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Total and size-resolved particle number and black carbon concentrations in urban areas near Schiphol airport (the Netherlands)

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HIGHLIGHTS

- Ultrafine particles are a factor 3 elevated 7 km downwind Schiphol airport.
- The size-distribution of these particles is dominated by particles of 10–20 nm.
- 45,000/60,000 addresses exposed to 5–10,000 (annual)/10–20,000 (hourly) #/cm^3.

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ABSTRACT

The presence of black carbon, and size-resolved and total particle number concentrations (PNC) were investigated in the vicinity of Schiphol airport in the Netherlands, the fourth busiest airport in Europe. Continuous measurements were conducted between March and May 2014 at Adamse Bos, located 7 km from Schiphol, and in 2012 at Cabauw, a regional background site 40 km south of Schiphol. No significantly elevated black carbon levels were found near Schiphol. However, PNC increased during periods in which the wind direction was from Schiphol: at Cabauw by 20% and at Adamse Bos by a factor of three, from 14,100 (other wind directions) to 42,000 # cm^{-3} between 06.00 and 23.00. The size distribution of Schiphol-related PNC was dominated by ultrafine particles, ranging from 10 to 20 nm. Four relevant particle number (PN) emission sources at Schiphol were identified as being responsible for the elevated PNC levels at Adamse Bos: take-off and climb-out on the Kaagbaan and Aalsmeerbaan runways, planes waiting at the gates, and landing on the Buitenveldertbaan runway. PN emissions from road traffic at and near the airport were less important than air traffic. The exposure to Schiphol-related PNC in urban areas northeast of Schiphol in Amsterdam and Amstelveen was estimated for 2012 using a Gaussian Plume model. The results showed that a considerable number of the 555,000 addresses in the modelling domain were exposed to elevated PNC. For example: 45,000 addresses suffered *long-term* exposure to an additional annual background PNC of 5–10,000 # cm^{-3} originating from Schiphol and 60,000 addresses suffered *short-term* exposure (14% of the time) of additional 10–15,000 # cm^{-3} originating from Schiphol. Further research on emission sources and the dispersion of PN is recommended and may support future studies on eventual health effects.

1. Introduction

Many epidemiological studies have established associations between exposure to the mass of ambient particulate matter (PM) and adverse health effects. Various physical and chemical fractions in PM have been proposed as the cause of these acute and chronic effects (Cassee et al., 2013). One of the suspect fractions concerns the large number of submicron particles because the latter may contain potential toxic species and can penetrate deep into the respiratory system (Loane et al., 2013; Oberdörster et al., 2005). The number of submicron particles is dominated by ultrafine particles, which are smaller than 100 nm (Sioutas et al., 2005). Submicron particles in ambient air originate in primary emissions from combustion processes and the secondary formation of sulphates, nitrates and organic PM from gas-to-particle conversion in ambient air (Chow and Watson, 2007). During initial cooling and further

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dispersion of combustion emissions in ambient air, the size-distribution and concentration of submicron particles are affected by nucleation, condensation/evaporation, coagulation, dry deposition and dilution (Ketzel and Berkowicz, 2004; Kumar et al., 2011). Despite concern about the health effects, epidemiological studies related to exposure to PNC are scarce due to the challenges involved in assessing population exposure (HEI, 2013; Hoek et al., 2010; Kumar et al., 2011; Sioutas et al., 2005).

Airports are important sources of submicron particles, as shown by studies on emissions from jet engines (Kinsey et al., 2010; Mazaheri et al., 2011; Vander Wal et al., 2014) and measurements at and near airports (Hudda et al., 2014; Keuken et al., 2012; Westerdahl et al., 2008). A recent review of airport emissions (Masiol and Harrison, 2014) concluded that 'despite the increased attention given to aircraft emissions at ground level and air pollution in the vicinity of airports, many research gaps remain', including emissions and dispersion of submicron particles. In our study, total and size-resolved PNC, and black carbon concentrations, were investigated near Schiphol airport. Black carbon (BC) was measured as a sensitive indicator of the *mass* of PM in combustion emissions (Maricq, 2007). Schiphol is the fourth busiest European airport, handling 52 million passengers in 2012. The airport is located in the densely populated west of the Netherlands and noise nuisance is considered the most important environmental impact (Schiphol, 2013). In contrast, emissions of air pollutants have been found to contribute less than 5% to ambient concentrations of the compliance indicators PM_{10} and NO_2 near the airport (Dassen et al., 2006), suggesting that airport emissions have a limited impact on local air quality. We tested this hypothesis by measuring PNC and BC, and using dispersion modelling near Schiphol.

2. Methodology

2.1. Approach used in the study

In 2012, total and size-resolved PNC were measured along with BC at a *regional background site* in the Netherlands ('Cabauw'). In addition, a monitoring site was set up near Schiphol from March to May 2014 ('Adamse Bos'). The hourly average PNC and BC measurements at both sites, combined with the prevailing wind direction over that period (36 directions in total), were plotted in 'pollution roses'. These pollution roses identified PN and BC emission sources upwind of both sites in wind directions with elevated PNC and BC. The size-resolved PNC measurements at both sites were used to analyse the size distribution of PNC in different wind directions.

Meteorological data were retrieved from the National Meteorological Monitoring Network at the Schiphol site (www.knmi.nl). Annual average air traffic data (e.g. the number of landings and take-offs [LTO] per hour at different runways) was available for 2012 (Schiphol, 2013). The total number of landings and take-offs at Schiphol in 2012 was about 440,000, an increase of 1.6% compared to 2011 (Schiphol, 2013). The meteorological and air traffic data were used as input for a dispersion model to estimate the contribution of airport emissions to PNC in urban areas near Schiphol. Modelling was conducted for 2012 as this was the most recent year for which open-source, air traffic data was available. Finally, the annual average exposure of the population to PNC in these areas was estimated ('long-term exposure') and compared to specific periods in which the wind was coming from Schiphol ('short-term exposure').

2.2. Monitoring sites and air traffic at Schiphol

The two monitoring sites were the regional background site of Cabauw (51° 58′13 N; 4° 55′34 E) and the Amsterdamse Bos, a park situated between Schiphol and the urban areas of Amsterdam-Amstelveen (52° 19′17 N; 4° 50′52 E). Cabauw is a site used by the National Air Quality Monitoring Network in the Netherlands and is situated about 40 km south of Schiphol. The Adamse Bos site, which was established as a temporary monitoring location, lies about 7 km east of Schiphol. The sites are shown in Fig. 1, including dominant flight paths of arriving and departing planes at Schiphol (Schiphol, 2013).

Fig. 1 illustrates that Schiphol is situated near densely populated areas, particularly to the west and northeast of the airport. The south and south westerly directions are the dominant flight paths at Schiphol (2013) due to the prevailing south-westerly winds, and because of the need (e.g. noise hindrance) to avoid the densely populated areas to the west and northeast of Schiphol. Fig. 2 shows the six runways and the distribution of hourly average wind directions in percentages ('wind rose') at Schiphol in 2012. More details on the geographical location of the runways at Schiphol, including their lengths are presented in Fig. 5.

