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Variation in Polycyclic Aromatic Compound (PAC) Concentrations in a Norway Spruce Stand Close to a Major Traffic Route—Influence of Distance and Season

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Abstract Air concentrations of polycyclic aromatic compounds (PACs) were measured in a Norway spruce (*Picea abies*) stand at four different distances from a major traffic route near Gothenburg, Sweden, during summer and winter 2019, using passive samplers. In total, the concentrations of 32 polycyclic aromatic hydrocarbons (PAHs) and six dibenzothiophenes (DBTs) and NO_2 were analysed. PAC air concentrations were on average 2.5 times higher during the winter compared to the summer. All investigated PAH categories and compounds as well as dibenzothiophene declined linearly with the logarithmic distance from the traffic route. This indicates that the length of pollutant pathways through vegetation is a valid proxy to assess health risks of traffic-related PAC pollutants. Concentrations of heavier PAHs declined faster (concentration at site furthest away from the road was 6% and 33% of concentration at site closest to road, for summer and winter respectively) with increasing distance from traffic compared to lighter PAHs (63% and 68% for summer and winter respectively). As a result, the mix of PAHs changed with the depth into the forest stand (i.e. distance from the road) with potential effects on the toxicity as well as on diagnostic ratios used for source attribution. This is likely caused by different deposition rates among the PAH compounds to the vegetation surfaces, although the exact mechanism needs to be further investigated. This study provides new information that can improve

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air quality risk assessment and limit the exposure of the population to toxic air pollutants such as PACs.

1 Introduction

Air pollution is associated with adverse health effects such as cardiovascular diseases (Bourdrel et al., 2017), lung cancer (Hamra et al., 2015), childhood asthma (Khreis et al., 2017), childhood respiratory diseases (Loukili et al., 2022) and increased risk for dementia (Peters et al., 2019). Road traffic is a major source of air pollutants, and accurate assessment of the exposure to traffic-related air pollutants is crucial when investigating the health effects of traffic emissions (Gilbert et al., 2003). In risk assessments of traffic-related air pollution, the distance to major roads is sometimes used to estimate pollution levels (e.g. Hamra et al., 2015; Hoek et al., 2002), which highlights the importance of understanding the variation in air pollution concentration near roads. In a study from Canada, Gilbert et al. (2003) found a significant decrease in NO_2 concentration with increasing logarithmic distance from a highway, indicating that distance can be used to estimate the decline of traffic-related air pollutant concentrations from roads. Corresponding results for NO_2 have been obtained in studies from Sweden (Klingberg et al., 2017; Pleijel et al., 2004).

There is an increasing concern about polycyclic aromatic hydrocarbons (PAHs) as they are ubiquitous in ambient air, persistent in the environment and because of their carcinogenicity and mutagenicity (Boström et al., 2002; Famiyeh et al., 2021; IARC, 2013; Kim et al., 2013). PAHs are formed through the incomplete combustion of organic material arising, partly, from natural sources such as wildfires, but with the majority due to anthropogenic emissions (Ravindra et al., 2008). In many urban areas, vehicle emission is the dominating source (Ravindra et al., 2008). PAHs belong to the wider group of polycyclic aromatic compounds (PACs) (Achten & Andersson, 2015).

PAHs constitute a diverse group of compounds, with different chemical-physical properties. Some are comparatively low molecular mass, (L-PAHs, consisting of 2–3 benzene rings), and mostly occur in the gaseous phase at temperatures typical of the lower atmosphere, while others, high molecular mass (H-PAHs, 4–7 benzene rings), are to a large extent bound to particles. The intermediate category of M-PAHs (3–4 benzene rings) can occur in both phases. The most well-studied PAH is the particle-bound (H-PAH) benzo(a)pyrene (BaP) which is used as an indicator of carcinogenic PAH exposure in the air (Boström et al., 2002; EEA, 2019).

Another category of PACs are the dibenzothiophenes (DBTs). DBTs are organosulphur compounds containing two benzene rings fused to a central thiophene ring, chemically related to the L-PAH anthracene. DBTs have not been monitored to the same extent in air as PAHs (Lammel, 2015). In a study from Toronto, Canada, Jariyasopit et al. (2019) investigated ambient levels of PACs associated with different source types and highlighted the importance of traffic as an emission source of PACs to urban air and the relevance of PAC categories other than just PAHs, such as DBTs.

NO_2 is a traffic-related air pollutant with strongly elevated levels in areas with abundant traffic (Pleijel et al., 2004). Air concentrations of NO_2 are routinely monitored and often used to characterise the air pollution levels and as a proxy for other air pollutants. Klingberg et al. (2022) found a strong correlation between air concentrations of NO_2 and PAHs comparing differently polluted sites in the City of Gothenburg, emphasising that these pollutants to a large degree originated from the same sources, essentially combustion engines of road traffic.

