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Assessment of the applicability of a model for aviation-related ultrafine particle concentrations for use in epidemiological studies

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HIGHLIGHTS

- High ultrafine particle number concentrations (PNC) are found near Schiphol Airport.
- A dispersion model for aircraft emissions is evaluated using measurements of PNC.
- The analysis includes road traffic and background contribution to PNC.
- The model is suited to study effects of long-term exposure to UFP from aviation.

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ABSTRACT

Background and objectives: In recent years it has been shown that aircraft emissions are a dominant source of ultrafine particles in the surroundings of airports. However, health effects of long-term (monthly to yearly) exposure to these particles are unknown. As part of an integrated research program into the health risks of ultrafine particles around Schiphol Airport, the applicability of the dispersion model STACKS+ to assess long-term exposure to ultrafine particles from aviation was assessed.

Methodology: A detailed comparison between modelled and measured particle number concentrations (PNC) due to aircraft emissions was carried out at ten locations in the surroundings of Schiphol Airport during two six-month periods in 2017 and 2018. In order to deduce the contribution of aviation to measured PNC, we applied a fitting method of the sum of the modelled contributions from aviation, the modelled contributions from traffic on main roads and the contributions from outside the study area estimated from the measurements, to the measured total PNC. The analysis yielded scaling factors and uncertainty estimates for each of the main contributions. We then subtracted the estimated background and modelled contributions of road traffic from the total measured PNC and took the remainder as an approximation of the measured contribution from aviation to PNC. We compared it to the modelled contribution from aviation, based on the averaged values for the six-month periods.

Results: Both six-month averaged modelled and measured PNC due to aircraft emissions (i.e., adjusted for background) showed a large range at the monitoring locations representative for population exposure (from close to zero to 10 000 particles/cm^3). Spearman and Pearson correlation coefficients between model and measurement results were high (>0.83).

Conclusions: The applied approach enabled us to obtain a robust estimate of the contribution of aviation to the measured PNC. The dispersion model is able to determine the spatially varying average concentrations due to

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aircraft emissions in residential areas over periods of 6 months, allowing for application in epidemiological studies into long-term exposure.

1. Introduction

Ultrafine particles (UFP) are defined as airborne particles smaller than 100 nm (0.1 μm) in aerodynamic diameter. Their composition is variable, containing organic compounds, black carbon and metals and they can be volatile (Stacey, 2019). Particle number concentration (PNC) is often used as an indicator for UFP, since the vast majority of airborne particles is smaller than 100 nm (Hinds, 1999). Stacey (2019) and Riley et al. (2021) presented literature reviews on measurements of UFP at and near airports. They show that results from multiple measurement campaigns performed for airports worldwide indicate that aircraft emissions are a dominant source of UFP at or close to airports (a. o. Westerdahl et al., 2008; Hudda and Fruin, 2016; Masiol et al., 2017). At distances up to 10 to even 40 km (according to Keuken et al., 2015), the influence of aircraft may still be detected above background levels when the location is downwind of the airport. Schiphol Airport is the main airport in the Netherlands with around 500 000 air transport movements and 80 million passengers in 2018 (Schiphol Group, 2019). Research into the concentrations of UFP around Schiphol Airport also revealed increased concentrations due to aircraft emissions (Keuken et al., 2015; Bezemer et al., 2015 (the latter in Dutch)).

Little is known about the long-term health effects of UFP, in particular from aviation. A systematic literature review by Ohlwein et al. (2019) identified only ten studies on long-term effects of UFP, with various health outcomes. None of these studies focused on UFP from aircraft emissions; therefore, studies on the long-term health effects of UFP for airport-related exposure are needed (Umweltbundesamt, 2018).

Research into the health effects of long-term exposure to UFP due to aircraft emissions requires information about the exposure of people living in the vicinity of airports. This is not feasible using measurements only. Modeling techniques are needed to estimate long-term averaged concentrations caused by aircraft emission over larger areas.

Different types of dispersion modelling methods have been applied for the assessment of the contribution of aircraft emissions to local concentrations of, for example, particulate matter (PM_{10}, $PM_{2,5}$) and nitrogen oxide (NO_x). According to Barrett et al. (2013), many of the models applied for aviation do not take into account aircraft plume dynamics in sufficient detail. They found that, according to a study at Heathrow Airport, this could result in an overestimation by a factor of 1.3–2.3 of the mean NO_x concentration depending on the location. They present a simplified concentration correction factor for dispersion models with a passive plume approach i.e., no plume dynamics and motion. This approach is assumed valid for distances larger than 1 km.

Models that take into account plume dynamics include the ADMS-Airport model (Carruthers et al., 2007) and the LASPORT model (Janicke Consulting, 2011). ADMS-Airport employs a quasi-Gaussian dispersion model nested within a trajectory model (Carruthers et al., 2007), in which aircraft sources are treated explicitly as accelerating jet sources. LASPORT is a Lagrangian model. In the Netherlands, the Gaussian dispersion model STACKS+ is frequently used for the dispersion of aircraft emissions for environmental impact studies. It was developed from the STACKS model for point or surface area sources (Erbrink, 1995a). Some changes were made to better represent the aircraft exhaust circumstances relating to plume rise and initial dispersion. However, it does not take into account more complex aircraft plume dynamics.

Dispersion modelling of UFP has only recently been started at some airport locations, often following monitoring campaigns in the near field to adjust model results. Keuken et al. (2015) used a Gaussian plume model intended for point and surface area sources to estimate concentrations around Schiphol Airport. In an exploratory study, Bezemer et al. (2015) applied the STACKS+ Gaussian dispersion model to aircraft emissions at Schiphol Airport. Modelling of PNC in 2015 around Brussels Airport (Belgium) was performed using the IFDM bi-gaussian plume model (Lefebvre et al., 2019; in Dutch). They applied the simplified correction factor approach by Barrett et al. (2013) on the aircraft emissions to account for the wake turbulence. Average PNC levels in 2015 at and around Frankfurt Airport (Germany) were estimated using a combination of LASPORT and large-scale modelling (Lorentz et al., 2019). Wing et al. (2020) used the US EPA AERMOD Gaussion plume model to estimate exposure of residents near Los Angeles International Airport (LAX). The AERMOD model was also used in a study into concentrations of several pollutants including UFP near Chania Airport in Greece (Makridis and Lazaridis, 2019). Zangh et al. (2020) applied a Lagrangian dispersion model fit for complex terrains like the inner-Alpine basin in which Zurich Airport is situated.

Some of these studies only modelled the dispersion of aircraft emissions (e.g. Keuken et al., 2015; Bezemer et al., 2015; Wing et al., 2020; Zangh et al., 2020), while others also took into account road traffic in the surroundings of the airport and background levels (e.g. Lefebvre et al., 2019; Lorentz et al., 2019). The studies use different approaches to account for the emission factors of particle numbers for the different aircraft flight phases. Lorentz et al. (2019) used data from the ICAO Engine Emission Databank, yielding emission factor estimates for the non-volatile particles only. However, it is well known that the majority of the UFP close to aircraft sources are volatile and form by nucleation or condensation (Beyersdorf et al., 2014). Therefore others use emission factors derived from monitoring campaigns in the field (Lefebvre et al., 2019; Winther et al., 2015; Zangh et al., 2020). The Dutch national aircraft emission database does not contain emission factors for particle number. Therefore, the recent Dutch studies used an arbitrary, total emissions strength of 1 kg/s (Keuken et al., 2015) and emission factors for PM_{10} (Bezemer et al., 2015) as a proxy to calculate concentrations due to aircraft emissions and translated them to PNC using measurements of PNC at several locations.

As part of an integrated research program into the health risks of UFP around Schiphol Airport, the applicability of the dispersion model STACKS+ to assess long-term (monthly to yearly) exposure to UFP from aviation was assessed. In a previous exploratory study (Bezemer et al., 2015), STACKS+ was used for the first time to model PNC due to aircraft emissions. In that study, emission factors from PM_{10} were used and the results were calibrated using measurements of PNC, conducted with different types of monitors during four to six weeks campaigns at ten sites. In this context, we use the term calibration for adjusting model results based on the comparison with measurements. In the current study, the model was enhanced by adding taxiing aircraft and applying emissions factors for PNC from literature. The investigation of the model performance was enhanced by using identical condensation particle counters and extending the measurement period. Furthermore, we applied a new approach to deduce the contribution of aviation to measured PNC, explicitly quantifying contributions from the background, road traffic and contributions from aviation.

The objective of the present study is to assess whether the model for dispersion of aviation-related ultrafine particles adjusted to measurements is suitable for long-term aviation-related PNC exposure estimation in support of epidemiological studies. We do this by comparing the correlation between six-month averaged measured and modelled contributions from aircraft emissions to PNC to correlations used in other epidemiological studies, taking into account the contrast in aviation related PNC within the study area. We used a dispersion model employed in de Dutch national air quality management system (Wesseling et al., 2011), to estimate the NO_x concentration contributions from road traffic as a proxy for the contribution from road traffic to PNC, in order to deduce the PNC contribution from aviation from the total measured PNC concentrations. In the epidemiological studies on the health effects of long-term exposure to UFP from aviation, that will be published separately, we will adjust for exposure to other (traffic-related) pollutants, by running multi-pollutants models with $PM_{2.5}$, NO_2 and EC (annual average concentrations from dispersion modelling).