The wind rose for 2012 in Fig. 2 shows that south-westerly winds occur with the greatest frequency at Schiphol. The flight codes in Fig. 2 indicate the flight directions of arriving and departing planes at a particular runway: *18R/36L (Polderbaan), 18C/36C (Zwanenburgbaan)), 27/09 (Buitenveldertbaan), 24/06 (Kaagbaan)), 18L/36R (Aalsmeerbaan) and 22/04 (Oostbaan)*. These codes are used to register the number of flights at each runway, as shown in Fig. 3 for 2012 (Schiphol, 2013).

Fig. 3 shows that most planes land on the Polderbaan runway in a southerly direction (code: 18R) and most planes take-off from the Kaagbaan runway in a south westerly direction (code: 24). Most air traffic at Schiphol occurs between 06.00 and 23.00, with around 5% of air traffic in the remaining night-time hours.

2.3. Monitoring equipment

Both at Cabauw in 2012 and at Adamse Bos between March and May 2014, size-resolved PNC was measured continuously with a Scanning Mobility Particle Sizer (SMPS 3034; TSI Inc.) covering a size range of up to 480 nm. A measurement scan takes about 3 min. The number of particles per size class were detected with a Condensation Particle Counter (CPC 3010; TSI Inc.) with a 50% cut-off at 10 nm. In addition, at Adamse Bos the total PNC was simultaneously measured using a CPC 3775 (TSI Inc.) and a 50% cut-off at 4 nm. Black carbon concentrations were also measured at both sites with a multi-angle absorption photometer, the MAAP model 5012 (Thermoscience). The black carbon measurements were converted to elemental carbon (EC) concentrations to facilitate comparison with thermal measurements of EC (Keuken et al., 2013). The detection limit of the MAAP is 0.1 μg EC per m^3.

The applied Quality Control procedures were derived from the European EUSAAR (European Supersites for Atmospheric Aerosol Research) research project (Asmi et al., 2011). These involved inter-comparison studies of monitoring instruments, 2-weekly checking of the sampling flow, and annual calibration of the PN monitors by the manufacturer and of the black carbon monitoring with thermal analysis of EC and OC (Keuken et al., 2013).

2.4. Dispersion modelling

The contribution of Schiphol emissions to PNC in the urban areas of Amsterdam and Amstelveen was modelled with an hourly version of the Gaussian Plume model (SRM3). This is the regulatory model used in the Netherlands to calculate the dispersion of air pollutants from point and surface area sources to a maximum distance of 25 km from the source (Infomil, 1998). The model is *not* suited to the calculation of particle numbers, as mass units are

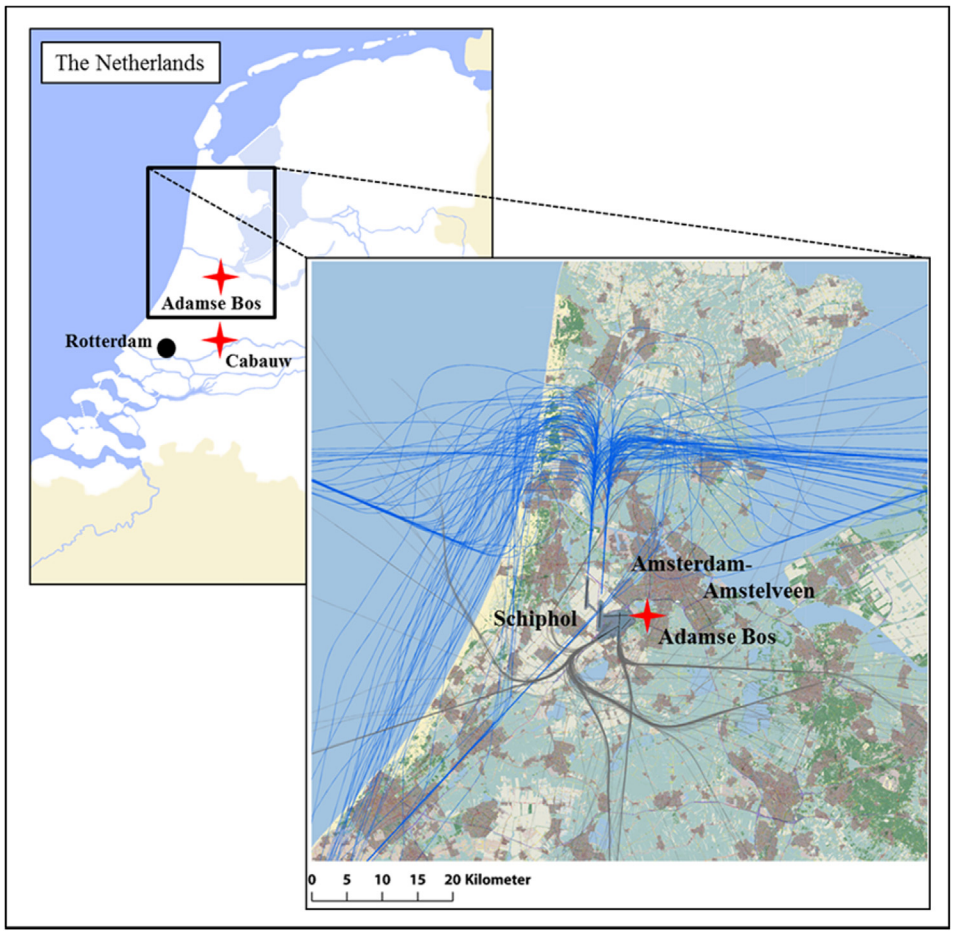

Fig. 1. Monitoring sites: Cabauw and Adamse Bos, the urban areas Amsterdam-Amstelveen and dominant flight paths for landing (blue) and take-off (grey) at Schiphol (2013).

