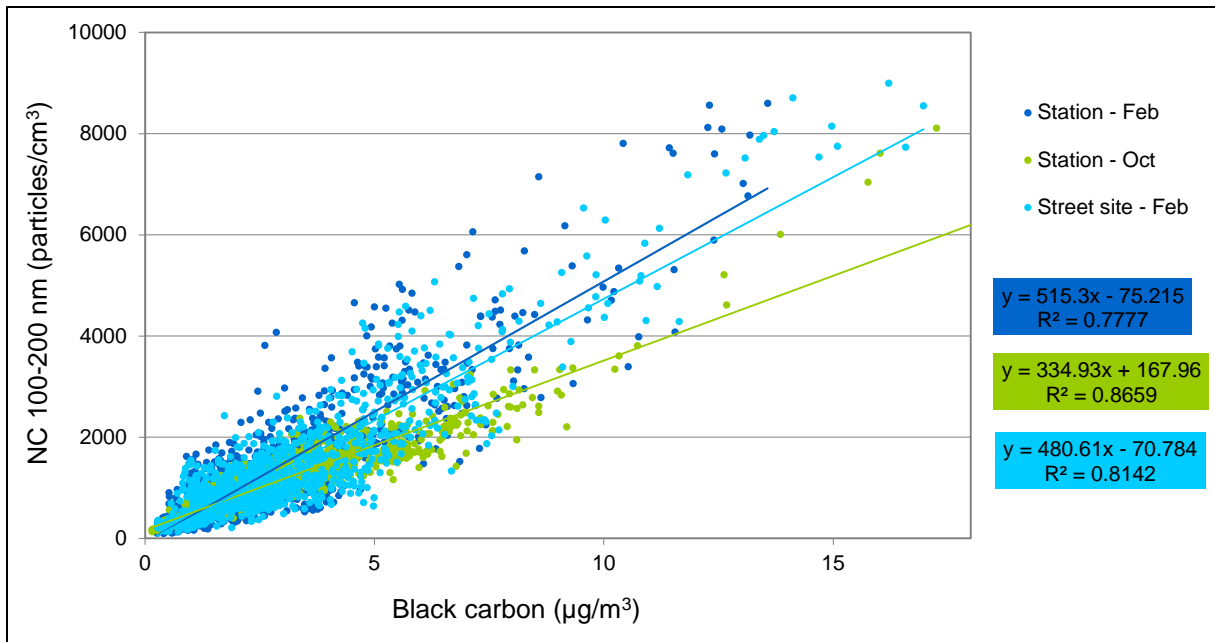


Figure 31: Half-hourly 100-200 nm particle number concentration (NC) in relation to the black carbon concentration at the monitoring station and the street site (10 m, February only)

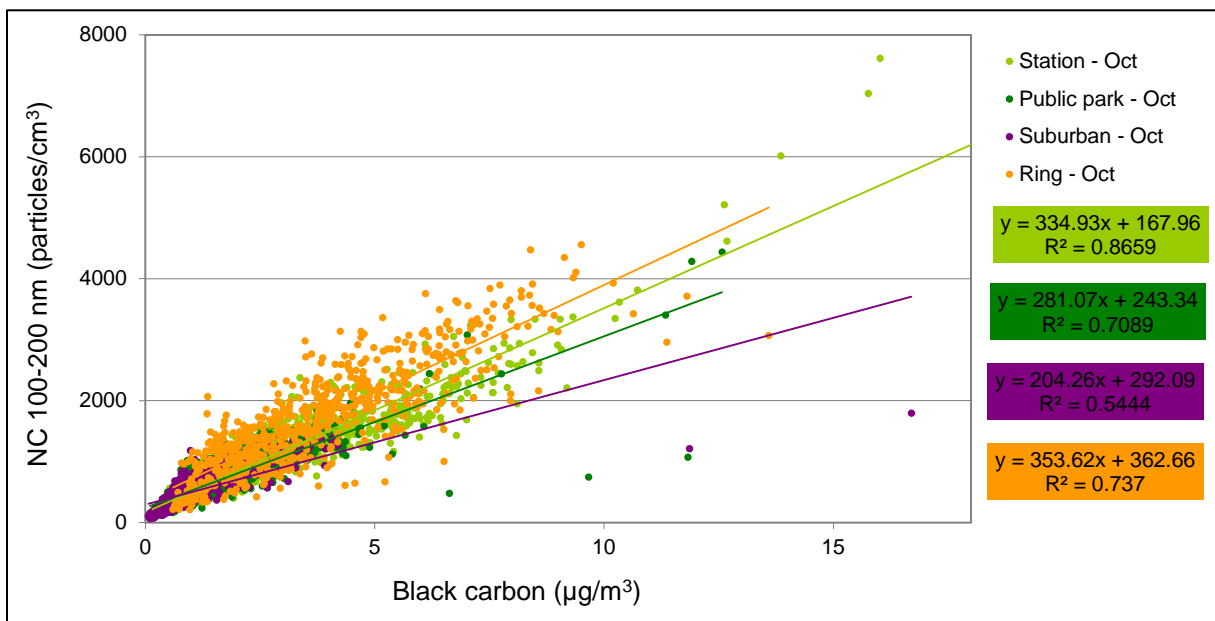


Figure 32: Half-hourly 100-200 nm particle number concentration (NC) in relation to the black carbon concentration per site during the October campaign