2. Methodology

2.1. Approach

We performed measurements of PNC at 10 locations in the area around Schiphol Airport during two six-month periods. Details about the measurements are described in Section 2.2. In order to investigate the model performance for aviation-related UFP, the contribution of aviation needs to be deduced from the total measured PNC. This is not straightforward since the total PNC at any location consists of contributions from many (types of) sources. Therefore, for the present analysis we fitted the sum of the main contributions (aviation, road traffic and contributions from outside the study area) to measurements of total PNC in the study area around Schiphol. We do not base the fit only on the 10 average concentrations at the measurement locations; instead, the fit is based on the concentration estimates for all 10-degree wind direction intervals at each of the measurement locations. So, we had 36 average measurement values at each of the measuring locations. The combined analysis of total PNC results in scaling factors and uncertainty estimates for each of the main contributions. The main contributions of UFP that were considered are.

- Measurement-based estimated background PNC from outside the area;

The background PNC was estimated by selecting wind directions with air coming from outside of the study area for the specific measurement locations near the outer boundaries of the study area (see section 2.2). The PNC measurements were aggregated in 10-degree wind direction intervals. This enabled us to select the data coming from wind directions from outside of the study area. This is described in more detail in Section 3.2.

- Modelled contributions from aviation;

The contributions from aviation activities were calculated using STACKS+. The model and its input data are described in detail in Section 2.3.1.

- Modelled traffic contributions from within the study area;

Finally, the contributions from road traffic on main roads to PNC were estimated by using NO_x concentrations calculated using a model specific for traffic contribution as a proxy with a conversion factor (from NO_x to PNC) fitted to the measurement data. The model is described in more detail in Section 2.3.2.

The combined analysis is followed by a correlation analysis for the model for aviation. For application in epidemiological studies of long-term exposure to UFP, the most critical requirement for a model is to correctly rank locations in terms of average concentration. The reason is that in epidemiological studies basically the health status of subjects living in locations with high and relatively low concentrations are compared (on a continuous scale). Hence, the correlation coefficient between model and measurement is the first key performance indicator of a model for epidemiological application. We investigated the independency of the main contributions, allowing to interpret the difference between the measured PNC and the sum of the estimated backgrounds and modelled traffic contributions as the "measured" contribution of aviation. The estimated "measured" contribution of aviation is then compared to the modelled contribution of aviation yielding correlation coefficients.

2.2. Measurements

For the model performance evaluation, it is important to have a number of measurement locations representing the spatial variation in resident exposure. In epidemiological studies into long-term exposure, periods of months (e.g. for effects on birth outcomes) or years (e.g. for effects on mortality) are used. As we had six identical environmental particle counters TSI Incorporated type EPC-3783 available for a one-year period, we decided to balance the number of locations and measurement period by measuring at ten locations at different distances and orientations from Schiphol Airport (Fig. 1) during two six-month periods (August 2017–January 2018 and March–August 2018). Two monitors were placed at locations to the south and north of Schiphol Airport and operated continuously for 12 months (locations 1 and 2); the other four monitors were placed at different locations in the surroundings for six months and moved to four new locations during the second period of six months. Locations 1, 2, 6, 9 and 10 are existing monitoring locations part of the air quality monitoring network managed by the Public Health Service (GGD) of Amsterdam; the other locations were set up specifically for the study. Locations 6 and 7 are not situated in residential areas but rather close to the runways. These locations were added to increase the variation in distance towards the runways. Especially location 6 in Fig. 1 can be highly exposed to aircraft emissions. With wind coming from northerly directions, aircraft depart from the frequently used runway "Polderbaan" just 400 m away from location 6. Location 7 is also situated within 1 km from a runway ("Buitenveldertbaan"). However, due to low use of this runway, location 7 is far less exposed. Further details on the measurement locations are presented in Table 1. Distance to the nearest runway ranged from 400 to 5600 m. We did not include monitoring sites further away as we wanted to determine the UFP levels at locations where the contributions from aircraft emissions to concentration levels were expected to be measured with high enough certainty.

The EPC-3783 water-based condensation particle counter measures the total number of airborne particles with a diameter of 7 nm or more ($D_{50} = 7$ nm). At the existing air quality monitoring locations, the particle counters were placed inside air-conditioned cabins. At the temporary locations, the particle counters were placed inside small, heated and ventilated shelters equipped with identical inlets. At ambient temperatures over 25 °C, there were some periods in which the particle counters inside the shelters did not function properly. All monitors used identical non-conductive inlets with a length of 1.5 m to sample outdoor air. Humidity control or correction for diffusion losses was not applied. The saturator wick was exchanged every three weeks. PNC was measured on a 1-min average basis. Hourly averages were derived from the raw data. Only hours with at least 45 min of data were used in the analysis.

As part of the quality control, we performed three side-by-side comparisons of the six EPC's: 1) before the measurement period, 2) halfway, between both six-month periods and 3) after the second six-month period. The EPC's were brought in from the field to location 6, close to the runway "Polderbaan", ensuring large variation in PNC. The measurements were carried out in the same configuration (housing, inlet) as in the field. The monitor with the readings closest to the average readings of all monitors at the start of the study was chosen as the reference monitor. For the calibration of the other five monitors to this reference monitor, linear regression analysis was performed on the hourly measurement data. Within one side-by-side comparison period, the individual monitors differed between 0 and 14% (when operated without malfunction). Between the side-by-side comparison periods, changes in the calibration factors of up to 10% were observed. A correction was applied to account for this drift using linear interpolation between calibration factors before and after the six-month periods. The comparative measurements and detected relationships are described in

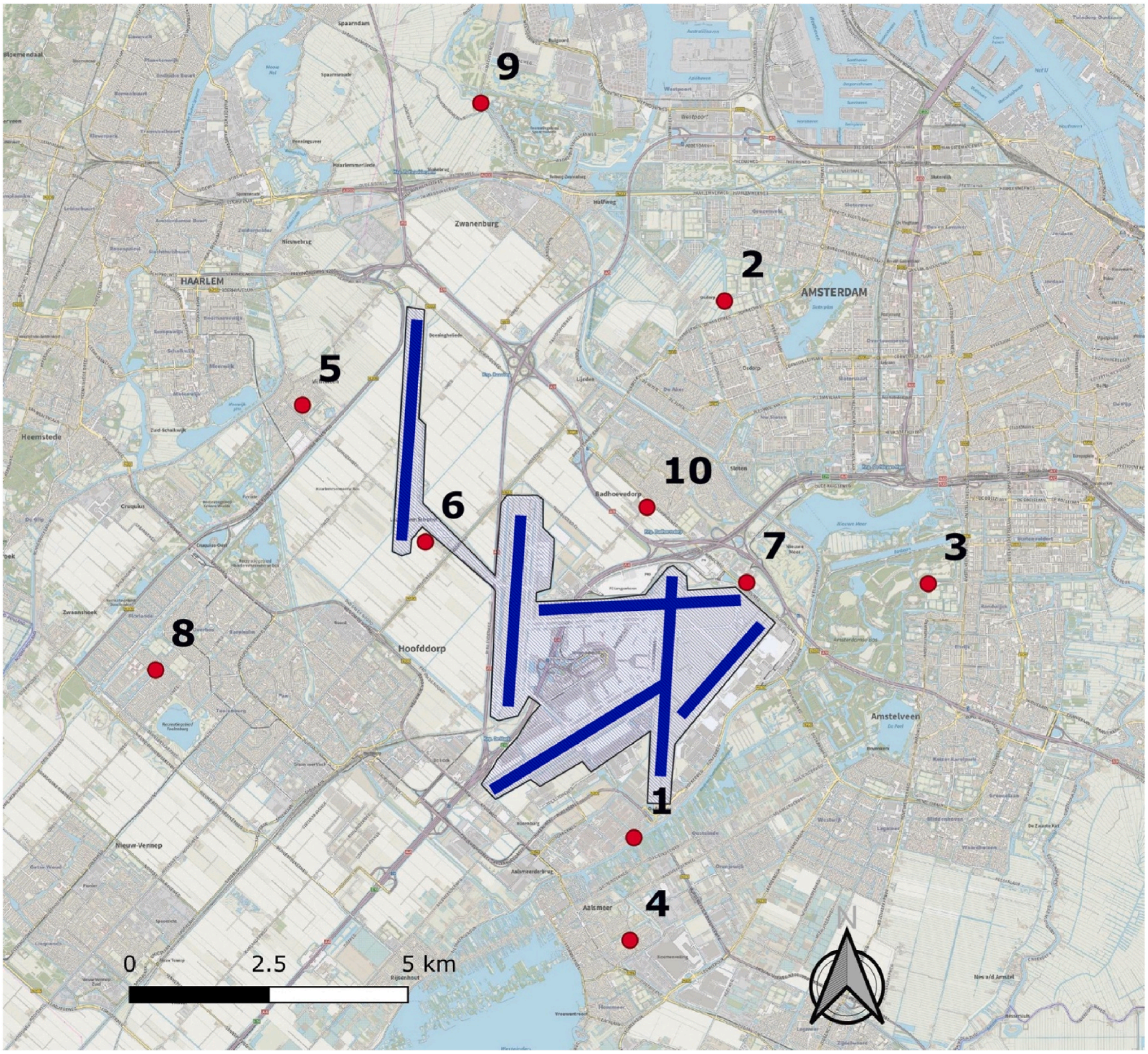

Fig. 1. Measurement locations. Temporary locations (nr 3, 4, 5, 7 and 8) are set up specifically for this study; existing locations (nr 1, 2, 6, 9 and 10) are part of the air quality monitoring network managed by the Public Health Service (GGD) of Amsterdam.

the Supplemental Materials.