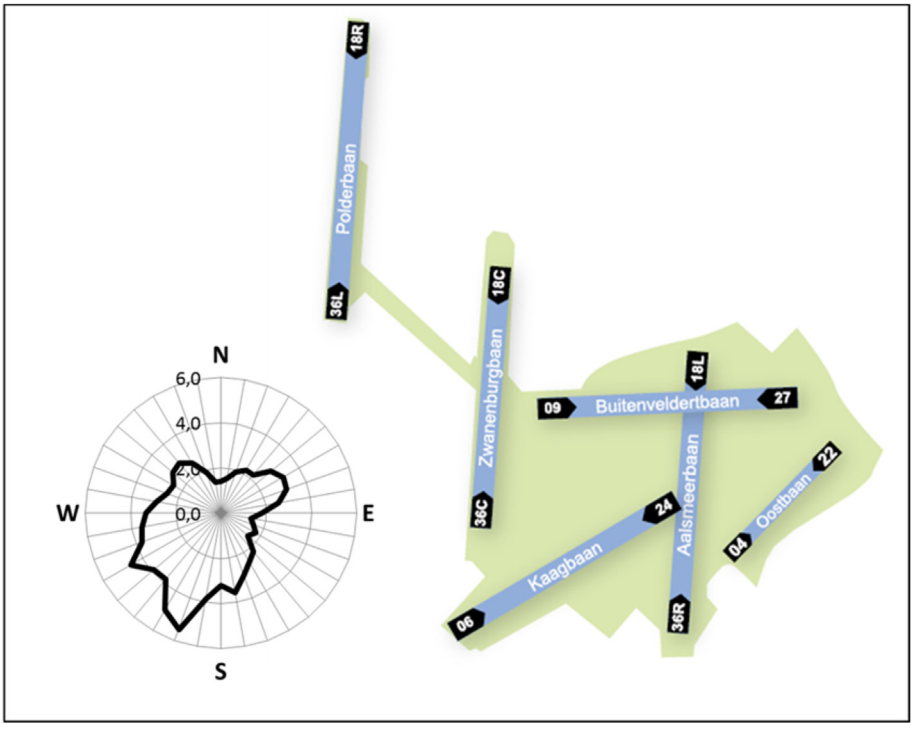

Fig. 2. The six runways, including flight codes and the wind rose (%) in 2012 at Schiphol.

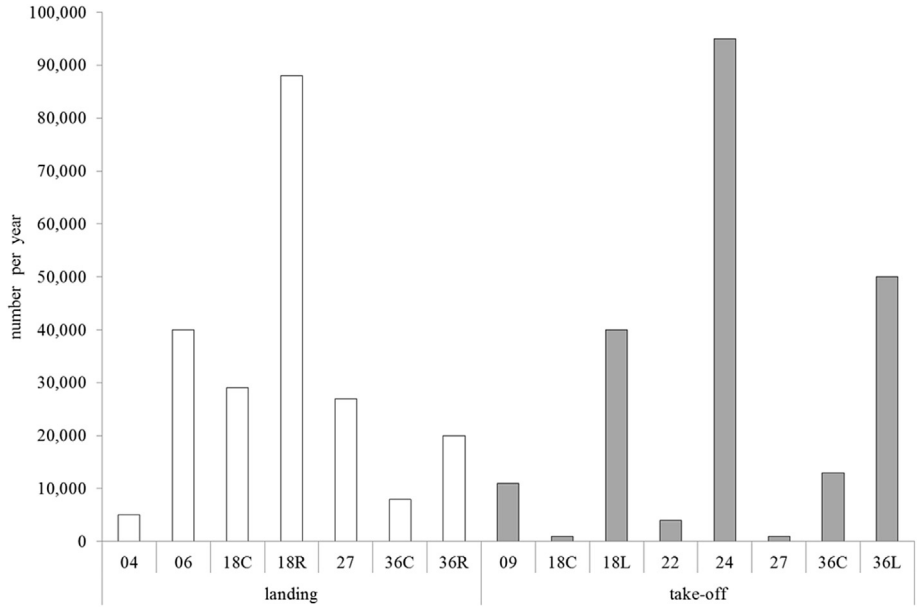

Fig. 3. The number of landings and take-offs at Schiphol runways in 2012.

embedded in the SRM3 software. Therefore, the dispersion of PN was treated as an inert gas ('X') as a *proxy* for the dispersion of PN and scaling the spatial distribution of X with the measurements of PNC at Adamse Bos, as follows:

1. *Emission strength of X (kg s^{-1}) at Schiphol*: four point sources were located at Schiphol with an *arbitrary*, total emission strength of 1 kg X per second and a 24-hourly emission profile which took into account the diurnal variation in air traffic intensity at Schiphol (see: Section 3.3).
2. *Conversion factor of X (μg m^{-3}) to PNC (# cm^{-3})*: the average hourly contribution to concentrations of X at Adamse Bos was calculated with SRM3 for hours with Adamse Bos downwind from Schiphol for the monitoring period in 2014 (see: Section 3.2). The modelled contribution to concentrations of X was compared with the measured contribution to PNC during these selected hours. This resulted in a conversion factor between the *modelled* X and the *measured* PN contribution for the monitoring period in 2014.
3. *Modelled contribution to X (μg m^{-3}) in 2012*: the contribution to concentrations of X in 2012 in the urban areas of Amsterdam and Amstelveen was modelled using the meteorological data from 2012, four emission locations and an emission strength of 1 kg X per second, as in Step 1. The grid size of the modelling output was 20*20 km and the grid resolution was 0.5*0.5 km. The spatial resolution of the output was increased to 50*50 m by interpolation between the modelled X concentrations. The interpolation was performed with the 'natural neighbour' algorithm from the spatial analyst tool in ArcGIS software (www.esri.com). The modelling was performed to determine the contribution to annual average concentrations and to concentrations when the wind was coming from Schiphol in 2012.
4. *Population exposure to PNC (# cm^{-3}) contribution in 2012*: the contribution to PNC concentrations was obtained by converting the contribution to concentrations of X from Step 3 with the conversion factor from Step 2. The long-term and short-term exposure of the population to Schiphol-related PNC was determined by matching the population density in the urban areas of

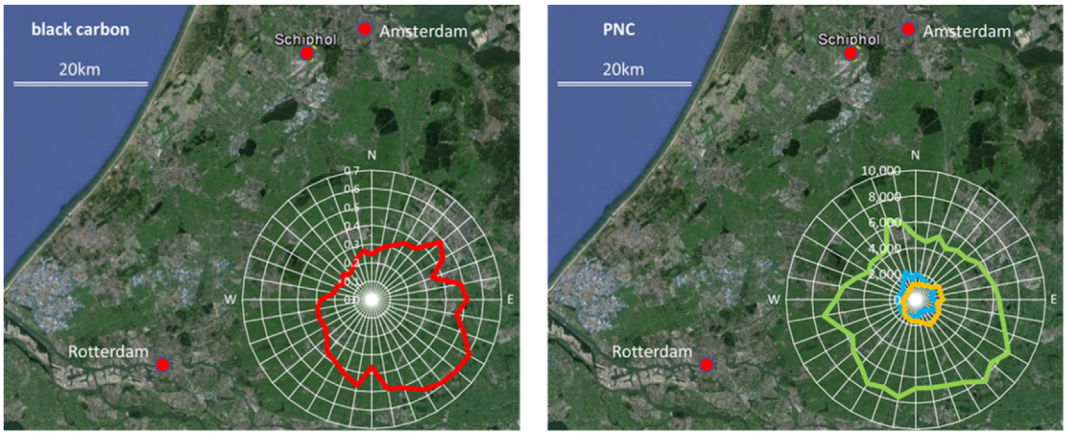

Fig. 4. Pollution roses of black carbon in μg EC per m^3 (left) and size-resolved PNC in # per cm^{-3} (right): 10–25 nm (blue), 25–100 nm (green) and 100–480 nm (yellow) at Cabauw in 2012 (© Google maps).