### 3.5.3 Nitrogen dioxide ( $\text{NO}_2$ )

The reports about the intra-urban UFP variation in February and October presented correlations between weekly  $\text{NO}_2$  concentrations and the particle number concentration in different size classes. For the gradient study in February, the correlation between  $\text{NO}_2$  and the total TNC per distance to the main road was given.

Here we first present a correlation analysis where the sites were subdivided into three sets. Figure 33 shows that the weekly TNC was strongly related with the  $\text{NO}_2$  concentration. In February, TNC was related in a similar way to  $\text{NO}_2$  for the sites used to study the intra-urban variability as for the sites of the gradient study. For October, in contrast, the slope of the linear regression between TNC and  $\text{NO}_2$  was lower than in February.

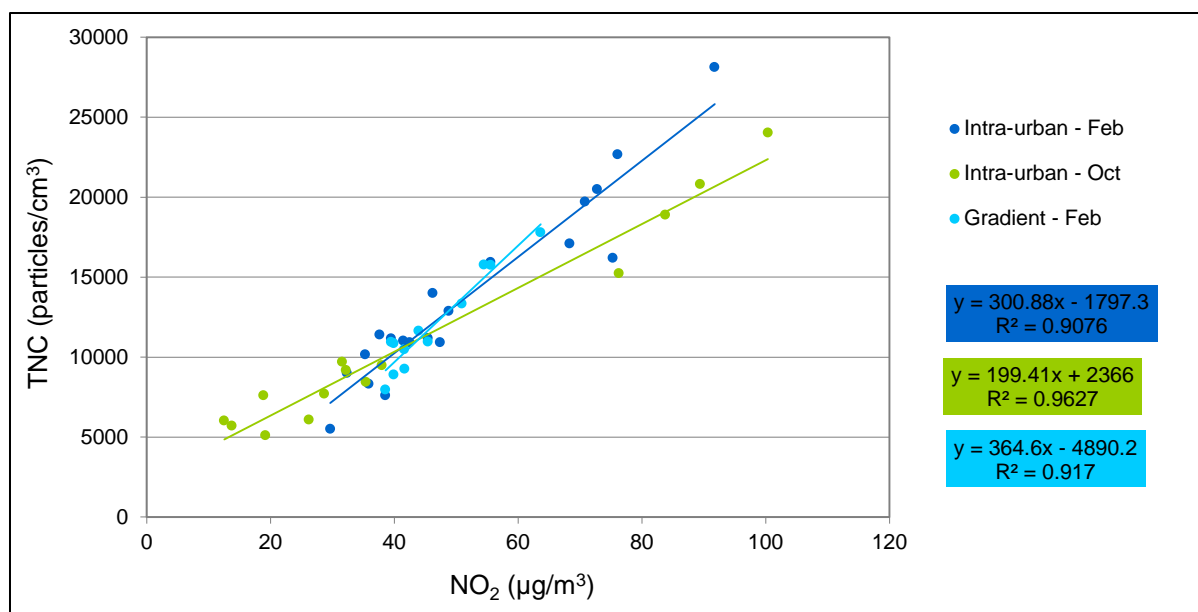


Figure 33: Weekly total particle number concentration (TNC) in relation to the  $\text{NO}_2$  concentration for the sites of (a) the intra-urban study in February and (b) October and (c) the gradient study in February

Then we repeated the correlation analysis per data set for the different particle size ranges. Table 15 summarizes the results of this analysis. The strongest correlations with  $\text{NO}_2$  were found for the particle number concentrations in the smallest size class (<20 nm). For all data sets the correlation with  $\text{NO}_2$  decreased for the larger particle size classes.

Table 15: Pearson correlation between weekly  $\text{NO}_2$  concentration and particle number concentration for different selections of sites

Study	Period	<20 nm	20-30 nm	30-50 nm	50-70 nm	70-100 nm	100-200 nm	>200 nm	TNC <sup>a</sup>
Gradient	Feb	0.90	0.94	0.93	0.94	0.91	0.89	0.65	0.96
Intra-urban	Feb	-	0.92	0.97	0.93	0.86	0.67	0.10	0.95
	Oct	0.97	0.97	0.97	0.96	0.95	0.93	0.80	0.98
Overall	Feb + Oct	0.94	0.83	0.89	0.88	0.84	0.72	0.29	0.94

<sup>a</sup> TNC is the total number concentration according to SMPS or EPC measurements

Figure 34 shows the relationships between  $\text{NO}_2$  and the particle numbers per size class for the sites of the October campaign. Figure 35 is a similar graph but now for all the available data in February and October.