During the field campaigns, there were periods in which some of the instruments did not function (properly). Data availability per measurement location is given in Table 1. The overall measurement data availability was about 90%, ranging from 66% to 100% for the individual locations.

2.3. Dispersion models and input

2.3.1. Aviation

The STACKS+ dispersion model is a frequently used model for the dispersion of aircraft emissions in the Netherlands. It has been used in the past for air quality calculations for various Environmental Impact Assessment studies about both Schiphol Airport and other airports (among others Hoolhorst et al., 2016; in Dutch). The underlying STACKS model is a Gaussian plume model intended to describe the dispersion of emissions from point and surface area sources (Erbrink, 1995a, 1995b). It is based on Monin-Obukhov boundary-layer scaling in combination with Taylor dispersion theory and uses amongst others sensible heat flux, friction velocity, Monin-Obukhov length, turbulence intensity and time scales, mixing height, vertical profiles of wind speed, wind direction and temperature. For STACKS+, some changes were made to better represent the aircraft exhaust conditions: (1) the description of plume rise due to the heat emission takes into account both aircraft and wind speed; and (2) dilution caused by the jet impulse is represented by an initial size of the plume with an initial spread/dispersion of 10–20m. However, the model does not take into account more complex aircraft plume dynamics, like horizontal displacement of the exhaust stream due to the high exhaust velocity or vertical displacement of emissions due to turbulence indicated by the aircraft (the effect of wake vortices as investigated by Unterstrasser et al., 2014). Several tests were performed varying the effect of the downwash by introducing an effective change in the height of the emissions. The agreement between measurements and model results did not improve. Therefore, the default setting of the model was used.

The model describes the dispersion of aircraft emissions through the atmosphere on an hourly basis. For this study, hourly values were averaged over the six-month measurement periods. The uncertainties in the hourly modelled contributions to the concentration by aircraft are relatively large due to small scale variations in the width and height of the plume. The model gives lower uncertainties when the results are averaged over longer periods. For the specific purpose of testing the dispersion model for aviation, at all locations the hourly values for which the measurement was available and the wind speed was 1.5 m/s or higher were taken into account in both the measurements and modelled aviation-related PNC. The latter is because wind directions

Table 1
Measurement location details.

Location	Monitoring period	Distance to nearest runway (m)	Distance to nearest major road (m)	Wind direction sector for estimation of regional background (degrees)	Measurement data availability (%)
1 Oude Meer	Period 1 (Aug 2017–Jan 2018)	1300	400	–	92
2 Ookmeer	Period 1 (Aug 2017–Jan 2018)	5100	2000	320–140	98
3 Amstelveen	Period 1 (Aug 2017–Jan 2018)	3500	1500	10–150	99
4 Aalsmeer	Period 1 (Aug 2017–Jan 2018)	3200	500	30–250	83
5 Vijfhuizen	Period 1 (Aug 2017–Jan 2018)	1900	600	220–360	86
6 Polderbaan	Period 1 (Aug 2017–Jan 2018)	400	1300	–	79
1 Oude Meer	Period 2 (Mar–Aug 2018)	1300	400	–	96
2 Ookmeer	Period 2 (Mar–Aug 2018)	5100	2000	320–140	99
7 Nieuwe Meer	Period 2 (Mar–Aug 2018)	500	100	–	66
8 Hoofddorp	Period 2 (Mar–Aug 2018)	5600	700	190–30	82
9 Spaarnwoude	Period 2 (Mar–Aug 2018)	4200	1200	320–50	100
10 Badhoevedorp	Period 2 (Mar–Aug 2018)	1500	50	–	96

might not be well defined when wind speed is low due to enhanced horizontal wind meandering at low wind speeds (Mortarini et al., 2016). Furthermore, the assumptions underlying Gaussian dispersion modelling become less valid at lower wind speeds. Applying this criterion excludes 8–10% of the hours in the measurement periods. In the actual epidemiological study, reported separately, the estimated contributions from the aircraft, calculated using the actual emissions and meteorological conditions in each year, are used. For the dispersion calculations in the epidemiological study, a lower limit for the wind speed of 1 m/s is used, as is prescribed for Dutch Gaussian models, like STACKS+.

Actual detailed flight movements are used to feed the dispersion model. The actual number of aircraft arrivals and departures per runway per hour, come from the FANOMOS database from Air Traffic Control The Netherlands (LVNL), made available by the Netherlands Aerospace Center (NLR). The model assumes an average length of the runway of 2 km, with a climbing slope of 9% and a descending slope of 4% for all (types of) aircraft. The sensitivity to different choices for the length of the runway was not investigated. The emissions from the aircraft are gridded in cells 500 × 500 × 250 m (LxWxH). The highest (average) concentrations were modelled at the locations of the take off. The path of the aircraft is a straight line over a distance of 12.5 km. For climbing aircraft this equates to around 1.1 km height and for approaching aircraft to a height of around 500 m. Applying STACKS+, particles or gases that are emitted around 500 m or higher do not or hardly reach the ground due to dilution in the atmosphere. Moreover, the emissions in the dispersion model often take place above the mixing layer of the atmosphere, so that they do not end up in the lower air layer. Further details of the modelling approach are given by Erbrink Stacks Consult, 2019) (in Dutch).

The model is applied on the assumption that UFP – once they are emitted from the aircraft exhaust and are formed by nucleation or condensation in the first moments after emission in the cooler ambient air (Timko et al., 2013; Beyersdorf et al., 2014) - disperse within the study area as an inert substance. This means that the substance itself does not undergo any changes but is only diluted through the atmosphere. We are aware that this is a simplification, since processes like (ongoing) nucleation and condensation, evaporation, coagulation and deposition probably play a role at the scale of our study area. However, apart from emission and near source nucleation and condensation, dilution is assumed to be the dominant factor for the change in PNC with increasing distance relative to ultrafine particle sources (Kumar et al., 2011).

Meteorological observations performed at Schiphol Airport have been made available by the Royal Netherlands Meteorological Institute (KNMI). The two measurement periods were quite different with respect to the meteorological circumstances. As indicated in Fig. 2, the first period had very few hours with wind from the north and east. During the second period the wind direction was more evenly distributed. Furthermore, the second period was on average warmer (15.4 vs 10.6 °C) and drier (223 vs 612 mm precipitation) compared to the first period. The meteorological conditions in the full measuring period, august 2017–august 2018, are quite similar to that of the longer period 2001–2020, wind data are shown in the Supplemental Materials.