Vegetation, with its large leaf surface area to which pollutants can deposit, has the potential to influence the local concentration of air pollution (Abhijith et al., 2017). Vegetation accumulates atmospheric lipophilic organic pollutants such as PAHs in leaves (Desalme et al., 2013; Simonich & Hites, 1994). Trees and other types of vegetation also influence the air mixing and may cause a reduction in ventilation and reduced dilution of the emitted air pollutants. In certain situations, this aerodynamic effect is stronger than the pollutant removal capacity of vegetation (Vos et al., 2013). The net effects of these processes on local scale atmospheric PAH concentrations can be both positive and negative with respect to air quality (Klingberg et al., 2017; Viippola et al., 2016) depending on the situation, season and compound studied. Increased knowledge of the influence of vegetation on air quality along busy roads can contribute

to sustainable urban planning strategies, efficient mitigation strategies and estimations of human exposure risks to carcinogenic pollutants.

PAHs are emitted as a mixture, and the relative concentration ratios are often assumed to be characteristic of a given emission source (Tobiszewski & Namiesnik, 2012). These so-called PAH diagnostic ratios have been used as a tool for the identification of emission sources, necessary for proper risk assessment and risk management, e.g. in Italy (Cecinato et al., 2014), China (Wang et al., 2020) and Turkey (Sari & Esen, 2022). However, the accuracy and reliability of PAH diagnostic ratios have been questioned (Famiyeh et al., 2021; Katsoyiannis & Breivik, 2014; Katsoyiannis et al., 2011; Tobiszewski & Namiesnik, 2012). Both vegetation and organic surface films in the environment can accumulate particles and gaseous phase chemicals from air (Csiszar et al., 2012; Simonich & Hites, 1994). If the capture efficiency of individual gaseous phase constituents or particle sizes in these organic matrices varies, it could affect a diagnostic ratio and thus the validity of such a ratio to characterise sources.

The variation in air concentrations of PACs is not as well studied as that of NO_2, and an improved understanding is needed to accurately assess the exposure to these traffic-related air pollutants, when investigating the health effects of traffic emissions. The decline in concentration of these compounds with distance from roads and the influence of vegetation is crucial for accurate risk assessments regarding human exposure as well as for accurate source attribution.

We investigated the atmospheric concentrations of PAHs (32 compounds), DBTs (six compounds) and NO_2 using passive air samplers inside a Norway spruce (*Picea abies*) stand at four distances from a major traffic route north of Gothenburg, Sweden. The aim of the study was to characterise the spatial and temporal variation of atmospheric PAC concentration with respect to the influence of distance from a road and the season of the year. Firstly, we aimed to investigate how the air concentrations of different PAC categories, such as particle-bound PAHs, gaseous PAHs and DBTs, change with increasing distance from a road inside a planted Norway spruce stand. A second aim was to study how the PAC composition changes with distance from the road and between summer and winter. Finally, we aimed to analyse how

PAH diagnostic ratios varied with increasing distance from the road, as well as between summer and winter, and to highlight the implications of the sensitivity of diagnostic ratios to distance and season for PAC source attribution.

2 Methods

2.1 Measurement Site

The study was conducted in a planted Norway spruce (*Picea abies*) stand, approximately 20 m tall, next to the major traffic route E6 north of the city of Gothenburg (57.828874° N, 12.003443° E). Approximately 50,000 vehicles pass each day (Trafikverket, 2020). Equipment for passive sampling of PACs and NO_2 was installed in the spruce stand at 1, 10, 40 and 80 m from the forest edge, which was located 35 m from the traffic route. Further description of the site can be found in Figure S1 in the supplementary information.

Gothenburg is located on the coast of south-west Sweden. It is the second largest city in Sweden with approximately 600,000 inhabitants (SCB, 2018). It has a maritime temperate climate with, for the latitude, moderately cool summers and mild winters.

2.2 PAC and NO_2 Air Measurements

The air concentrations of the two PAC categories, PAHs and DBTs, were measured using passive PUF (polyurethane foam) disc samplers (14 cm in diameter, 1.2 cm thickness, surface area 360 cm^2, density 0.035 g cm^{-3}, Klaus Ziemer GmbH, Germany) housed in two stainless steel domes (Tisch Environmental, Inc., OH, USA). This sampler design has been calibrated for both gaseous and particulate associated PAC compounds including alkylated species (Bohlin et al., 2014; Harner et al., 2013). Prior to deployment, the PUF discs were precleaned by Soxhlet extraction for 24 h using dichloromethane (DCM), dried under vacuum and stored in multiple layers of solvent rinsed aluminium foil inside airtight polyethylene zip bags. After air sampling, the PUF discs were enveloped in rinsed aluminium foil, placed in airtight polyethylene zip bags and brought back to the laboratory and stored at −18 °C until extraction.

NO_2 was measured using passive diffusion samplers of the IVL type (Ferm, 2001; Sjöberg et al.,

2001). The samplers have been used in past measurement campaigns in Gothenburg and shown reliable results (Klingberg et al., 2017). The sample analysis was made by the IVL accredited laboratory (www.ivl.se).

The PAC and NO_2 samplers were deployed at ca 1.8 m height and exposed for 26–28 days (Table 1). Sample design, wind speed and direction during the measurement periods are presented in Figure S1 in the supplementary information. Duplicate sampling of PAC and NO_2 was performed at two locations (1 m and 80 m from the forest edge), and three PUF field blanks were taken during each sampling period. The field blanks, i.e. samplers unopened during the measurement campaign, were processed in parallel with the samples in the analytical procedure to estimate the residual levels of target compounds.