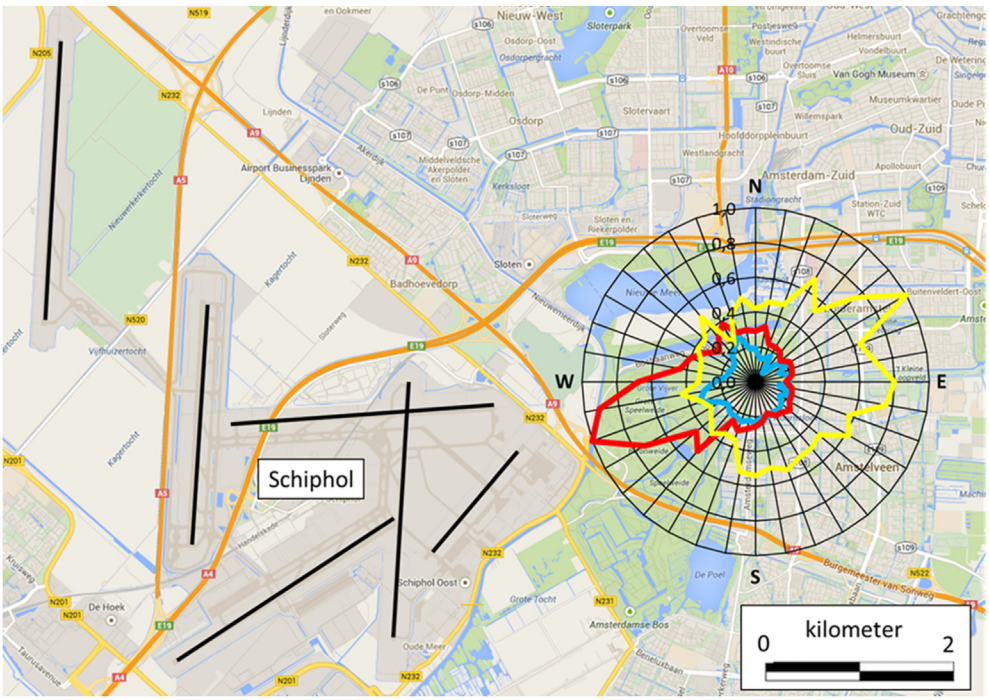

Fig. 5. Normalized pollution roses of EC and PNC at Adamse Bos: for EC all hours (n = 1486) in yellow and for PNC during day-time hours (n = 1112) in red and night-time hours (n = 460) in blue (© Google maps).

Amsterdam and Amstelveen with the spatial distribution of PNC.

It should be noted that using a different *value* than that of 1 kg X per second for the emission strength would have changed the X–PNC conversion factor but also the spatial distribution of X in 2012. Consequently, for the spatial distribution of PNC in 2012 – based on the conversion factor and the dispersion of X in 2012 – the *value* of the emission strength of X at Schiphol is not relevant in Steps 1–4 of the modelling approach.

3. Results

3.1. BC and PNC at the regional background

The annual average black carbon and PN concentrations in 2012 at Cabauw were 0.5 μg EC per m^3 and 9600 # cm^{-3}, respectively. For data analysis, the annual average PNC were grouped in three size classes: i) 1400 # cm^{-3} (10–25 nm), ii) 6100 # cm^{-3} (25–100 nm) and iii) 1400 # cm^{-3} (100–480 nm). These size classes were selected to represent: nucleation particles smaller than 25 nm, ultrafine particles smaller than 100 nm and accumulation particles larger than 100 nm. From these measurements, pollution roses for EC and PNC were derived and presented in Fig. 4.

Upwind emission sources relative to Cabauw can be traced in wind directions with elevated EC and PNC. For both black carbon and PN, sources were identified in the south-easterly direction (e.g. heavy traffic and the industrialized Ruhr Area in Germany), while sources for PN were also identified in the westerly direction, with polluted air from the industrial and harbour areas near the city of Rotterdam. Moreover, in particular, in the size range of 25–100 nm, PNC were elevated during hours when the wind was blowing from the direction of Schiphol. It was considered unlikely that emissions from Schiphol could be detected at a distance of almost 40 km (at Cabauw). However, recent research has shown that PNC may be elevated by a factor of four, 10 km downwind of the Los Angeles Airport (Hudda et al., 2014). The area between Cabauw and Schiphol is mainly pasture land, with no specific PN emission sources other than motorways. The latter were also present in other wind directions but were not detected at Cabauw. These findings at Cabauw triggered further research on dispersion of PN from Schiphol starting with the monitoring campaign at Adamse Bos 7 km from Schiphol.

3.2. Black carbon and PNC at Adamse Bos near Schiphol

Black carbon and PNC were measured at Adamse Bos in the period from 12 March to 19 May 2014. In total, 1572 h with a wind speed higher than 1 m s^{-1} were selected. A lower wind speed occurred in only 5% of the total number of hours and these were excluded from the data analysis. The average black carbon and PNC during the monitoring period were 0.7 μg EC per m^3 and 17,300 # cm^{-3}, respectively. The average black carbon and PNC for periods with different wind directions are shown in Fig. 5. In order to present the results for black carbon and PNC in one graph, the data were normalized by dividing by the maximum value. The data for PNC are presented separately for day-time (06.00–23.00) and night-time (23.00–06.00) periods.

Fig. 5 shows that black carbon was mainly elevated when the wind was blowing from the urban areas of Amsterdam-Amstelveen with black carbon emission from road traffic, but there were no elevated concentrations downwind of Schiphol. The latter is in agreement with black carbon measurements near a runway at Schiphol in 2010, from which it was concluded that planes have limited black carbon emissions during landing and take-off (Keuken et al., 2012). In contrast, PNC were especially elevated during day-time hours downwind of Schiphol for wind directions in the 240°–270° range. The limited elevated PNC during night-time hours with these wind directions reflect the limited air traffic during night-time with less than 5% of total air traffic. The

average night-time and day-time PNC were 10,200 # cm^{-3} (n = 460 h) and 20,200 # cm^{-3} (n = 1112 h), respectively. Fig. 6 presents the average day-time and night-time PNC for periods in which the wind was blowing from Schiphol and from other directions.

Fig. 6 shows that during the night (14,000 # cm^{-3}) and particularly during the day (42,000 # cm^{-3}), PNC in the Schiphol wind sector were elevated. The average PNC from all wind directions was 17,300 # cm^{-3} (n = 1572), while when the wind was blowing from directions other than from Schiphol, PNC was 12,800 # cm^{-3} (n = 1282). Hence, PN emissions at Schiphol resulted in an average increase in PNC of 4500 # cm^{-3} in the study period. During daytime hours in the Schiphol wind sector, average hourly PNC may increase by a factor of three (42,000 # cm^{-3}) compared to the daytime background (14,100 # cm^{-3}). The wind blew from these directions in the Schiphol wind sector for about 15% of the time during the study period. This percentage closely reflects the annual average frequency of these wind directions at Schiphol as measured in 2012 (see Fig. 2). Details of the PN emission sources at Schiphol (e.g. departing and arriving planes, ground-based operations and road traffic) are given below.