Emission factors for UFP were derived from a field study performed under real-world conditions at Brisbane Airport (Brisbane, Australia) using measurement equipment with the same D_{50} value as our equipment (Mazaheri et al., 2009). From co-located measurements of carbon dioxide (CO_2), they estimated emission factors for taxiing, and idling, taking off and landing. The representativeness of the fleet in Brisbane Airport more than 10 years ago (mainly Boeing 737) for the present situation at Schiphol Airport is not exactly known, and there were no emission factors derived for climbing and approaching aircraft. Therefore, an estimate was made for the situation at Schiphol from the emission factors presented by Mazaheri et al. (2009). The emission factors for the B737 and B767 types, that had the highest number of measured aircraft (207 and 40 respectively) in their study, were

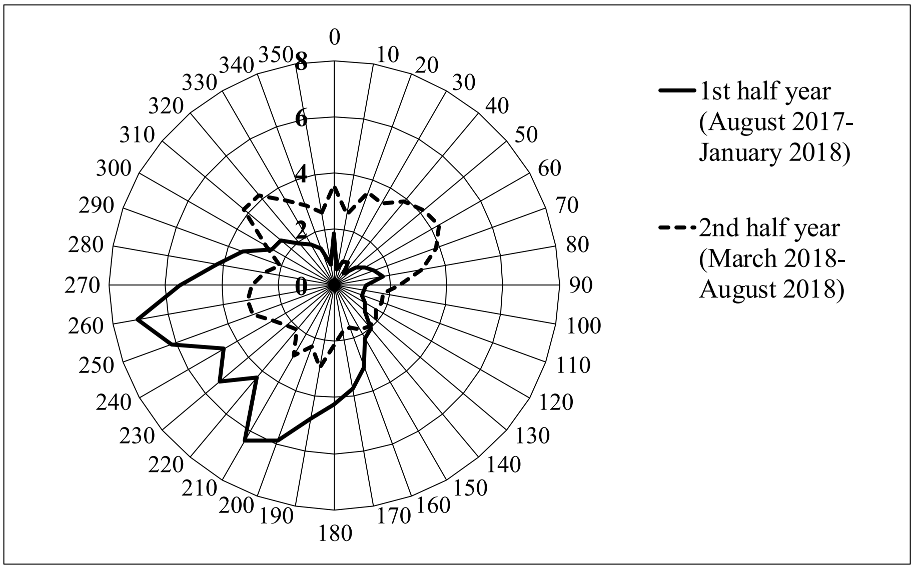

Fig. 2. The distribution of wind direction in % for both periods (0 = wind from the north, 90 = wind from the east, 180 = wind from the south, 270 = wind from the west).

averaged weighted by the number of measured aircraft. It was assumed that emission factors (in number per kg fuel) for climbing/approaching were the same as for taking off/landing. The emission factors are presented in Table 2, also expressed in number per second, taking into account differences in the amount of fuel consumed in the different flight phases.

We also investigated the use of emission factors for Particulate Matter (PM_{10}) and NO_x as proxies, provided by NLR retrieved from the national aircraft emission database. However, the physical formation of UFP in aircraft emissions is likely not very similar to the formation of PM_{10} or NO_x and initial analyses showed that the emissions factors from Mazaheri et al. (2009) showed slightly better agreement with the measurements (Voogt et al., 2019).

2.3.2. Road traffic

The NO_x concentration contributions from the traffic on the main roads were used as a proxy for the contribution from road traffic to PNC. As we did for the aircraft emissions, the model is applied on the assumption that UFP, once they are emitted from the traffic and are formed by nucleation or condensation in the first moments after emission in the cooler ambient air, disperse within the study area as an inert substance. For the analysis of the UFP measurements and model results, a linear relation is assumed between the NO_x concentration contributions from traffic and the PNC contributions from traffic. By fitting the calculated NO_x contributions to the estimated measured PNC contributions, the (linear) conversion from NO_x to PNC is estimated. This assumption introduces uncertainties that are discussed later. The NO_x contributions were calculated using the model employed in the Dutch national air quality management system, "Standard Calculation Method 2" (Wesseling et al., 2011; Wesseling and van Velze, 2014; Wesseling et al., 2016). For the yearly national air quality assessment, all local authorities must provide detailed traffic data (numbers and types of cars, speeds, stagnation, road details, etc.). Emission factors for the Dutch mix of cars are estimated on a yearly basis and reported by the Ministry of Infrastructure and Water management. With these data it is possible to calculate reasonably accurate contributions from road traffic for (among others) NO_x (Wesseling et al., 2016).

In contrast to the standard calculations of air quality in the Netherlands, the contributions from roads up to 7 km from the measurement location have been calculated. In standard calculations for air quality management this is 4 km, as the effects of emissions from further away are taken into account in a background model. In the present analysis we do not use a separate model for the background but use actual measurements. The 7 km chosen here is a balance between completeness of the emissions on the roads and too much double counting in the background.

As the dispersion model calculates the concentration contributions from traffic on a yearly average basis, it was not possible to estimate separate sets of NO_x contributions for the two measuring periods. The yearly average concentration contributions from traffic, calculated for 2018 and averaged per 10-degree wind direction interval, were used as estimates for both measuring periods. This creates some additional uncertainty.

Table 2
Flight phase estimates of fuel consumption and emissions of ultrafine particle numbers estimated from Mazaheri et al. (2009).

Flight phase	Fuel (kg/s)	UFP (#/kg)	UFP (#/s)
Taxiing	0.24	$3*10^{16}$	$7.2*10^{15}$
Landing	0.32	$4*10^{16}$	$1.3*10^{16}$
Approaching	0.80	$4*10^{16}$	$3.1*10^{16}$
Climbing	2.22	$5*10^{16}$	$1.1*10^{17}$
Taking off	2.80	$5*10^{16}$	$1.4*10^{17}$

3. Results

3.1. Measurements

The measured PNC for each location and period, averaged per 10-degree wind direction interval are presented in Fig. 3 as pollution roses. The measured PNC at the outer boundaries of the study area are shown in red and the results at locations in the center of the area are presented in blue. In Fig. 3, we aim to show the main wind directions of the measured PNC contributions. It is evident that at all locations the PNC levels increase when the wind is coming from the direction of aircraft activity related to Schiphol Airport. Note that the plots for the individual pollution roses have different scales, since the scale needed for the highest levels at location 6 would eliminate details in the other figures. These scales are not shown in Fig. 3 but can be found in Figure S7 of the Supplemental Materials. For the locations in the center (presented in blue), highest concentrations in the downwind sector ranged from 45 000 to 180 000 particles per cm^3. At the outer locations (presented in

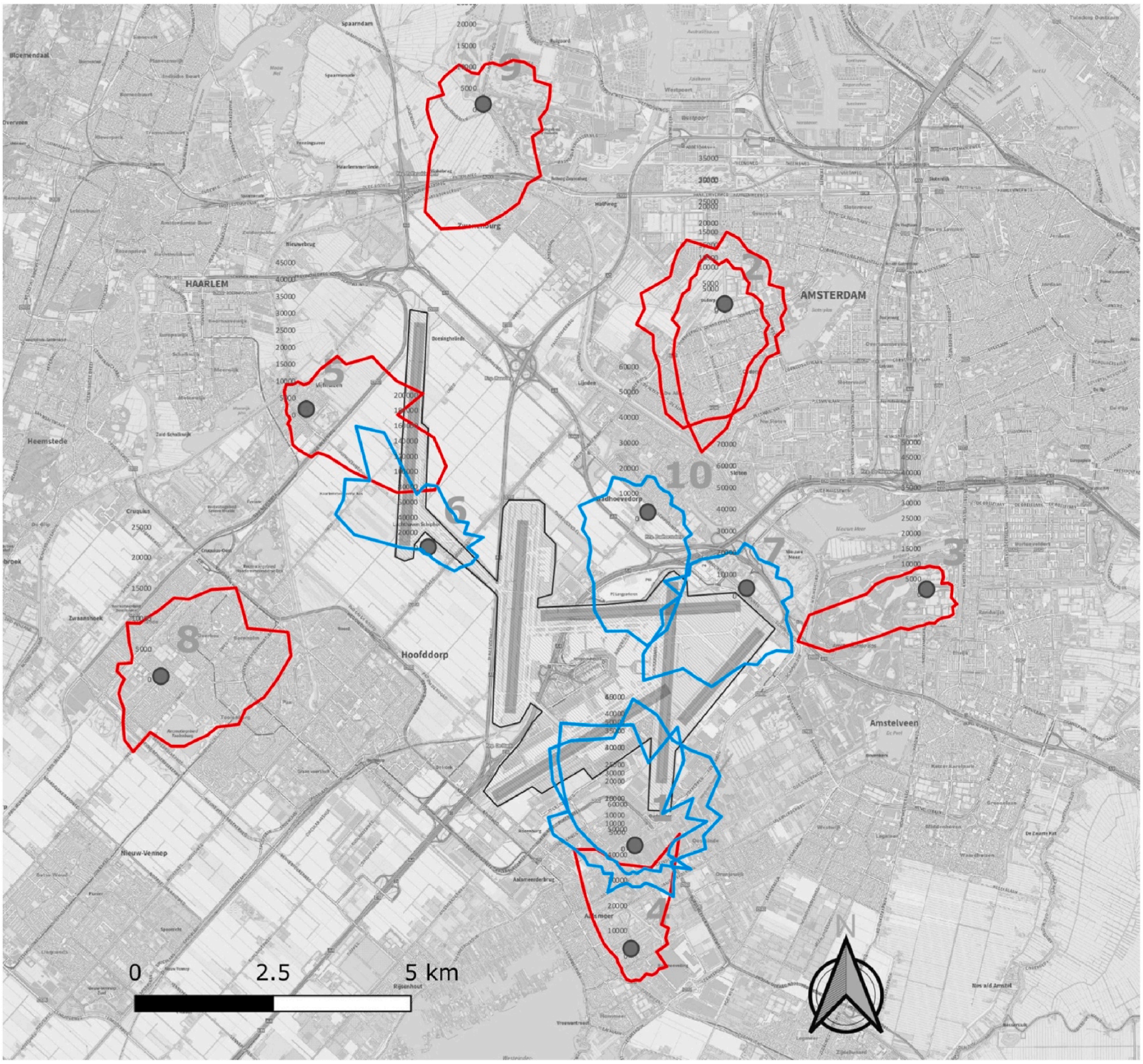

Fig. 3. Six-month averaged measured PNC (particles/cm^3) per 10-degree wind direction interval. Note the plots have different scales. The pollution roses shown in red are those of the “outer stations”.

red), the range is 22 000 to 50 000 particles per cm^3.