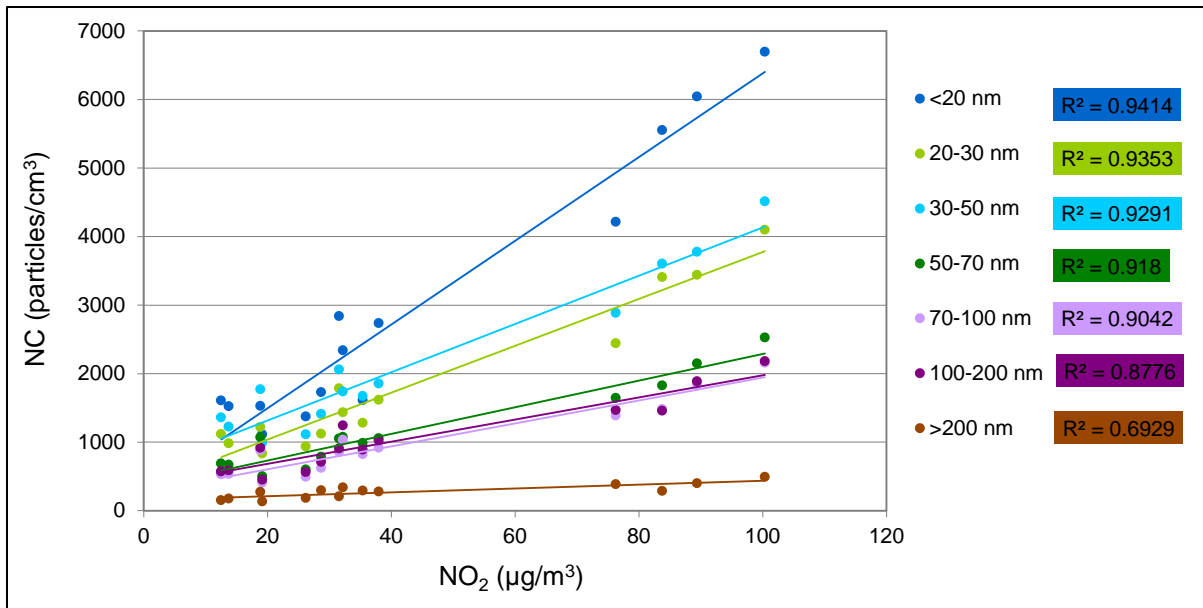


Figure 34: Weekly particle number concentration (NC) per class in relation to the  $\text{NO}_2$  concentration in October

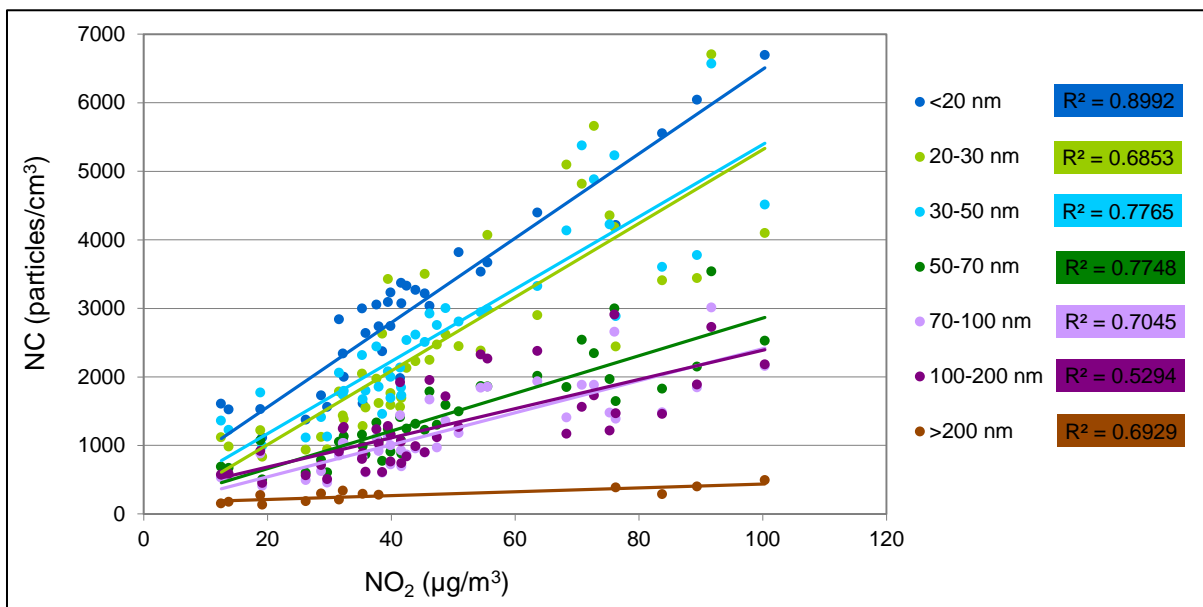


Figure 35: Weekly particle number concentration (NC) per class in relation to the  $\text{NO}_2$  concentration in February and October

### 3.6 Particle concentration in relation to traffic intensity

Finally the relationship between particle number concentrations and traffic volumes was investigated. As for the air quality variables, correlations were calculated both for the total particle number concentration and the number concentration of the six distinguished size classes. We used half-hourly data for all the sites in February and October.