3.3. PN emission sources at Schiphol

Aircraft PN emissions increase with the thrust level of the jet engine. This level varies depending on the type of operation involved: take-off (100%), climb-out (85%), landing (30%) and idling during ground-based operations, such as taxiing to and from runways and parking at the gate (7%) (Mazaheri et al., 2011). The related PN emissions are highest during take-off (1) and decrease at lower thrust levels: climb-out (0.5), landing (0.1) and ground-based operations (0.01). These figures show that the PN emissions of planes are most elevated during take-off and climb-out. However, these operations are of relatively short duration compared to ground-based operations. Thus, despite relatively low PN emissions, the latter operations may also be important factors affecting PNC near airports. In addition, road transport at and near the airport is also a source of PN emissions. PNC measurements at Adamse Bos were used to analyse the significance of these various PN sources at Schiphol, focussing on:

a) *The relationship between PNC and the wind speed.* Carslaw et al. (2006) have shown that ground-level concentrations downwind of buoyant plumes (e.g. emissions from aircrafts climbing out and landing) reach a maximum at lower wind speeds and decrease little with increasing wind speeds, due to these buoyant plumes being brought down to ground-level by increasing winds. In contrast, downwind concentrations of plumes with little or no buoyancy (e.g. road traffic emissions and ground-based aircraft operations) decrease with increasing wind speed. This different relationship between downwind concentrations and wind speed has been used to distinguish ground-based emissions from take-off/landing emissions. The average PNC contribution at Adamse Bos for three wind speed classes when the wind was blowing from the direction of Schiphol in day-time hours were: 31,400 # cm^{-3} for wind speeds from 1 to 3 m s^{-1} (n = 29), 27,000 # cm^{-3} for wind speeds between 4 and 7 m s^{-1} (n = 99) and 27,000 # cm^{-3} for wind speeds from 8 to 13 m s^{-1} (n = 94). This observed relationship is typical for buoyant plumes, indicating that take-off/landing are likely to be more important sources of PN emissions at Schiphol than ground-based operations and road traffic at and near the airport.
b) *Temporal variation in PNC contribution.* The contribution of Schiphol emissions to hourly PNC at Adamse Bos was calculated by subtracting the night-time and day-time background PNC from the measurements: 9600 and 14,100 # cm^{-3}, respectively (see: Fig. 6). The diurnal variation in this contribution during periods when the wind was blowing from the direction of Schiphol is presented in Fig. 7.

The contribution of Schiphol emissions to PNC at Adamse Bos and the air traffic intensity at Schiphol were correlated with the square of the correlation coefficient (R^2) of 0.77. This means that 77% of the elevated PNC in the Schiphol wind sector can be explained by air traffic at Schiphol. Fig. 7 shows that from 08.00 to 12.00 and from 20.00 to 22.00, in particular, the contributions to PNC were elevated in accordance with high air traffic intensity. The

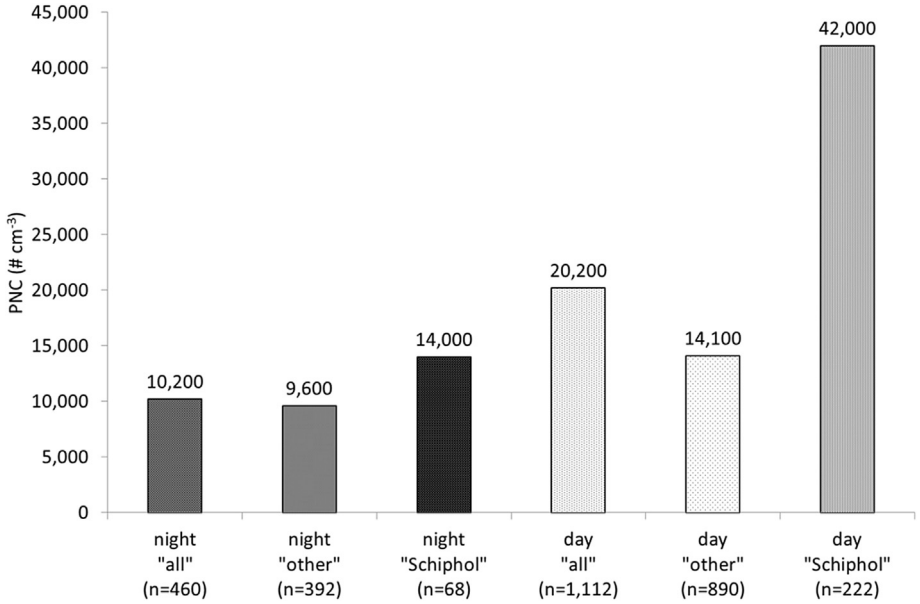

Fig. 6. Average PNC at Adamse Bos during night-time and day-time hours (n) for all wind directions ('all'), for wind directions other than from Schiphol ('other') and for wind directions from Schiphol ('Schiphol').

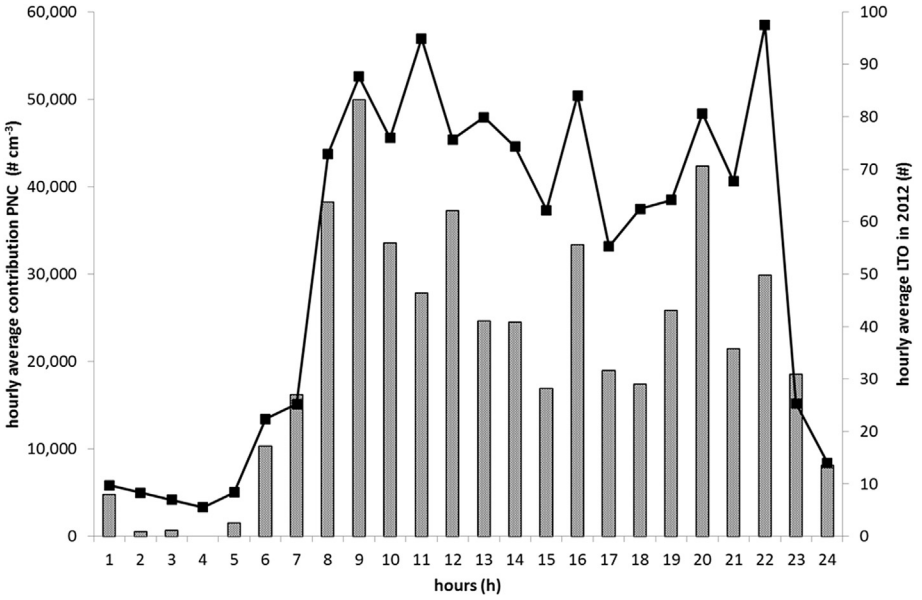

Fig. 7. Hourly average contribution to PNC in # cm^{-3} at Adamse Bos with wind directions from Schiphol (n = 290) (left Y-axis) and the average number of LTO per hour (■) in 2012 (right Y-axis).

latter was derived from the hourly average sum of landings and take-offs (LTO) in 2012 (Schiphol, 2013), because air traffic data in the monitoring period (2014) was not available. It is assumed that average air traffic intensity in 2012 is a good *proxy* for air traffic in the monitoring period, as the growth of air traffic per year is only a few percentages (Schiphol, 2013). The lower PNC contribution in the afternoon hours as compared to the morning hours is attributed to the relatively higher mixing height at Schiphol, as presented in Fig. 8.