3.2. Estimated PNC background

During the study, the outer measurement locations were located in a wide ring around Schiphol, see Figs. 1 and 3. This makes it possible to determine the contributions of UFP from outside the study area for the different wind direction intervals. For every location in the outer ring, the wind angles were selected where the air was coming from outside the study area. Wind directions from traffic on nearby busy roads were not considered as background. In other words, there is limited “double counting” between the background determined in this way and the separately estimated contributions from traffic and aviation. Due to the positioning of the outer locations, there is some overlap in background-directions. For instance, at the locations 5 and 8 the wind directions of 180–360 are treated as background. The averaged PNC, plotted as a function of the wind direction, is shown in Fig. 4.

For most of the wind directions shown in Fig. 4, there are several locations where background concentrations from that direction were measured. In general, the differences between these measurements are limited, neighboring locations in the outer ring of stations observe roughly the same background concentrations. The largest variation in background PNC is seen for easterly wind directions, from the area where the cities of Amsterdam and Amstelveen are located. For the analysis, we have taken the average value for each wind direction.

3.3. Model results for road traffic

Contributions from road traffic were calculated for each 10-degree wind direction interval for each measuring location in the measurement campaign. As the measurement periods do not cover an entire calendar year and the model for road contributions is optimized for annual average traffic contributions, results obtained for 2018 are used in all analyses (see Methods section 2.3.2).

At a number of measurement locations, substantial traffic contributions are expected in certain wind directions, with only limited contributions from aviation. An example is measurement location 7 “Nieuwe Meer”, where aviation contributions are expected from southern, western and northwestern directions, but hardly or not at all from the other directions. On the other hand, substantial traffic contributions are expected from the north and the east. We compared the calculated NO_x contributions with the measured PNC for all location/wind direction combinations with low or no aviation contributions (calculated PNC from aircraft <50 particles per cm^3) and with high enough NO_x

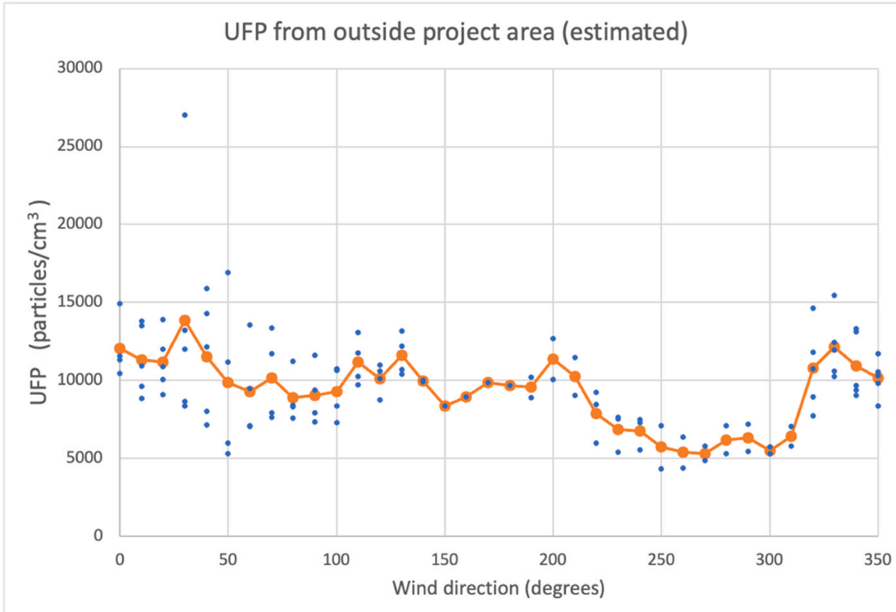

Fig. 4. Averaged PNC (particles/cm^3) per 10-degree wind direction interval when the wind comes from outside of the study area, shown in blue. The orange curve is the mean of the background values at the selected locations. The data covers the whole period of the measurements.

contributions from road traffic (calculated NO_x from road traffic >2 μg/m^3), to determine the correlation between the modelled NO_x and measured PNC from traffic. With these criteria, data from the locations 2, 3, 4, 5, 7 and 10 were used in the analysis. The sum of the background concentrations and the contributions from road traffic was fitted to the measured PNC using the "lm" function of the package R (R, 2021). The results are shown in Fig. 5.

There clearly is a reasonably high correlation between the averaged PNC and the modelled NO_X concentrations at the measurement locations. From the fit to the data in our analysis, the relation between NO_x from traffic and PNC was found to be 662 ± 67 particles per cm^3 for each μg/m^3 of NO_X.

3.4. Combined analysis of the total PNC

In the previous sections we described how the background PNC and contributions from road traffic were estimated from the available measurements. As the background and contributions from road traffic were estimated relatively independent from each other, we can now fit the sum of the estimated background, the modelled contributions from road traffic and the modelled contributions from aviation to the total measured PNC using the averaged values per 10-degree wind direction interval for the measurement locations.

The measurements at location 6, close to the start of the frequently used runway "Polderbaan", were found to show very high peaks, substantially higher than the other stations. The location was only 400 m from and directly in line with the starting point of aircraft taking-off on the runway. Furthermore, it was close to a busy route for taxiing air planes. As a result, there was some doubt concerning the validity of the model to reproduce contributions from aviation at location 6. Given that the location was hardly/not representative for exposure of the population we decided not to include the measurements at location 6 in the

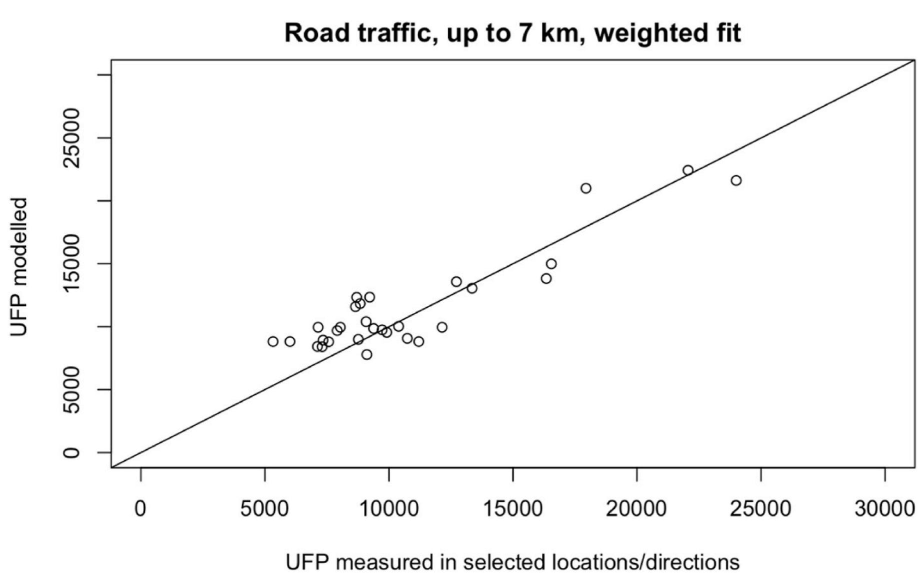

Fig. 5. Measured averaged PNC (particles/cm^3) on the x-axis and the fitted modelled concentrations from road traffic plus estimated background (particles/cm^3) at the y-axis at the measurement locations for the hours and wind directions with low aircraft contributions and high road contributions.

analysis.

For the contributions from aviation we use the PNC calculated using the Gaussian dispersion model STACKS+ described in section 2.3.1. For the combined analysis we want to determine the α, β and γ in the sum of the contributions, as shown below.

Measured PNC = α Estimated Background + β Modelled Traffic + γ Modelled Aviation (1)

The fit of the parameters to the total measured PNC was performed using the "lm" function of the package R. As the number of measurements coming from different wind directions varied substantially during the measurement periods, we used the number of hours of data as weight in the "lm" function. The result of the fit is shown in Fig. 6 and in Table 3. There is a satisfactory agreement between the measured and modelled PNC. The background PNC is scaled by 1.04, the traffic NO_X is converted to PNC with a factor of 675 particles/cm^3 per μg NO_x/m^3 and the modelled contribution by aviation activities is scaled by a factor of 0.78.

The results for the scaling of background and traffic are practically the same as for the data without contributions from aviation (for traffic 662 particles/cm^3 per mg NO_x/m^3). This indicates that the three contributions are quite independent from each other. The possible collinearity was checked using the "Performance" package in R. With a fitted offset in the analysis, the Variance Inflation Factors (James et al., 2017) are just over 1.0, whereas without an offset in the analysis the Variance Inflation Factors are between 1.35 and 2.05, all indicating low potential collinearity issues.