2.3 Analysis of PAHs and DBTs

In total, the concentrations of 32 PAHs and 6 DBTs were determined, among them 15 US EPA priority PAHs and 14 alkylated species. The following five PAH categories are highlighted in this paper: SUM-PAH refers to the sum of the 32 PAH compounds included, L-PAHs refers to low molecular mass PAHs (>90% in gaseous phase), M-PAHs refers to intermediate (medium) PAHs (fractions in both gaseous and particulate phases), H-PAHs refers to particle bound, high molecular mass PAHs (>90% on particles) and DBTs (sulphur-containing PAH derivates). In addition, the individual compounds phenanthrene (an abundant gaseous PAH), the particle-bound benzo(a)pyrene and dibenzothiophene are discussed separately. Dibenzothiophene does not contain any methyl groups, in contrast to the other DBTs included. Benzo(a)pyrene was selected since there is

a health-based guideline value of 0.12 ng m^{-3}, corresponding to an excess lifetime lung cancer risk of 1/100,000 (WHO, 2000). See Table S1 in the supplementary information for more details on which PACs are included in the different categories, their molecular mass and number of benzene rings. All PAC and NO_2 data are available in the supplementary information.

The retrieved field-exposed PUF discs were extracted and cleaned up following the procedure used earlier for the PUF (Bohlin et al., 2008). In brief, prior to extraction, the samples were spiked with internal standard (IS) containing deuterated US EPA 16 PAHs. Recovery standard (RS) containing octachloronaphthalene was spiked in each sample during the last step before analysis. PAHs were then separated and detected by means of high-resolution gas chromatography/low-resolution mass spectrometry (HRGC/LRMS). The MS instrument was an Agilent 5975C connected to a 7890A GC (Agilent Technologies, Inc., Santa Clara, CA, USA). The MS was operated in electron impact (EI) ionisation using the selected ion monitoring (SIM) mode, and the GC column was a non-polar capillary column (60 m × 0.32 mm id and 0.25-μm film thickness; J&WDB-5, Folsom, USA). The published uptake rates by Bohlin et al. (2014) were used to quantify air concentrations of 15 US EPA PAHs.

Two-ring PAHs, such as naphthalene, may, after a 28-day sampling period, be in the curvilinear phase of uptake or have reached saturation in the sampling material (Bohlin et al., 2014). Thus, accurate quantification of this compound could not be made, which is therefore excluded from analysis and further discussions. The alkylated PAHs were quantified using published uptake rates by Harner et al. (2013) or using the uptake rates for a corresponding US EPA PAH compound.

Table 1 Overview of start, stop and duration of the measurement periods. Background NO_2 concentrations (μg m^{-3}), temperature (°C) and precipitation (mm) data were retrieved from the City of Gothenburg, Environment Administration rooftop measurement station Femman, situated in the centre of the town, approximately 15 km south of the PAC measurement site (https://goteborg.se/wps/portal/start/kommun-o-politik/kommunfakta/oppna-data/oppna-data-soksida/oppna-data-datamangd#esc_entry=690&esc_context=6; accessed 11 August 2021)

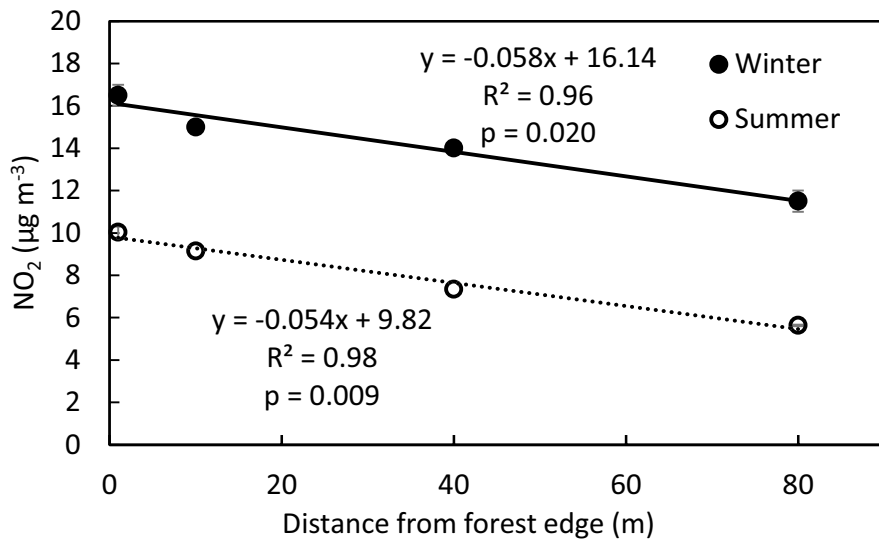| Time period	Start (yyyy–mm–dd)	Stop (yyyy–mm–dd)	Duration (days)	NO_2 (μg m^{-3})	Temperature (°C)	Precipitation (mm)
1 (summer)	2019–07–25	2019–08–20	26	11.2	18.9	108
2 (winter)	2019–11–11	2019–12–09	28	17.2	5.6	55