The contribution of Schiphol emissions to PNC at Adamse Bos and the mixing height were correlated with the square of the correlation coefficient (R^2) of 0.32. This means that 32% of the diurnal variation in the contribution of PNC at Adamse Bos may be explained by variation in the mixing height at Schiphol. Hence, from Figs. 7 and 8, it can be concluded that the diurnal variability in elevated PNC at Adamse Bos can mainly be accounted for by air traffic intensity and by the mixing height involved.

c) *Different PN emission sources at Schiphol.* As well as overall air traffic intensity, the use of particular runways for take-off and landing—in combination with wind direction — are important parameters for the contribution to PNC downwind of Schiphol. The average contribution of PNC at Adamse Bos in four relevant wind directions during 222 day-time hours were: 23,100 # cm^{-3}

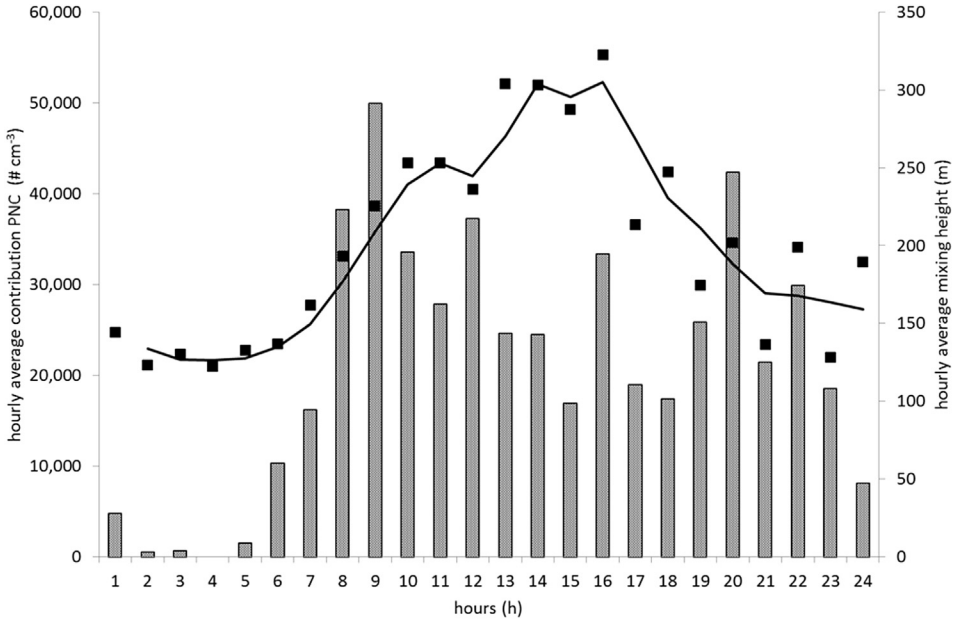

Fig. 8. Hourly average contribution to PNC in # cm^{-3} at Adamse Bos with wind directions from Schiphol (n = 290) (left Y-axis) and the average mixing height (2-period moving average) per hour (■) at Schiphol (right Y-axis).

(n = 82), wind direction 240°; 36,500 # cm^{-3} (n = 53), wind direction 250°; 31,700 # cm^{-3} (n = 47), wind direction 260°; and 20,000 # cm^{-3} (n = 40), wind direction 270°. The upwind sources at Schiphol associated with these wind directions (see: Figs. 3 and 5) are the Aalsmeerbaan runway for take-off (240°), the Kaagbaan runway for take-off (250°), the gates at the centre of Schiphol (260°) and the Buitenveldertbaan runway for landing (270°). These sources, with a total source strength of 1 kg X per second and a relative source strength of 0.2, 0.4, 0.25 and 0.15, respectively (based on their contributions to each of the four wind directions (240°–270°) and their distance to Adamse Bos) were used in the dispersion modelling of PN near Schiphol in Section 3.5 (below).

The size-resolved distributions of PNC at Cabauw and Adamse Bos are presented below.

3.4. Size-resolved PNC at Cabauw and Adamse Bos

The results of the size-resolved PNC measurements were divided into two groups at Cabauw: i) periods in which the wind was blowing from the direction of Schiphol (340°–350°) and ii) periods in which the wind was blowing from other directions, and into three groups at Adamse Bos: i) night-time hours, ii) day-time hours in which the wind was blowing from the direction of Schiphol (240°–270°) and iii) day-time hours in which the wind was blowing from other directions. The size distributions of these five groups are presented as normalized concentrations ($dN/dlog(D_p)$) against the diameter of the particles (D_p) in Fig. 9.

Fig. 9 shows that the size distribution of PNC downwind of Schiphol at both sites differs from other wind directions and from measurements taken during the night at Adamse Bos. The size distribution at Adamse Bos –during wind directions from Schiphol - is dominated by nucleation particles in the size range of 10–20 nm, which is in agreement with other studies near airports (Masiol and Harrison, 2014). At Cabauw, the size distribution downwind of Schiphol shifts to particles in the size range of 20–40 nm due to the more remote location of Cabauw relative to Schiphol (40 km), compared to Adamse Bos (7 km). For other wind directions, the size distributions at both sites are comparable but with relatively more nucleation particles at Adamse Bos due to the vicinity of combustion sources in Amsterdam and Amstelveen (e.g. road traffic, domestic heating, power station), compared to Cabauw.

3.5. Dispersion modelling of total PNC near Schiphol

The contribution of Schiphol emissions to PNC in the urban areas of Amsterdam-Amstelveen that lie northeast of Schiphol was estimated using a dispersion model. As described in Section 2.4, the dispersion of the tracer X emitted from sources at Schiphol with a total strength of 1 kg X per second was first modelled to estimate the contribution to hourly concentrations of X in μg m^{-3} at Adamse Bos during the monitoring period (March–May 2014). Based on the findings described in Section 3.3, four emissions sources relevant for dispersion to Amsterdam-Amstelveen were identified: 1. Buitenveldertbaan (0.15 kg X s^{-1}), 2. Schiphol gates (0.25 kg X s^{-1}), 3. Kaagbaan (0.4 kg X s^{-1}) and 4. Aalsmeerbaan (0.2 kg X s^{-1}). In addition, a source profile was similarly applied for all sources, based on the contribution of PNC at Adamse Bos during day-time hours (see Fig. 7). Based on these emission parameters (e.g. source location and strength) and *current* meteorological conditions (e.g. wind speed and stability), the contribution to hourly average X concentrations at Adamse Bos was modelled for 222 day-time hours with the wind directions of 240°–270° from March to May 2014. On this basis, an average conversion factor was derived between hourly *modelled* X and *measured* PNC contribution. Then, using the same sites and source strength of X as were used during the 2014 monitoring period, the contribution to concentrations of X in Amsterdam-Amstelveen was modelled for 2012: for all hours (n = 8784) and for hours with wind directions ranging from 240° to 270° (n = 1275). Finally, the modelled contribution to concentrations of X in μg m^{-3} was scaled to PNC in # cm^{-3} using the X/PNC conversion factor. The results for PNC in 2012 are presented in Fig. 10.