As mentioned, measurements were taken in two different periods, with two fixed locations and four rotating locations. The fits can be made separately for both periods, allowing us to test the robustness of the fitted values. The results of the fits for the α, β and γ are shown in Table 3. The uncertainties shown are standard deviations determined with a bootstrap routine (the R-routine simpleboot/lboot, https://github.com/rdpeng/simpleboot).

In Fig. 7 the modelled contributions from aviation at the measurement locations are compared to those from the background and road traffic. There is quite some variation in both the contributions from aviation and road traffic.

3.5. Comparison between aviation model and measurements

The performance of the dispersion model for aviation was evaluated using correlation analysis with the aim to assess the applicability of the model in epidemiological studies of long-term (monthly to yearly) exposure to UFP due to aviation. Therefore, estimated measured and calculated six-month average contributions from aircraft to PNC at the measurement locations were compared and correlation coefficients were derived.

We have taken the total measured PNC and subtracted the estimated background and modelled contributions of road traffic. The remaining difference in PNC is taken as an approximation to the "measured contributions from aviation". For all measuring locations the weighted average values over the wind direction intervals of the "measured" and modelled PNC are calculated. The results are shown in Fig. 8. Correlation coefficients are presented in Table 4. For the averaged data, the Pearson correlation coefficient is 0.88 and the Spearman correlation coefficient is 0.83, indicating good agreement between the estimated "measured" and modelled PNC. When the correlations are calculated for the almost 400 underlying data points, without averaging for every location, the Pearson correlation coefficient is 0.79 and the Spearman correlation coefficient is 0.80.

Fig. 8 illustrates that the ranking of the monitoring sites by modelling and monitoring agrees well. Modelling and monitoring suggest a range of average aviation related PNC from close to zero to roughly 10 000 particles/cm^3.

Table 3
Fitted values of the parameters in Equation (1) for all data, data from period 1 only and data from period 2 only.

Parameter	Total	Period 1	Period 2
Background (α)	1.04 ± 0.04	0.88 ± 0.08	1.06 ± 0.04
Traffic (β)	675 ± 75	1018 ± 268	620 ± 70
Aviation (γ)	0.78 ± 0.07	0.95 ± 0.18	0.69 ± 0.06

3.6. Yearly average PNC from aviation

As an illustration, a map of the yearly averaged contributions of aircraft emissions at Schiphol Airport to PNC was constructed using the scaled dispersion model for the year 2018 (Fig. 9). The spatial resolution of the model grid was 500 m for the area of 10 by 10 km surrounding Schiphol Airport and 1 km for the areas further away. The figures were made by interpolation of the values at the aforementioned grid, using an interpolation factor of 1.5 and smoothing. As input to the model, actual

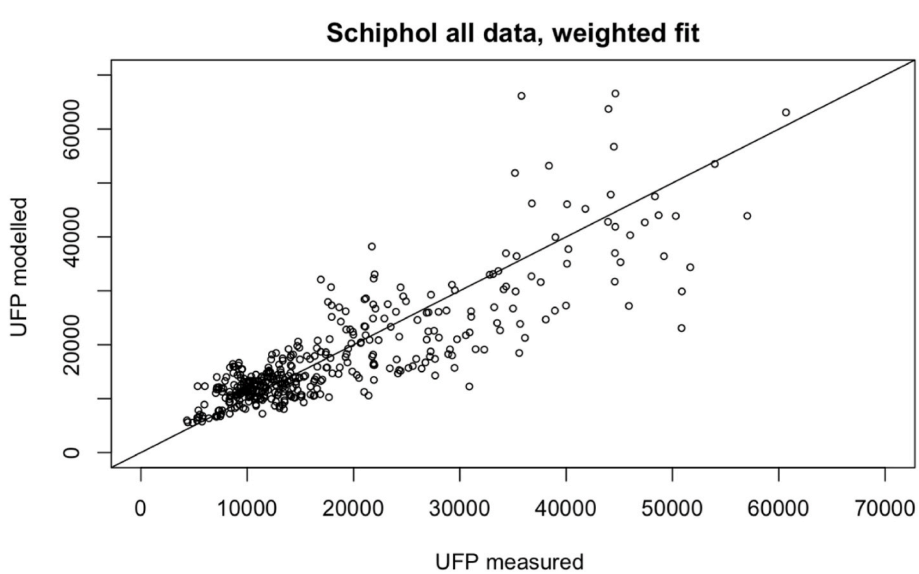

Fig. 6. Measured averaged PNC (particles/cm^3) on the x-axis versus fitted estimated background PNC plus modelled contributions from traffic and aviation (particles/cm^3) on the y-axis at the measurement locations per 10-degree wind direction interval.

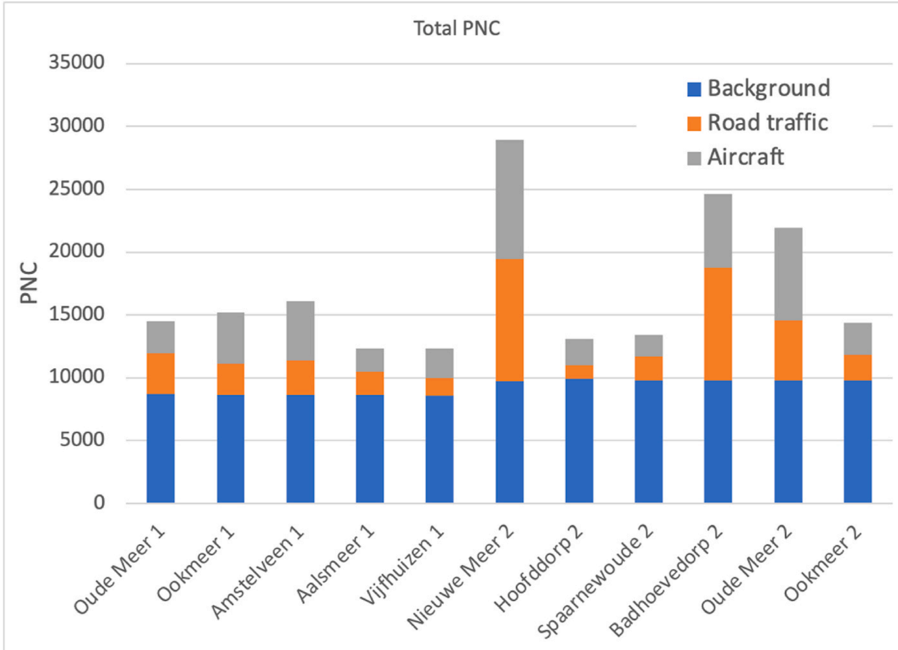

Fig. 7. The modelled six-month average contribution of aircraft emissions, road traffic and backgrounds to PNC in the measurement periods (in number per cm^3).

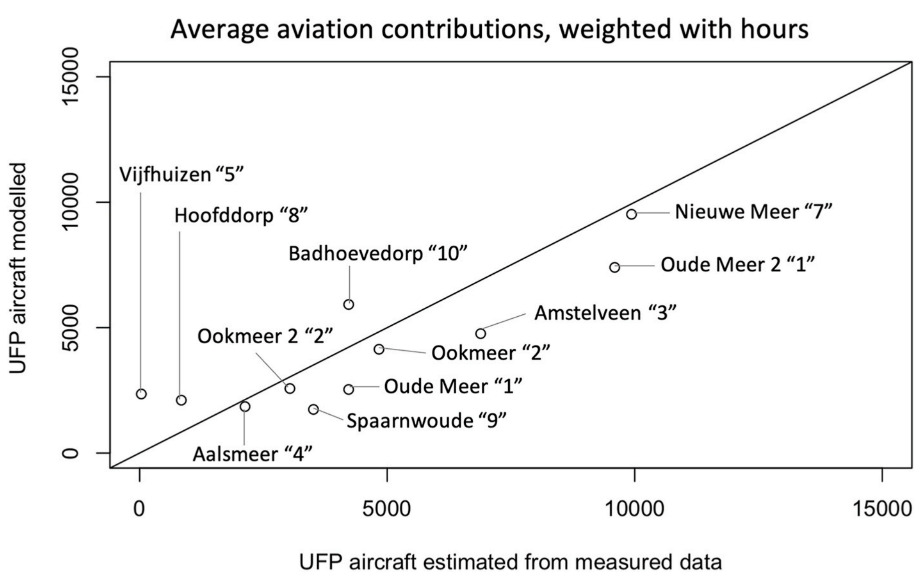

Fig. 8. Comparison of six-month averaged measured (x-axis) and modelled (y-axis) contribution from aircraft emissions to PNC in number per cm^3. Data points are labelled with location name and, where relevant, monitoring period. The numbers of the sites are shown in parentheses.

Table 4
Correlation coefficients between modelled and estimated measured contribution from aircraft emissions to PNC.