Fig. 1 NO_2 air concentrations in relation to distance from the edge of the spruce stand during the approximately 4-week long summer and winter measurement periods. Error bars show standard deviation of two parallel (duplicate) measurements

2.4 Quality Assurance/Quality Control

There were good agreements between the duplicates; the deviations from their means were about 5–25% for all PAC target compounds, verifying the applicability of PUF disc to monitor both gaseous and particle-associated compounds. Some PAC residues were found in the blanks, but in no case was the amount of any compound in any blank >5% of the amount found in any sample. All PAC results were corrected to the blanks. The limits of detection (LODs) were calculated as three times the SD of the blanks. A certified reference material (SRM 1649a urban dust) was used as a quality control (QC) sample. The measured levels of 12 PAHs rarely deviated >10% from the certified levels. Recoveries were routinely monitored, and ranged from 50 to 120% for all internal standards.

2.5 Analysis of Pollution Concentration Decline, Statistics and PAH Diagnostic Ratios

The decline in air pollution concentration from road sources has previously been described as a linear function versus the logarithmic distance from the road source, e.g. in studies conducted for NO_2 in Canada (Gilbert et al., 2003), in the USA (Rodes & Holland, 1981) and in Sweden (Pleijel et al., 2004)

as well as for PAH in an earlier study from Gothenburg, Sweden (Klingberg et al., 2017). An exponential decline of the concentration with increasing distance is theoretically more appealing since it moves towards background levels at large distances and will never obtain negative values. However, in this study, we use the function resulting in the best fit to the measured data as the aim was to describe the concentration decline within the range of distances from forest edge, and not predict the concentrations outside this range. For the decline of NO_2 concentrations with distance, a linear regression provided the best fit (see Fig. 1 and Figure S3a and S3b in the supplementary information showing an exponential and a logarithmic function, respectively). For PACs, the best fit was obtained using a linear regression of PAC concentrations versus the logarithmic distance from the forest edge.

The significance of the relationship between pollution concentration and distance was tested using regression analysis. Summer and winter concentrations were compared using paired t test. A probability value (p) below 0.05 was used to indicate statistical significance. Microsoft Excel (version 2202) was used for the statistical analysis. PAC values below LOD were set to zero.

Five PAH diagnostic ratios from several literature sources, compiled by Tobiszewski & Namiesnik (2012), were chosen to investigate
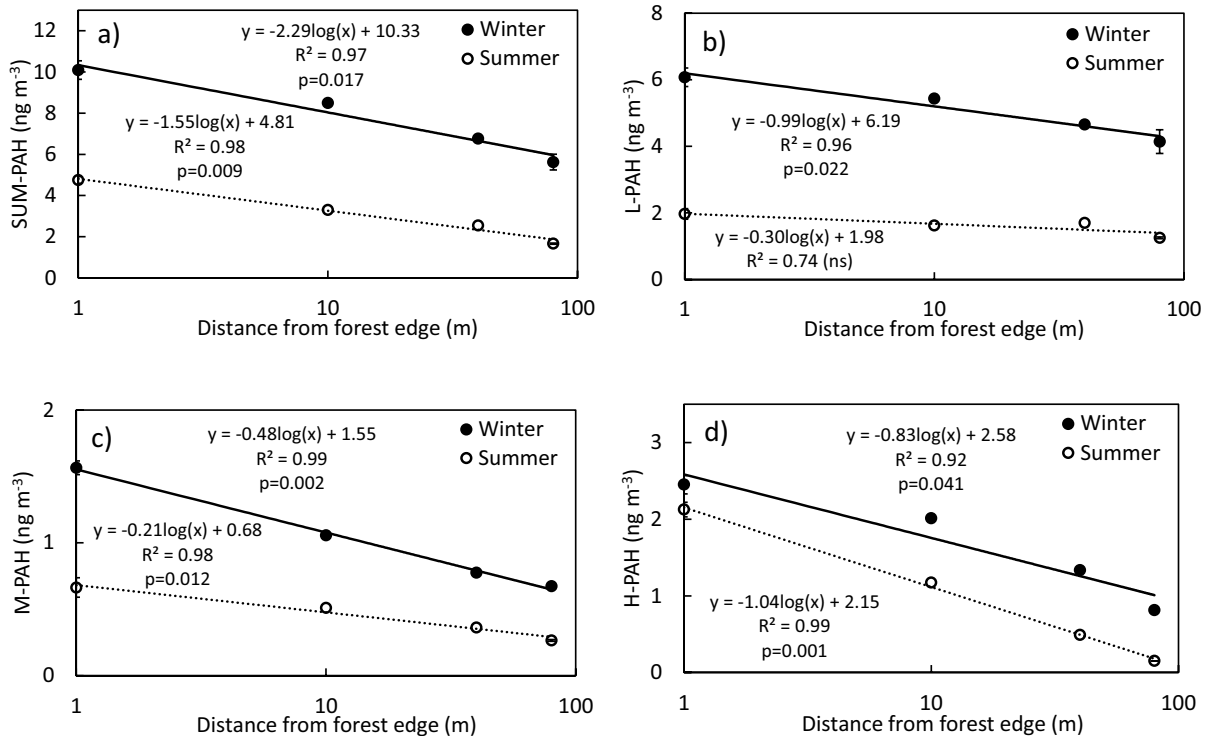
Fig. 2 SUM-PAH (**a**), L-PAH (**b**), M-PAH (**c**) and H-PAH (**d**) air concentrations in relation to distance from the edge of the spruce stand during the approximately 4-week long summer and winter measurement periods. Note the logarithmic scale on the x-axis. Error bars show standard deviation of two parallel measurements

possible PAHs source attribution and for tracking differences between PAH patterns, between summer and winter measurements as well as at different distances from the road. The diagnostic ratios and their reported typical values for different emission sources are shown in Table S2 in the supplementary information.