Table 16 summarizes the Pearson correlation coefficients between particle concentrations and traffic intensities per site and period. Overall, the highest correlations with traffic were found for the smallest size classes (NC<20 nm or NC20-30 nm). From 50 nm on, the correlation was rather poor. The correlation between TNC and the traffic intensity (half-hourly level) varied from 0.22 to 0.60.

Table 16: 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 <sup>a</sup>	R <sup>2</sup> <sub>max</sub> <sup>b</sup>
Mon. station	Feb	<b>0.42</b>	0.42	0.21	0.09	0.01	-0.05	-0.07	0.22	0.18
	Oct	<b>0.51</b>	0.41	0.33	0.30	0.28	0.26	0.20	0.41	0.26
Public parc	Feb	<b>0.66</b>	0.62	0.55	0.46	0.39	0.29	-0.08	0.60	0.43
	Oct	<b>0.51</b>	0.30	0.17	0.13	0.11	0.10	0.07	0.30	0.26
Suburban	Feb	<b>0.44</b>	0.37	0.30	0.21	0.15	0.11	-0.14	0.34	0.20
	Oct	<b>0.40</b>	0.25	0.14	0.07	0.04	0.02	0.01	0.29	0.16
Urban	Feb	-	<b>0.33</b>	0.31	0.28	0.25	0.16	0.00	0.31	0.11
Ring	Feb	-	<b>0.43</b>	0.34	0.23	0.17	0.06	0.06	0.32	0.19
	Oct	0.39	<b>0.40</b>	0.42	0.46	0.47	0.47	0.39	0.44	0.16

<sup>a</sup> TNC is the total number concentration according to SMPS measurements

<sup>b</sup> The maximum correlation is given in bold and expressed as R<sup>2</sup><sub>max</sub> in the last column

For the monitoring station, the correlation between traffic intensity and the number of particles <20 nm was equal to 0.42 in February and 0.51 in October. The regression lines appeared to be rather similar during the two measuring periods (Figure 36).

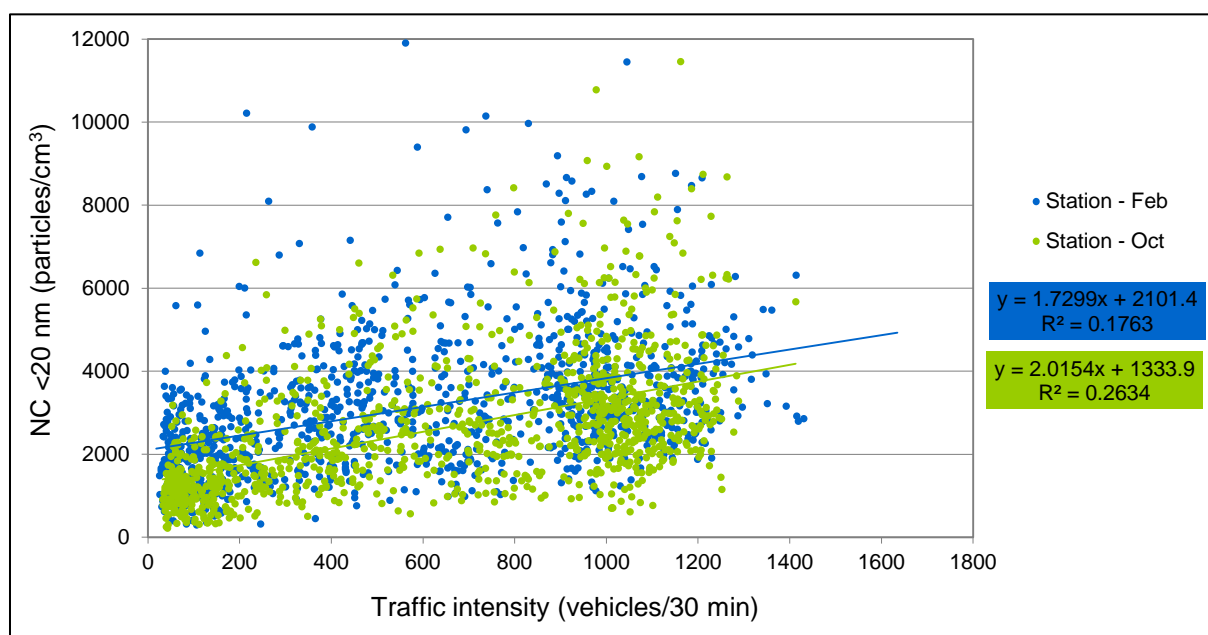


Figure 36: Half-hourly &lt;20 nm particle number concentration (NC) in relation to traffic intensity for the monitoring station during the February and October campaigns