	Pearson	Spearman
Per 10-degree wind direction interval per location (n = 396)	0.79	0.80
Weighted average per location (n = 11)	0.88	0.83

meteorology, actual flight movements and the emission factors for UFP deduced from Mazaheri et al. (2009) were used. The model results are calibrated to the measurements by applying the scaling factor of 0.78. The map is illustrative for the maps that will be used in epidemiological studies among residents around Schiphol Airport.

The map shows that in general, the annual average exposure due to aircraft emissions rapidly decreases with the distance from Schiphol. For residential areas that are closest to Schiphol, the annual average contribution is around 10 000 particles per cm^{3,} with contributions up to 30 000 particles per cm^3 for individual residences closest to the airport. A contribution of 2 000 particles per cm^3 is possible up to a distance of 15 km from Schiphol Airport. The highest contributions in 2018 are modelled to the west of Schiphol Airport due to (exceptionally) frequent wind from easterly directions in 2018.

4. Discussion

4.1. Numerical results

Schiphol Airport is situated in an urban region in the Netherlands

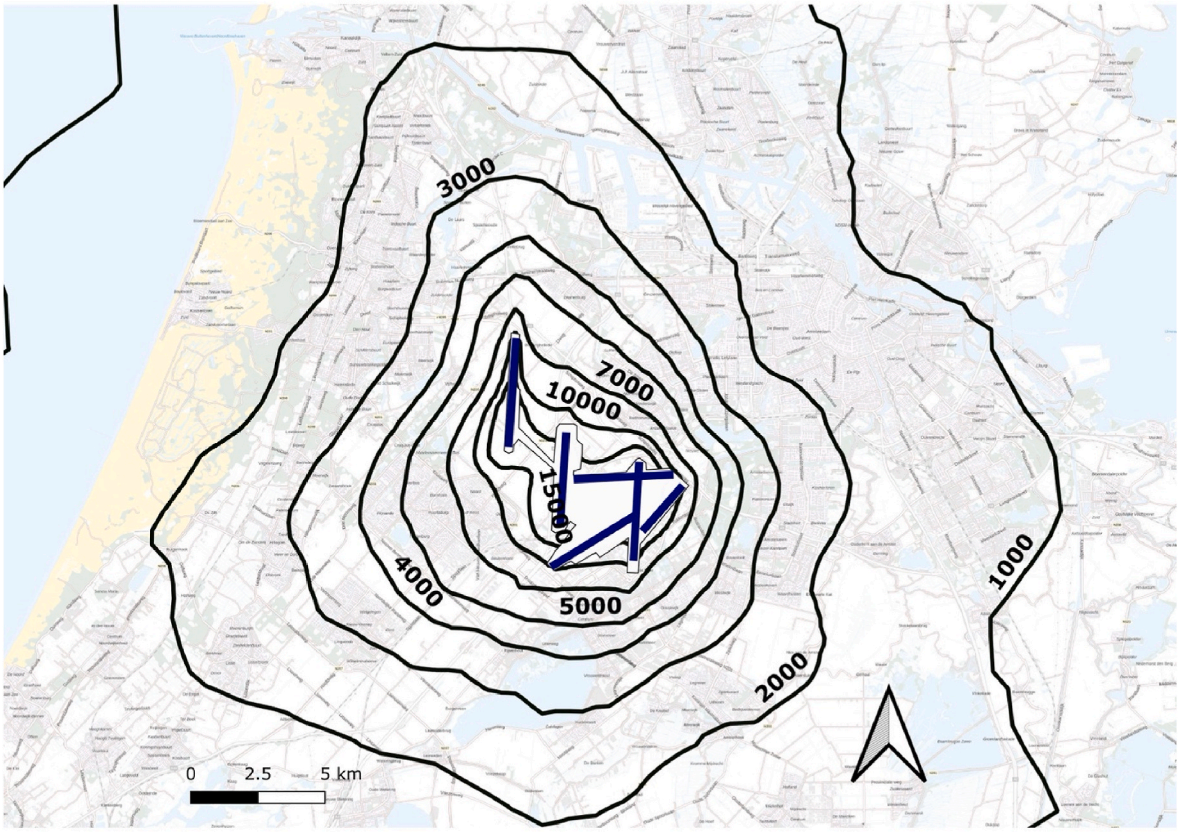

Fig. 9. The modelled annual average contribution of aircraft emissions at Schiphol Airport to PNC in 2018 (in number per cm^3). Note the larger, varying interval values between the contours higher than 5000.

that has many sources of UFP. It is therefore not straightforward to specifically measure the contribution by aircraft emissions. We applied a new method that combines modelled or estimated contributions from the main sources (aviation, road traffic on main roads and regional background) in a linear regression fit to measured PNC. The fit is based on the concentration estimates for all 10-degree wind direction intervals at each of the measurement locations. The resulting fitted value (scaling factor) of 1.04 ± 0.04 (1σ) for the background is close to 1. For aviation the scaling factor is 0.78 ± 0.07 (1σ). The deviation from 1 may at least partly be explained by bias in the applied emission factors that were deduced from measurements at Brisbane Airport more than 10 years ago (Mazaheri et al., 2009) and were applied to the present situation at Schiphol Airport. Taking the representativeness of these emission factors and possible biases in absolute measured PNC due to instrumentational differences into account, the results are quite satisfactory. Regarding road traffic, the scaling factor represents the conversion from NO_x to PNC, as the modelled contribution of road traffic is based on emissions of NO_x. We obtained a fitted value for the conversion factor of 675 ± 75 (1σ) particles/cm^3 per μg/m^3 in the combined analysis of road traffic, aircraft and backgrounds. When looking only at specific situations with high traffic and low aviation contributions, we found a conversion factor of 662 particles/cm^3 per μg/m^3. It is difficult to compare this number to values from literature due to differences in approach, types of roads/traffic and monitoring equipment. Converted to the same units, Keuken et al. (2016) found PNC to NO_x ratios in the order of 300–400 for *measured* difference between an *urban* traffic and background location in Amsterdam, The Netherlands. Gani et al. (2021) found ratios in the order of 300–600 for the *total measured* concentration at a highway location in California, USA, the highest ratio found during the summer season. Comparing with data from official emission databases like COPERT (https://www.emisia.com/utilities/copert-data/) or HBEFA (https://www.hbefa.net/e/index.html) is not useful as they provide only non-volatile particles whereas it is known that the vast majority of ultrafine particles from traffic emissions are volatile and form by nucleation or condensation (Keuken et al., 2016; Lefebvre et al., 2019).

Apart from the analysis for the combined measuring periods, we also performed the fit for the two six-month measuring periods separately, allowing us to test the robustness of the fitted values. For all contributors, the uncertainty is larger for period 1 than for period 2. The background has the smallest relative difference between the periods; traffic the largest. The latter is partly understandable because of the use of a single annual average contribution in the two periods. Furthermore, Gani et al. (2021) showed that measured PNC to NO_x ratios can vary substantially between seasons and moment of the day. If twice the standard deviation is interpreted as a 95% confidence interval, then it is clear that the different results between the periods are within each other's confidence interval for all contributors.

For the modelled contribution to PNC from aircraft emissions related to Schiphol Airport, the 95% confidence interval uncertainty based on the analytical fit is around 20%. However, when taking into account uncertainties in choices made in the numerical analysis, the uncertainty is assumed to be larger.

4.2. *Strengths and limitations*

A strength of our approach for the performance assessment of the model for aviation-related UFP is that it allows us to deduce the contribution of aviation to the total measured PNC by performing a combined analysis with other sources (road traffic on main roads and regional background). Such an approach adds to the scientific knowledge, especially in the context of aviation-related UFP. In a previous exploratory study around Schiphol Airport the contribution of aviation was deduced by expert judgement (Bezemer et al., 2015). The present study yields approximately 30% smaller contributions from aviation compared to the expert judgement method. The present analysis not only allows for a better estimate of the contributions of sources, it also allows for a better uncertainty assessment.

The assumption of using NO_x as a proxy for UFP from road traffic introduces uncertainties. Gani et al. (2021) reported that spatiotemporal variation in NO_x concentrations may differ from that of PNC. In a study by Kwasny et al. (2010) it was concluded that NO_x proxy indicators may be used in temperate regions, but are unsuitable parameters for warmer or even tropic regions, which the Netherlands clearly is not. As we furthermore use PNC that are averaged for every wind direction, we expect the NO_x to be an acceptable proxy for the UFP contributions from traffic, especially at the higher contributions.

A limitation of our approach is that we were forced to combine data available for slightly different periods in time. For the background, the measurements of period 1 and 2 are combined to an aggregated background pollution rose for the total period. Since the dispersion model for road traffic calculates the concentrations on a yearly basis, the 2018 pollution rose values are used as estimates for both six-month measurement periods (partly 2017, partly 2018). The occurrence of different wind directions in both measuring periods was taken into account by working with averaged values for each 10-degree wind direction interval. However, the wind speed from each direction was the average of the speeds from that direction in both periods. In addition to other differences between the two periods, this creates some additional uncertainty. However, the differences in results when using only data from period 1, period 2 or combining all data in our analysis are all smaller than the 95% confidence interval for the parameters.