3 Results

3.1 PAC and NO_2 Concentrations

The NO_2 concentrations were significantly higher during the winter measurement period compared to the summer according to the paired t test ($p<0.001$).
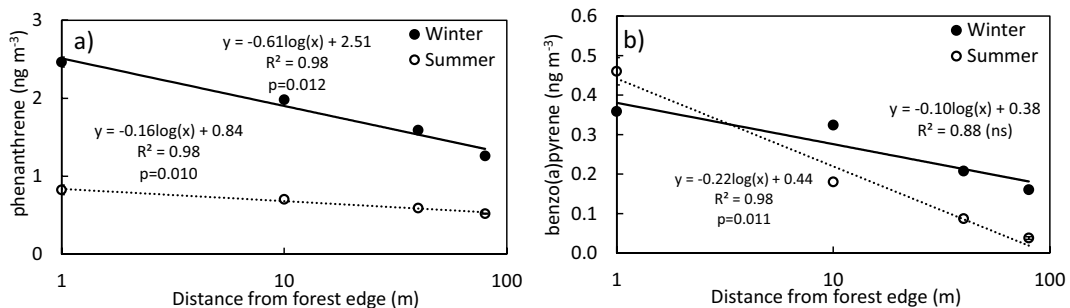

Fig. 3 Phenanthrene (**a**) and benzo(a)pyrene (**b**) air concentrations in relation to distance from the edge of the spruce stand during the approximately 4-week long summer and winter meas-urement periods. Note the logarithmic scale on the x-axis. Error bars show standard deviation of two parallel measurements

NO$_2$ concentrations were on average 1.8 times higher during the winter compared to the summer. During both seasons, the NO$_2$ concentration decreased linearly with increasing distance from the road (Fig. 1).

In Fig. 2, the PAH concentration of the categories SUM-PAH (sum of all 32 investigated PAH compounds), L-PAH (mainly gaseous), M-PAH (both gaseous and particle-bound) and H-PAH (mainly particle-bound) are shown in relation to the distance from the road. As for NO$_2$, the PAH concentrations were significantly higher during winter compared to the summer measurement period (p=0.008). SUM-PAH concentrations were on average 2.5 times higher during the winter period compared to the summer. During both seasons, the PAH concentrations decreased with increasing logarithmic distance from the road. The regression was statistically significant ($p \leq 0.041$) for all PAH categories except L-PAH during summer. Noteworthy is the difference in rate of decrease between the different PAH categories. Based on the regression function, we calculated at what distance the concentration had decreased with 25% compared to the concentration at 1 m from the forest edge. The L-PAHs reached a 25% reduction in 48 m (summer) and 45 m (winter). The H-PAHs decreased much faster with distance and was estimated to reach a 25% reduction at 3.4 m (summer) and 7.8 m (winter).

In Fig. 3, the concentration of the most abundant PAH, phenanthrene (an L-PAH), which consisted 23% of total PAH and 38% of L-PAHs on average, and the highly toxic benzo(a)pyrene (an H-PAH)
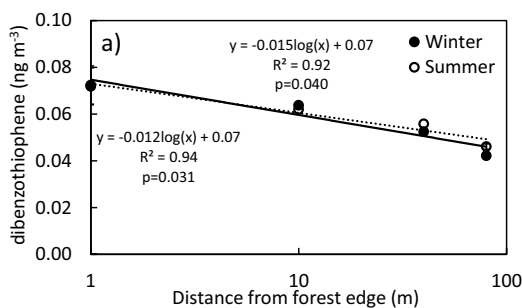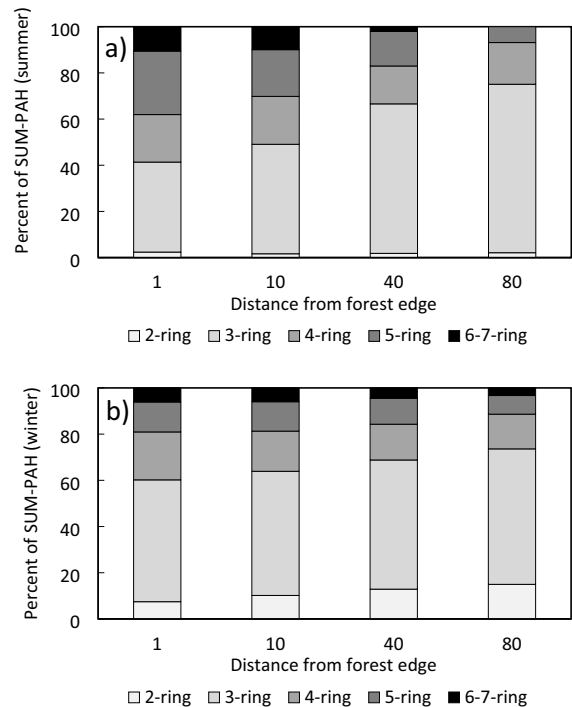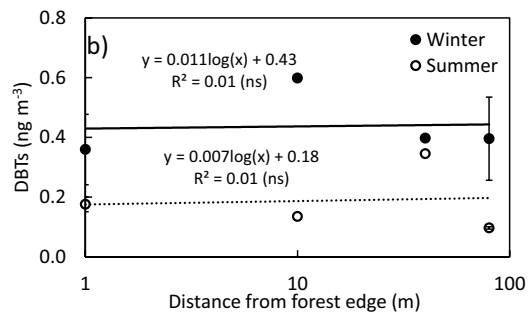
Fig. 5 Percentage of total SUM-PAH that consists of PAHs with different numbers of benzene rings during the approximately 4-week long summer (**a**) and winter (**b**) measurement periods at different distances from the edge of the spruce stand