For the public parc site, the Pearson correlation between traffic intensity and the NC <20 nm was 0.66 and 0.51 in February and October, respectively. The slope of the regression was lower in October than in February (Figure 37). So, for a given traffic intensity higher 20-30 nm particle concentrations were measured in February than in October. This may be due to differences in wind direction between the two periods. The public parc site is located at the north of the parc. The road on which the traffic intensity was measured is north from the site, and there is another major road northwards. During the February campaign, north-northeast wind directions were dominant. In October, the dominant wind direction was south to southwest. Therefore, the combustion emissions from both roads may be more blown away from the sampling site during the October campaign than in February.

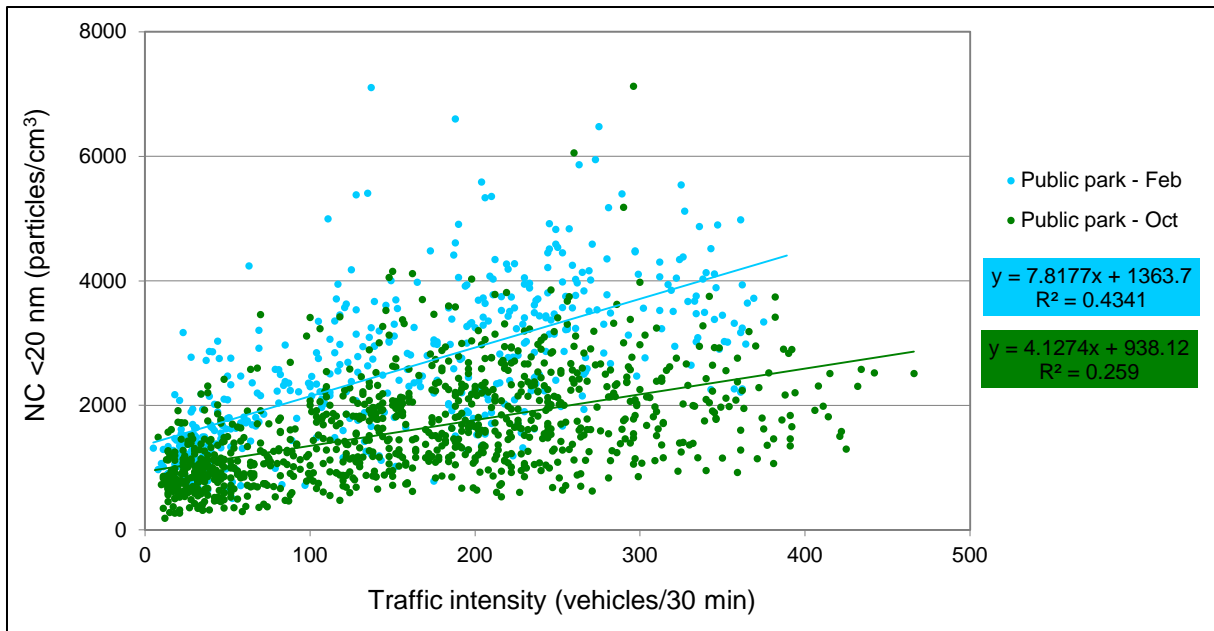


Figure 37: Half-hourly <20 nm particle number concentration (NC) in relation to traffic intensity for the public parc site during the February and October campaigns

For the suburban site, the correlation between traffic intensity and NC20-30 nm was 0.44 and 0.40 in February and October, respectively. The regression lines were similar in both periods (Figure 38).