Evaluation of the performance of a model for assessing spatial variation in long-term averaged concentrations is challenging as it requires long-term monitoring at a large number of locations differing in concentrations. For this study, six high-quality condensation particle counters of the same manufacturer and model-type were available for more than a year. In the Netherlands (and to our knowledge also abroad), there have been no other studies that used so many identical high-quality monitors for such a long period. However, aiming at the assessment of the applicability of the model for long-term (monthly to yearly) exposure, the number is still limited. Our measurement strategy was balanced between the number of locations and the duration of the measurements, allowing for an optimal robustness of the measured and modelled concentrations being representative for long-term exposure. However, data from one location (location 6 "Polderbaan", situated quite close to aircraft taxiing and taking off) needed to be excluded after a more detailed analysis leading to some doubt about the suitability of the model to reproduce contributions from aviation at location 6. The lack of a description in the model for horizontal displacement of the exhaust stream due to the high exhaust velocity may play a role in this case.

The dispersion model for aviation-related UFP itself is based on some simplifying assumptions and has limitations, described in Section 2.3.1. We are well aware of the complexity of the dispersion of jet plumes and the limitations of the model calculations. Calibrating the model results with actual measurements will (partially) help to overcome the shortcomings of the dispersion model.

A further limitation is the fact that the model performance was tested and the scaling factor was determined based on measurements in 2017 and 2018, whereas the model will be used for epidemiological studies starting approximately 10 years earlier. This introduces uncertainty in the representativeness of the adjusted emission factors for the fleet of aircraft in the years in the past. Our study is the first in which the performance of the dispersion model is explicitly tested for aircraft emissions. PNC measurements have only recently been performed in the surroundings of Schiphol Airport. The effect of aircraft emissions on most other aspects of air quality is limited. PNC measurements are much more suitable for the assessment of the model performance for aircraft emissions than measurements of other pollutants since the signal-noise ratio is much higher. However, for a number of reasons we think that the present results can be useful also for calculations in the recent past. Most importantly, the spatial variation of the exposure is expected not to be impacted much by changes in emission factors since the layout of the runways at Schiphol Airport has not changed since the end of 2003 and the operational procedures (landing and takeoff trajectories and routing of airplanes through the sky) have also not changed significantly since then (Ministerie van Infrastructuur en Milieu, 2012; MovingDot, 2018). While a change in emission factors over time due to changes in the fleet composition has an impact on the absolute PNC levels, it does not significantly impact the spatial contrast in PNC. For epidemiological studies, correctly classifying the absolute concentration levels is important when statements about the concentration response function are needed, e.g. at what concentration level a health effect of a certain magnitude occurs. However, at the current state of knowledge, the key question is *whether* long-term aviation related UFP is related to health effects and much less *at what exact* concentration levels.

4.3. Comparison with other airports

We compared the resulting contributions from aviation to PNC in the surroundings of Schiphol Airport as calculated for 2018 with yearly averaged PNC values obtained in other modelling studies. We estimated concentration levels calculated at 1 and 15 km from the nearest runway in these other studies. For this, we looked up the values using figures showing contours of aircraft contribution to PNC presented in these studies. Results are presented in Table 5.

At 1 km, the contribution by aviation to yearly averaged PNC ranges from 5 to 25*10^3 particles per cm^3. At 15 km, the range is almost zero to 5*10^3 particles per cm^3. Our study's values fall between the lower and higher end values.

A quantitative comparison of the PNC levels resulting from aircraft emissions with modelled values from other studies is not straightforward. First, the way the values from the other studies were estimated introduces some uncertainty: when looking them up in contour plots, we were dependent on the interval widths selected by the authors and information about the spatial scale. Furthermore, the studies differ in their modelling approach using different emission datasets and different

Table 5
Contributions from aviation to yearly averaged PNC values in recent modelling studies.

Study	Airport	Year of data	Annual mean PNC at 1 km distance (10^3 per cm^3)	Highest annual mean PNC at 15 km distance (10^3 per cm^3)	D_{50} value of measurement equipment for model calibration (nm)
Present study	Schiphol Amsterdam	2018	10	2	7
Bezemer et al. (2015)	Schiphol Airport	2015	15	3	7
Keuken et al. (2015)	Schiphol Amsterdam	2012	20	3	4
Lefebvre et al., 2019	Zaventem Brussels	2015	8–13	1	10
Lorentz et al. (2019)	Frankfurt	2015	5–12	0.5	No calibration
Zangh et al. (2020)	Zurich	2017	10–25	0.2	No calibration
Wing et al. (2020)	LAX Los Angeles	2008–2016	12–15	5	4

dispersion models (see Section 1). For example, Lorentz et al. (2019) modelled the non-volatile particles only for their Frankfurt study. Also, equipment and duration of the measurements that were used for model calibration differ. Stacey (2019) and Riley et al. (2021) showed that differences in (handling of) equipment can have large impact on the absolute measurement values. For example, the lowest particle sizes (D_{50}) that can be detected by the equipment used in the studies range from 4 to 10 nm. Finally, the geographical and meteorological situations can be very specific for some airports. Zangh et al. (2020) performed their study for Zurich Airport that is situated in an inner Alpine basin. This may partly explain higher PNC in this area. The region of LAX Airport in Los Angeles as studied by Wing et al. (2020) is known for steady wind direction (onshore breeze). This may explain the relatively high value at 15 km from the airport. Giving all these variations, we conclude that our modelled contribution from aviation to yearly PNC is quite comparable to values found in other recent studies.

4.4. Suitability of the model for epidemiological studies

The model performance for the dispersion of aviation-related UFP was assessed for the application of long-term (monthly to yearly) PNC exposure estimations in support of an epidemiological study. The comparison of the six-month averaged modelled contributions by aviation to PNC using emission factors for the different flight phases deduced from Mazaheri et al. (2009) with measurements showed a good agreement. The ranking of the modelled and measured concentrations at monitoring sites agreed well. The correlation coefficients between the six-month averaged modelled and measured contribution from aircraft emissions at locations representative for population exposure were high (>0.83). Using all 396 individual data points resulted in marginally lower correlations (>0.79). The correlation found in our study is as high or higher than reported in recent epidemiological studies for other major air pollutants (Carey et al., 2013; Hvidtfeldt et al., 2018).

There is no well-defined threshold of a correlation below which a model is no longer useful for epidemiological studies. With lower model performance, more misclassification of exposure occurs, generally resulting in bias of estimated health effects (Armstrong, 1998). Furthermore, the correlation coefficient between modelled and measured concentrations can be used to express how much bias may occur in an epidemiological study using an imperfect exposure assessment. The resulting bias in an epidemiological study also depends on the exposure contrast in the study: with larger contrast, the same measurement error results in less bias (Armstrong, 1998). At the monitoring locations representative for population exposure in our study, a range of average aviation related PNC from about from almost zero to roughly 10 000 particles/cm^3 was observed. This demonstrates that large contrasts in concentration exist in the study area.

5. Conclusion

We developed a method in which the contribution of traffic on main roads and the background concentration are explicitly taken into account in the analysis of UFP contributions from aviation. We conclude that this method, although it has its limitations, enabled us to make a robust estimate of the contribution of aviation to the measured PNC. The applied approach may be valuable for studies evaluating the performance of other models for assessing PNC due to aircraft emissions in areas with several other large emission sources.

Based on 1) the high correlation coefficients between the modelled contribution of aircraft emissions to PNC and the contribution estimated from the measurements and 2) the large contrast in aviation related PNC within the study area, we conclude that the dispersion model is able to determine the spatially varying average concentrations due to aircraft emissions in residential areas over periods of (at least) 6 months. This give us confidence that the model is suitable for application in epidemiological studies into long-term exposure.

CRediT authorship contribution statement

Marita Voogt: Conceptualization, Methodology, Formal analysis, Data curation, Writing – original draft, Project administration. **Peter Zandveld:** Validation, Formal analysis, Data curation. **Hans Erbrink:** Methodology, Software, Writing – review & editing. **Danielle van Dinther:** Investigation, Data curation, Writing – review & editing. **Pim van den Bulk:** Investigation. **Gerard Kos:** Investigation. **Marcus Blom:** Investigation. **Dave de Jonge:** Investigation. **Harald Helmink:** Investigation. **Jennes Meydam:** Investigation. **Jaap Visser:** Writing – review & editing. **Jan Middel:** Resources, Writing – review & editing. **Gerard Hoek:** Writing – review & editing. **Sjoerd van Ratingen:** Formal analysis, Validation, Writing – review & editing. **Joost Wesseling:** Methodology, Formal analysis, Conceptualization, Writing – original draft. **Nicole AH. Janssen:** Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data are available from the project website on rivm.nl (links provided in the manuscript)

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2023.119884.

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