are presented. Both are on the list of the 16 priority PAHs by the United States Environmental Protection Agency (US EPA) (Ravindra et al., 2008). Phenanthrene was significantly higher during winter (p=0.005). This was not the case for benzo(a)pyrene, which at the location closest to the road had a

Fig. 4 Air concentrations of dibenzothiophene (**a**) and DBTs (**b**) in relation to distance from the edge of the spruce stand during the approximately 4-week long summer and winter measurement periods. Note the logarithmic scale on the *x*-axis. Error bars show standard deviation of two parallel measurements
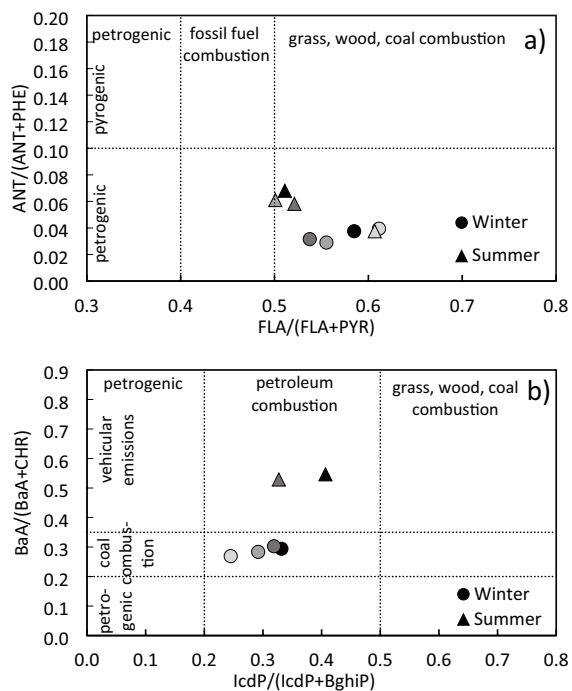
Fig. 6 Diagnostic ratios calculated for air samples at four distances from a road during the approximately 4-week long summer (triangles) and winter (circles) measurement periods. Darker colour indicates measurement location closer to road. ANT, Anthracene; PHE, phenanthrene; FLA, fluoranthene; PYR, pyrene; BaA, benzo(a)anthracene; CHR, chrysene; IcdP, indeno(1,2,3-c,d)pyrene; BghiP, benzo(g,h,i)perylene

in concentration between summer and winter for this compound. The other five compounds included in the DBTs category varied greatly. No conclusions regarding the influence of distance from the road or season could be drawn even though the category as a whole tended to have higher concentrations during winter than summer.

3.2 PAH Composition

The relative abundances of PAHs, with different numbers of benzene rings, in the air are presented in Fig. 5. Compounds with three rings were the most abundant PAHs in the air and accounted for 39% (1 m from forest edge) to 73% (80 m from forest edge) of SUM-PAH in the summer and 53–59% in winter. Phenanthrene was the most abundant three-ring PAH followed by flourene. Compounds with 6–7 rings accounted for 11% (1 m from forest edge) and 0% (compounds not detected, 80 m from forest edge) of SUM-PAH in the summer but 6% (1 m) and 3% (80 m) in winter. During the summer, the decrease in the heavier 5–7 ring PAHs with increasing distance from the traffic route was much faster compared to the winter.

3.3 PAH Diagnostic Ratios

In the present study, diagnostic ratios were analysed to investigate origin (sources) of PAHs at the measurement site and as a tool to evaluate differences in composition patterns with distance from the road, as well as between summer and winter. Cross plots for four PAH diagnostic ratios are shown in Fig. 6, and the values of all calculated diagnostic ratios are given in Table 2. BghiP was below the detection limit at the two locations furthest away from the road during summer, and therefore only two locations are shown in Fig. 6b for summer. Typical PAH diagnostic ratios for emission source attribution shown in Fig. 6 are

slightly higher concentration during summer compared to winter. During both summer and winter, the phenanthrene and benzo(a)pyrene concentrations decreased with increasing logarithmic distance from the road. The decline was statistically significant ($p \leq 0.012$) except for benzo(a)pyrene during winter ($p = 0.064$).