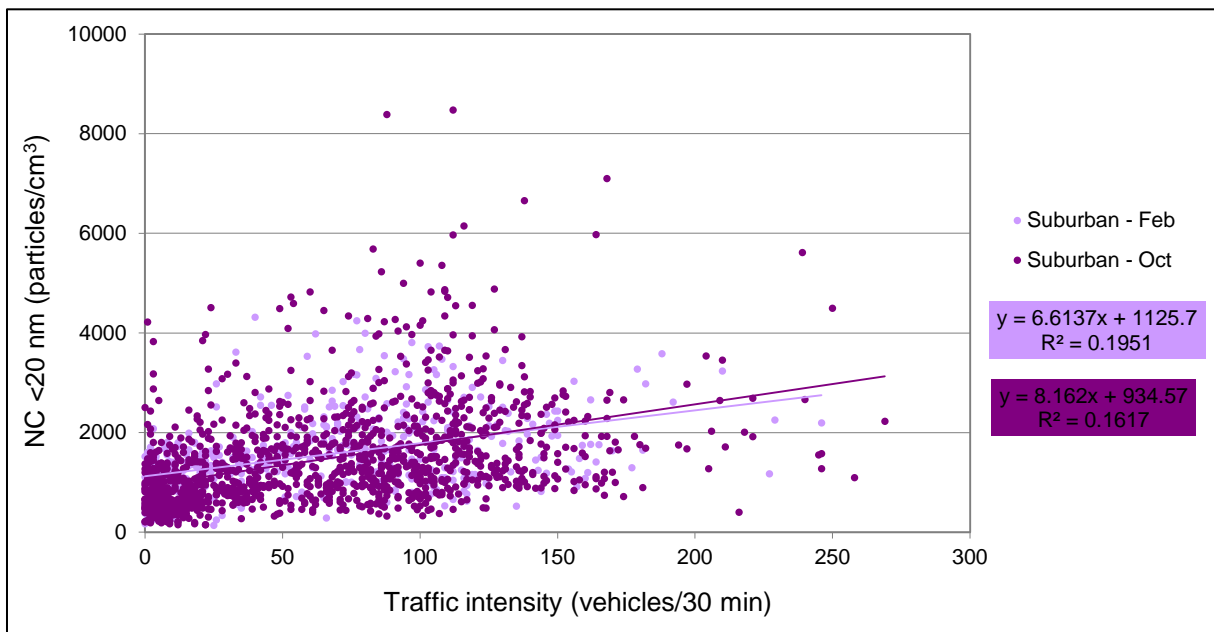


Figure 38: Half-hourly <20 nm particle number concentration (NC) in relation to traffic intensity for the suburban site during the February and October campaigns

At the urban site and ring road site, no particles <20 nm were measured in February. For the urban site in February, the correlation between the number of 20-30 nm particles was only 0.33 (Figure 39). During week 1 and 2 of the February campaign, and using EPC TNC data, the correlation between traffic and TNC was 0.52, however. In October no measurements occurred at this site.

For the ring road site, the correlation between traffic intensity and NC20-30 nm was 0.43 in February and 0.40 in October. The regression slope was higher in February than in October (Figure 40).

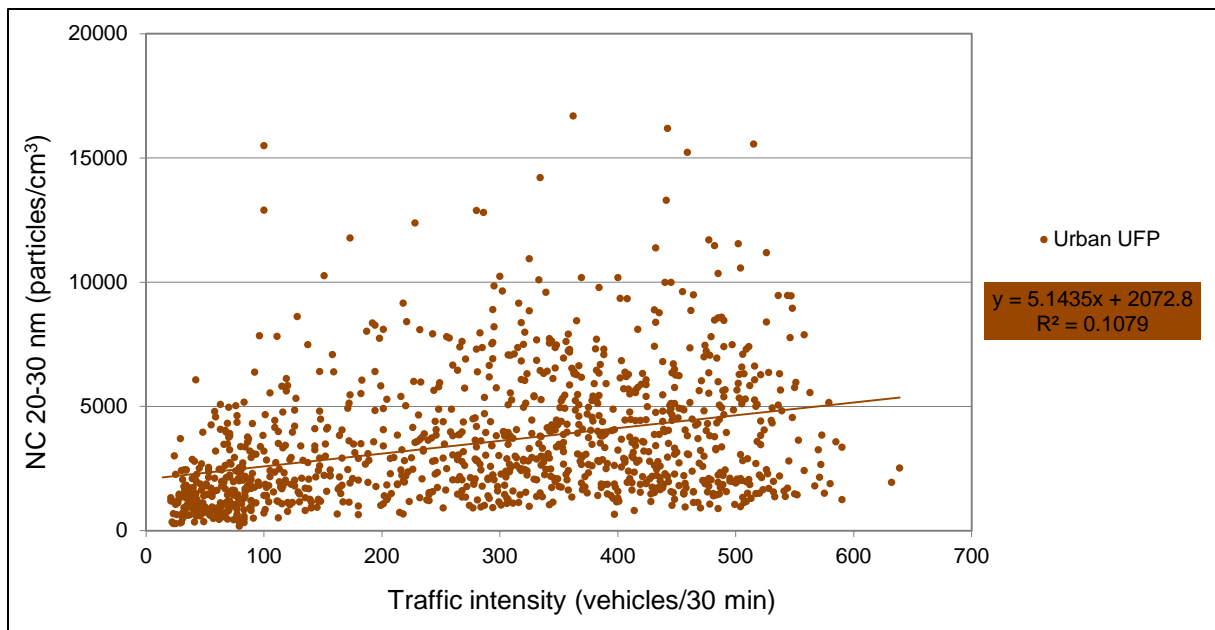


Figure 39: Half-hourly 20-30 nm particle number concentration (NC) in relation to traffic intensity for the urban site during the February campaign