In Fig. 10, the wind rose of PNC measurements at the Tropenmuseum is presented for a wind speed higher than 1 m s^{-1}. These measurements were conducted from October 2002 to March

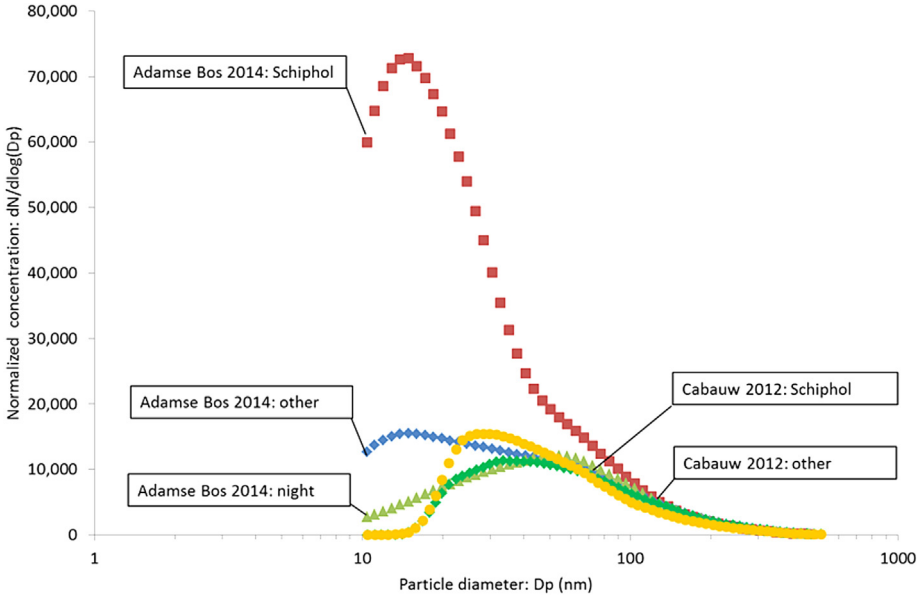

Fig. 9. Normalized size distribution (dN/dlog(Dp)) versus particle size (Dp in nm) at Adamse Bos (March–May 2014): night-time hours (▲; n = 460), day-time hours with wind directions from Schiphol (■; n = 222) and remaining day-time hours (■; n = 890), and at Cabauw (2012): with wind directions from Schiphol (●; n = 131) and remaining hours (■; n = 8461).

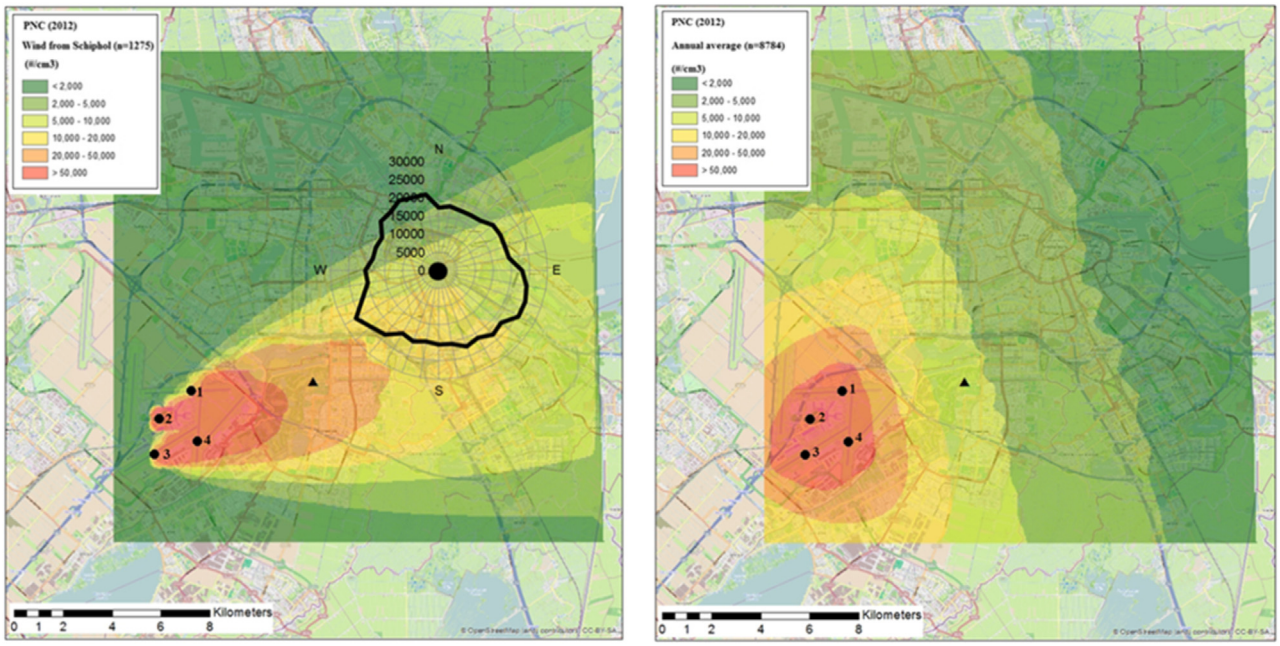

Fig. 10. Contribution of Schiphol emissions from (1) Buitenveldertbaan (2) Schiphol gates (3) Kaagbaan and (4) Aalsmeerbaan to PNC (#.cm^{-3}) in the urban areas of Amsterdam-Amstelveen in 2012, including the monitoring site in Adamse Bos (▲). Left: with wind directions from Schiphol (n = 1275), including the pollution rose of PNC for the period 2002–2004 at the Tropenmuseum (●). Right: annual average for all wind directions (n = 8784) (© Google maps).

2004 at 15 m height with a CPC 3022 (TSI Inc.) with a 50% cut-off of 7 nm (Puustinen et al., 2007). The PNC was elevated with 5000 # cm^{-3} in the wind direction from Schiphol (240°) as compared to the average PNC of 21,000 # cm^{-3} in other wind directions. These measurements confirm the results of the dispersion modelling and also indicate that Schiphol airport is contributing to elevated PNC in the urban areas of Amsterdam and Amstelveen already over a considerable period. The modelled contribution to PNC at the Adamse Bos in 2012, as presented in Fig. 10 was also compared to the measured contribution in the 2014 monitoring period. For the annual average, the figures were 6200 (2012) and 4500 (2014) with wind directions from Schiphol, they were 34,700 (2012) and 27,900 (2014). The agreement between the modelled and measured contributions (even though these are for different periods in 2002–2004, 2012 and 2014) supports the modelling approach used in this study.

The spatial distribution of PNC presented in Fig. 10 was used to estimate the exposure of the population in Amsterdam-Amstelveen. The grid cells of 50*50 m for the spatial distribution were matched with the number of addresses (e.g. residential, shops and offices) in these cells. These addresses and their geographical locations were derived from the national addresses database available in the Netherlands. The results for the annual average ('long-term' exposure) and during specific periods in which the wind was blowing from the direction of Schiphol ('short-term' exposure) are shown in Fig. 11. The latter accounts for 14% of the hours in 2012.