Dibenzothiophene concentrations also decreased significantly with increasing logarithmic distance from the road ($p = 0.031$ for summer and $p = 0.040$ for winter) (Fig. 4a). However, there was no difference

Table 2 Diagnostic ratios at different distances from the forest edge within a planted spruce stand close to a major traffic route during summer and winter

Diagnostic ratios summer/winter	1 m	10 m	40 m	80 m
ANT/(ANT+PHE)	0.068/0.038	0.058/0.032	0.061/0.029	0.038/0.039
FLA/(FLA+PYR)	0.511/0.585	0.521/0.538	0.501/0.555	0.607/0.611
BaA/(BaA+CHR)	0.546/0.294	0.529/0.303	0.602/0.284	0.619/0.269
IcdP/(IcdP+BghiP)	0.407/0.331	0.327/0.319	NA/0.292	NA/0.245
BaP/BghiP	1.544/0.953	0.816/1.005	NA/1.024	NA/1.190

taken from the literature as compiled by Tobiszewski and Namiesnik (2012), see also Table S2 in the supplementary information. The calculated ANT/(ANT + PHE) ratios were below 0.1 which suggested a low temperature petroleum source (petrogenic), higher than 0.1, would indicate a combustion source (pyrogenic). The calculated FLA/(FLA + PYR) ratio > 0.5 indicated biomass/coal combustion. BaA/(BaA + CHR) ratios were > 0.35 during summer, indicating vehicular emissions, but between 0.2 and 0.35 during winter, suggested to indicate coal combustion. IcdP/(IcdP + BghiP) ratios were in the range 0.2–0.5 during both summer and winter, indicating petroleum combustion and BaP/BghiP ratios were > 0.6 both during summer and winter, indicating traffic emissions. The investigated diagnostic ratios also tended to change with distance from the forest edge, but in this study, it did not lead to a change in the indicated source. Due to the vicinity to traffic at the study site, and lack of other sources in the near surroundings, one can expect fossil fuel combustion in vehicle engines to be the dominating source at the site. However, the sources suggested by the different diagnostic ratios are not consistent and, in the case of BaA/(BaA+CHR), differ between seasons.

4 Discussion

In this study, the highest SUM-PAH concentration was measured during winter at the forest edge (10 ng m^{-3}). It was about half the concentration of the most polluted site in a study by Klingberg et al. (2022), where air concentration of SUM-PAH (including the same 32 compounds as in this study) was measured at eight sites in the city of Gothenburg during the summer of 2018, covering a wide range of air pollution conditions from the centre of the city (23 ng m^{-3}) to a peri-urban park (1.9 ng m^{-3}). An earlier wintertime study from Gothenburg, with the same type of passive sampler, showed somewhat higher levels where the sum of 13 PAHs ranged between 7.6 and 68 ng m^{-3} (median 37) (Bohlin et al., 2008). The phenanthrene concentration (2.5 ng m^{-3} in winter, 1 m from the forest edge) in this study was lower than concentrations measured in near-road environments in Lahti and Helsinki, Finland, observed by Viippola et al. (2016), ranging from on average ca 4 ng m^{-3} in summer to ca 8 ng m^{-3} in winter. The comparison indicates that the measurement

site in this study was not extremely polluted despite the vicinity to the major traffic route. The forest edge was 35 m from the traffic route permitting some dilution and dispersion of pollutants before the air mass reached the closest measurement location. However, the benzo(a)pyrene concentration (0.46 ng m^{-3} in summer and 0.36 ng m^{-3} in winter) was clearly elevated above the rural background concentrations of 0.144 ± 0.174 ng m^{-3}, reported by Garrido et al. (2014) for central and northern Europe. Compared to another European study (Saarnio et al., 2008), our results are in the same range as in Western European cities (0.1–1.1 ng m^{-3}) but lower than Eastern and Central European cities (3.0–3.2 ng m^{-3}). A study from the city of Kaunas, Lithuania, reported as high benzo(a)pyrene levels as 6.2 ng m^{-3}, measured in the PM$_{2.5}$ fraction at a site close to dense traffic (Kliucininkas et al., 2011).

The European Union (EU) has set a target value for PAH for the protection of human health expressed as the benzo(a)pyrene annual mean value, which may not exceed 1 ng m^{-3} with lower and upper assessment thresholds of 0.4 and 0.6 ng m^{-3} (EU directive 2004/107/EC). WHO estimated the reference level for benzo(a)pyrene in the ambient air to be 0.12 ng m^{-3}, which corresponds to an increase in lifetime cancer risk of 1 case per 100,000 (WHO, 2000). In this study, the concentrations of benzo(a)pyrene were below 1 ng m^{-3}, but during summer at the location closest to the road, the concentration exceeded the lower EU assessment threshold of 0.4 ng m^{-3}. Only during summer at the two locations furthest away from the road, the benzo(a)pyrene concentrations were below the WHO reference level.