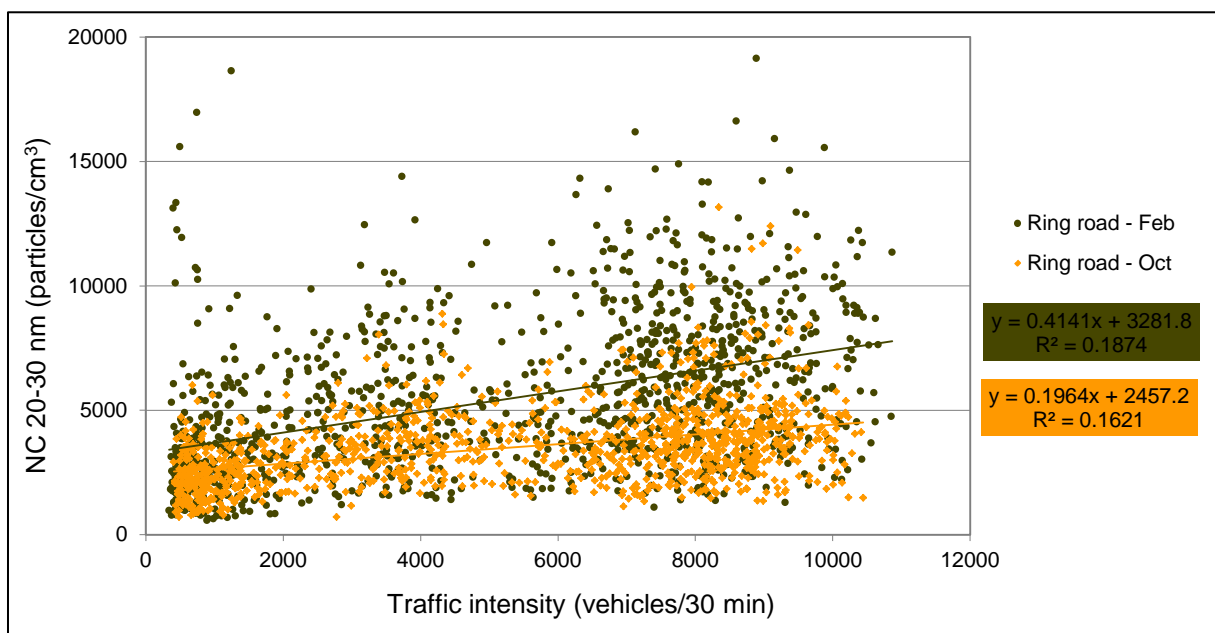


Figure 40: Half-hourly 20-30 nm particle number concentration (NC) in relation to traffic intensity for the ring road site during the February and October campaigns

As shown in the above figures, the slope and intercept of the regressions between particle numbers and traffic intensity varied largely between the different sampling sites and periods. This means that the relationship between traffic intensity and particle numbers depended strongly on the site and period. This is not unexpected, and indicates the influence of other factors such as meteorology and site characteristics. This does not only include the proximity and direction to the main road, but also the mean speed and composition of the vehicles passing by.

A drawback of the current results is that only the number of vehicles has been taken into account. It is known that trucks and buses have larger particle emissions per vehicle unit than passenger cars. It would therefore be relevant to investigate the particle number concentrations in relation to the traffic intensities per vehicle type and/or speed class.

## 4 Conclusions

This report on the number and size distribution of particles  $<1 \mu\text{m}$  in Antwerp presents the results of a gradient study in February 2013 and a study on intra-urban variation in February and October 2013.

The gradient study evaluated the number concentration and the size distribution of particles at 3 sampling sites at 10, 30 and 55 m distance of a main road (2 x 2 lanes). The main results were:

- The total number concentration of particles  $< 1 \mu\text{m}$  (TNC) was higher at 10 m from the road than at 30 and 55 m. The concentration at 55 m was slightly higher than at 30 m, so that for the total 4-week measuring period no clear gradient was observed;
- For downwind conditions the TNC was on average 17% lower at 30 m from the road than at 10 m and 22% lower at 55 m from the road than at 10 m;
- For downwind conditions the number of particles  $<25 \text{ nm}$  decreased with increasing distance to the road. The larger particles differed less between the three distances.

The intra-urban study analysed the spatial-temporal variability of the number concentration and size distribution of particles at 5 sites (February) or 4 sites (October). It was found that:

- The mean TNC varied by a factor of five between the sites and increased with an increasing traffic exposure of the site. On week days all sites showed a diurnal variation in the TNC, with traffic-related peaks during the morning and evening rush hours.
- The TNC was strongly correlated between the urban background site and the 4 other sites. However, this relationship differed between the February and October campaigns.
- The particle number in the distinguished size classes was moderately to strongly correlated between the monitoring station and the 4 other sites. The correlation was lower for particles of 20-30 and 30-50 nm and increased for the larger size classes.
- The particle size distribution generally had a similar trend for all sites and periods. The highest number concentrations were found on week days for particles  $<20 \text{ nm}$  (on the average 26% of the TNC). In the weekends the size classes  $<20 \text{ nm}$  and 30-50 nm contributed equally to the TNC (about 20%). Ultrafine particles dominated the TNC, accounting on the average for 87% of the TNC on week days and 81% in the weekend.
- In comparison with the urban background site, the spatial divergence in the TNC was lowest at the public park site and highest at the street canyon site.