Fig. 11 shows that a considerable number of the total of 555,000 addresses in the modelling domain of 20*20 km in Amsterdam-Amstelveen (see: Fig. 10) were exposed to elevated PNC from emissions at Schiphol: for 480,000 addresses less than 5000 particles per year and for 75,000 addresses more than 5000 particles per year. For example, in 2012, 45,000 addresses suffered *long-term* exposure to an additional, annual background PNC of 5–7500 # cm^{-3} from Schiphol, while 60,000 addresses suffered *short-term* exposure (i.e. 14% of the time) to additional 10–15,000 # cm^{-3} from Schiphol.

4. Discussion and conclusion

From this study, it was concluded that in particular air traffic contributes to elevated PNC downwind of Schiphol but not to the mass concentrations of black carbon. At the sites Adamse Bos (7 km downwind) and Cabauw (40 km downwind), PNC was elevated by a factor of 3 and 20% downwind of Schiphol, respectively, compared to other wind directions. The size distribution of PNC downwind of the airport at Adamse Bos was dominated by particle sizes of 10 nm–20 nm, while at Cabauw the size distribution downwind of Schiphol shifted to larger particles of 20–40 nm. The size distribution of PNC downwind of Schiphol is typical for combustion emissions (Kittelson et al., 2004) but the chemical composition and PN emissions per kg fuel differ with the type of fuel. In particularly, the sulphur content of (fossil carbon-based) fuels is important: PN emissions increase exponentially with the sulphur content and also affects the chemical composition (Maricq, 2007). In kerosene, the average sulphur content is about 400 mg per kg (Kinsey et al., 2010), which is considerable lower in fuel for road traffic with less than 10 mg per kg diesel in Europe. Consequently, PN emissions from jet engines are a mixture of sulphuric acid, EC and OC (Kinsey et al., 2010; Vander Wal et al., 2014), while PN emissions from road traffic contains contain relatively less sulphuric acid but are mainly a mixture of lubrication oil, EC and OC (Sakurai et al., 2013; Maricq, 2007). Take-off and climb-out from the Kaagbaan and Aalsmeerbaan runways, followed by taxiing and waiting at the gates, and landing at the Buitenveldertbaan runway were identified as the four most important sources of elevated PNC downwind of Schiphol at the Adamse Bos. The significance of PN emissions from take-off and climb-out at Schiphol is in agreement with other studies (e.g. Mazaheri et al., 2011). Other runways (i.e. Polderbaan and Zwanenburgbaan in the northerly direction) are also frequently used for take-off at Schiphol. However, the prevailing wind directions during the use of these runways does not transport PN to Adamse Bos. Measurements downwind of these runways are recommended to investigate the contribution of these sources to PNC in other urban areas near Schiphol.

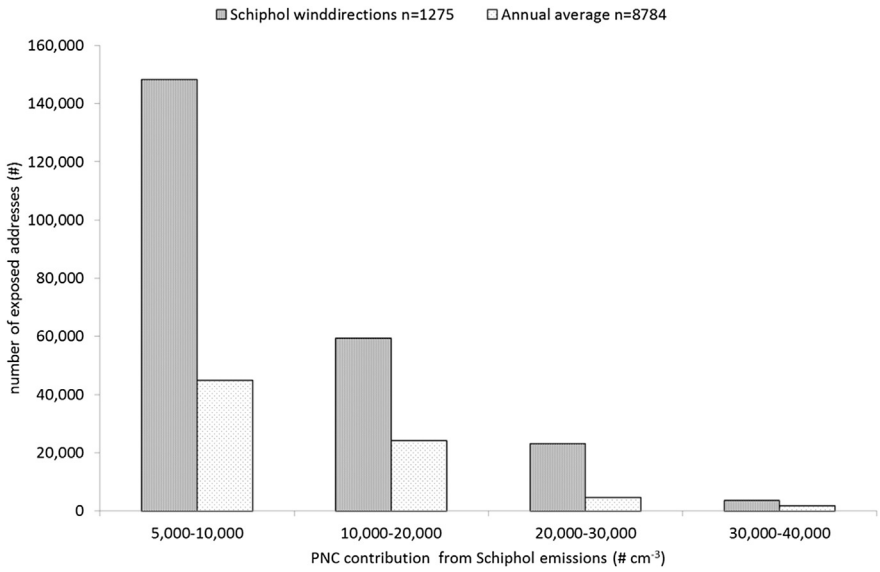

Fig. 11. Exposure of the population to PNC (# cm^{-3}) from Schiphol emissions: when the wind was blowing from the direction of Schiphol (grey) and the annual average in 2012 (light).

A Gaussian Plume model was used to model the dispersion of PN from the airport to urban areas northeast of Schiphol in Amsterdam and Amstelveen. The modelling was based on the assumption that PN would act as an inert pollutant, while neglecting atmospheric processes such as coagulation, condensation/evaporation and dry deposition. The first two processes are particularly important when combustion emissions initially cool down in ambient air, while dry deposition is a more important process during dispersion of PNC over longer distances (Ketzel and Berkowicz, 2004). In this study, a conversion factor for relating modelled concentrations of X (as a tracer for PN) in μg m^{-3} to measured PNC in # cm^{-3} was established at Adamse Bos 7 km downwind of Schiphol. Consequently, coagulation and condensation/evaporation were considered less important processes during dispersion of PN further downwind from Adamse Bos to the urban areas of Amsterdam-Amstelveen. A second assumption was that meteorological conditions and air traffic intensity during the monitoring campaign in 2014 were representative for the modelling year 2012. This is a likely assumption as shown as follows. The frequency of the wind directions from Schiphol (240–270°) were 12% during the monitoring period in 2014 and 14% in 2012. The air traffic only changes a few percentage from year-to-year and hardly varies from month-to-month within a year (Schiphol, 2013). The agreement between the measured contribution in 2002–2004 at the Tropenmuseum and in 2014 at Adamse Bos supported the modelling approach, including the assumptions of dispersion of PN as an inert gas and the representativeness of the monitoring period in 2014 for modelling the dispersion for a whole year. Still, more PNC measurements up and downwind from Schiphol are recommended to further validate the outcomes of the dispersion modelling. In addition, more specific information on the use of runways (e.g. number of take-offs, type of plane) may further improve research on the contribution of different sources at Schiphol.

The exposure of the population in urban areas of Amsterdam and Amstelveen was estimated from the results of the dispersion modelling for 2012. More than 200,000 addresses were found to be exposed to increased annual average and hourly average PNC due to Schiphol emissions. There are no air quality standards for PNC to evaluate these elevated levels, and the potential health effects due to this exposure are unknown, as no reliable concentration-response-function for PNC is available. In view of the growing number of studies indicating elevated levels of PNC in urban areas near airports and the suspected health effects of exposure to elevated PNC, it is recommended that further research on potential health effects be carried out at and near airports.

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