We also examined relationships between particle numbers, 3 other air quality variables and the traffic intensity. It was found that:

- The TNC per site was correlated with  $\text{PM}_{10}$  in October, based on daily means, but the data were limited and the relationship varied per site and most likely also over time;
- The TNC per site and period was strongly correlated with BC and for all sites a maximum correlation value was found for particles of 100-200 nm. The relationship varied per site and period;
- The TNC at all sites was strongly correlated with  $\text{NO}_2$  (weekly data), and smaller particles were more strongly correlated with  $\text{NO}_2$  than bigger particles;
- The TNC per site and period was correlated with the traffic intensity, and the highest correlations were found for small particles.

From this short-term study we conclude that:

- The urban background station can be considered as a community-representative monitoring site for UFP, but it is less representative for rural background areas and more traffic-exposed sites;
- The particle number at 5 sites and during 2 periods was influenced by traffic intensity, but also depended on other factors such as the site characteristics, the proximity to local sources and meteorological conditions;
- During this short-term study positive relationships were found between UFP and  $\text{PM}_{10}$ , BC and  $\text{NO}_2$ , but the relationships varied by site and period;
- More data are needed to examine whether and which relationships exist on the long-term between these parameters and the number and size distribution of ambient particles.



## **ANNEXES**

## Annex 1: Information on accredited measurements (ISO/IEC 17025:2005)

Parameter	SAROAD code	Unit	Device type	Measurement principle of analysis	According to norm	Measurement uncertainty	Determination of measurement uncertainty	Under accreditation	Subcontracted	Outsourced	Type approval
PM <sub>10</sub>	81102	µg/m <sup>3</sup>	Leckel	Gravimetric	EN12341	7.1% at annual mean of 40 µg/m <sup>3</sup> 7.7% at daily mean of 50 µg/m <sup>3</sup>	According to EN12341	yes <sup>1</sup>	no	no	n.a.
PM <sub>10</sub>	81102	µg/m <sup>3</sup>	ESM FH 62 I-R	Beta absorption	-	16% at daily mean of 50 µg/m <sup>3</sup>	Guide to demonstration of equivalence of ambient air monitoring methods (Jan 2010)	yes <sup>1</sup>	no	no	n.a.
NO <sub>2</sub>	42602	µg/m <sup>3</sup>	TS 42i	Chemiluminescence	EN14211	15% at annual mean of 40 µg/m <sup>3</sup> 15% at hourly mean of 200 µg/m <sup>3</sup>	According to EN14211	yes <sup>1</sup>	no	no	yes
BC	16111	µg/m <sup>3</sup>	MAAP 5012	Multi-angle absorption photometry	-	-	-	no	no	no	n.a.
Wind direction	61102	°	Thies Clima 4.3324.31.000	Wind vane	-	-	-	no	no	no	n.a.
Wind speed	61110	m/s	Thies Clima 4.3324.31.000	3-cup anemometer	-	-	-	no	no	no	n.a.
Temperature	62101	°C	RM Young 41342	Platina resistance	-	-	-	no	no	no	n.a.
Relative humidity	62106	%	Campbell Scientific CS215 CS100	Capacitive sensor	-	-	-	no	no	no	n.a.
Precipitation	65102	mm	Casella 103589D	Tipping bucket	-	-	-	no	no	no	n.a.
Pressure	64102	hPa	Campbell Scientific CS100	Capacitive sensor	-	-	-	no	no	no	n.a.

n.a.: not applicable

<sup>1</sup> : BELAC 456-TEST - VMM Dienst Lucht



## Annex 2: Instrument correction factors

Instrument correction factors were determined during the following campaigns:

- 13/12/2012 till 30/01/2013 in Wilrijk (Vuurkruisenplein): 12 instruments were compared during 3-6 weeks, depending on the type of instrument, as described in the comparison report:
  - 5 x EPC;
  - 3 x UFP monitor;
  - 3 x Grimm SMPS;
  - 1 x IFT customized SMPS.
- 04/04 till 24/04/2014 at the urban background site (monitoring station): 3 instruments were compared during 3 weeks:
  - 1 x MAAP;
  - 1 x AE22;
  - 1 x AE33.
- 30/09 till 08/10/2013 at the urban background site (monitoring station): 7 instruments were compared during 1 week:
  - 2 x EPC;
  - 1 x Grimm SMPS;
  - 2 x IFT customized SMPS;
  - 2 x MAAP.
- 28/11 till 04/11/2013 in Mol (VITO site): 2 instruments were compared during 1 week:
  - 1 x EPC;
  - 1 x CPC.
- 04/11 till 19/11/2013 at the urban background site (monitoring station): 4 instruments were compared during 2 weeks:
  - 2 x Grimm SMPS;
  - 2 x MAAP.