Except for benzo(a)pyrene at the location closest to the road and dibenzothiophene, the PAC and NO$_2$ concentrations were higher in the winter compared to the summer measurement period. Garrido et al. (2014) also reported a consistent pattern of higher PAH concentrations in the colder months, based on measurements from 15 EMEP stations (European Monitoring and Evaluation Programme). This pattern is explained mainly by emissions, which in turn are influenced by meteorological conditions which differ between winter and summer. Since PAH is a product of the combustion of fossil fuels, a reduction of the air temperature leads to an increased heating of buildings and road traffic, two strong contributors to PAH air pollution. Eleren and Tasdemir (2021) found ambient air PAH concentrations to be higher

in the winter season in a suburban-industrial area in Turkey, due to heating and meteorological conditions restricting air mixing. Wang et al. (2020) reported the same pattern of higher concentrations during winter in two medium-size industrial cities in central China. According to the Swedish Transport Administration, the number of cars on the road at the measurement site was fewer in winter compared to summer (Trafikverket, 2020). However, an increase in background PAH concentrations (i.e. the ambient level of pollution that is not affected by local sources) during winter could have contributed to the higher concentrations at this particular site, in combination with meteorological conditions suppressing air mixing and dilution. In addition, the physiologically less active vegetation during winter can lead to decreased removal of PAHs by deposition.

Both NO_2 and PACs, with the exception of DBTs, decreased with increasing distance from the road, probably a consequence of both dilution (distance effect) and deposition to the trees. Previous studies have measured lower NO_2 concentrations at tree-covered locations compared to non-vegetated or open field locations at the same distance from nearby roads (Fantozzi et al., 2015; Grundström & Pleijel, 2014; Klingberg et al., 2017), indicating a significant effect of trees in removing NO_2 from the air. Klingberg et al. (2017) also showed this effect for particle-bound PAHs. This indicates that distance from road (permitting dilution of air pollutants) in combination with trees (promoting deposition of air pollutants), is important to reduce the risk for negative health effects in humans. The importance of different types of vegetation in mitigating air pollution of PACs needs to be further evaluated.

NO_2 is a secondary air pollutant, to a substantial extent produced from oxidation of car exhaust emissions of NO that react with O_3 in the atmosphere (Rodes & Holland, 1981). The flatter curve of concentration decline with increasing distance for NO_2 compared to PAC in this study is likely related to the influence of fast chemical reactions on the NO_2 concentration in combination with the planted spruce stand causing reduced air mixing, but needs further investigation. For PAC, the logarithmic function better reflected the fast decline close to the emission source, and the measurements were performed within a distance where the logarithmic function did not reach negative values. This finding indicates that the length

of the pollutant pathway through vegetation (i.e. distance from the roadway) is a valid proxy variable to assess the risk for health effects of traffic-related air pollutants. However, the rate of decline differed between compound and season. Therefore, the use of relationships between PAC concentrations and distance in risk assessment needs validation and calibration to local conditions, in order to avoid systematic errors.

The rate of decline in concentration with distance from the road, i.e. the slope of the regression lines in Fig. 2, differed between the different PAC categories, where H-PAH decreased much faster with distance compared to L-PAH. This result is consistent with the pattern highlighted in Fig. 5, which shows that the concentration of heavier (consisting of 5–7 benzene rings) particle-bound PAHs decreased much faster with distance compared to the gaseous PAHs, especially during summer. As a result, a person at the edge of the forest close to the road will be exposed to a substantially different mixture of PAHs compared to a few tenths of metres further into the spruce stand. Several processes influence the rate of decline with distance. Transport and dilution of pollutants released in the atmospheric boundary layer depend on the vertical distributions of wind speed, wind direction and turbulence, all strongly dependent on atmospheric stability (Arya, 1999). Differences in the roughness of the landscape can also cause differences in the rate of decline of the PAH concentration with distance from the road. High roughness increases mechanical turbulence and promotes vertical air mixing, thus increasing the rate of dispersion, leading to a steeper gradient. In addition, pollutants are deposited to vegetation and thereby removed from the atmosphere (Abhijith et al., 2017). This effect is larger during summer when the vegetation is physiologically active. The exact relative importance of these processes cannot be determined within this study. However, it would be consistent with our observations that while both particle-bound and gaseous PAHs are subject to dilution, the particle-bound PAHs appear to be deposited to the vegetation at a higher rate than the gaseous PAHs resulting in a faster decline with distance of particle-bound PAHs in this environment.

The accumulation of PAHs in leaves and needles has been shown to be proportional to the air concentrations (Klingberg et al., 2022) thereby improving the air quality. However, it has also been shown that PAH concentrations in the soil

are elevated closer to roads (e.g. Gateuille et al., 2014) as a result of the litter formed from leaves in which PAHs were accumulated. Accumulated PAHs in the soil may disrupt the food web and pose adverse impacts on soil and human health (Thakur et al., 2023).

PAH diagnostic ratios are commonly used for source attribution of PAHs (Katsoyiannis et al., 2011; Sari & Esen, 2022; Tobiszewski & Namiesnik, 2012; Wang et al., 2020). In this study, the measurements were performed close to one of the busiest roads in the Gothenburg region, and no other large combustion sources are located nearby. Despite the vicinity to traffic, the diagnostic ratios ANT/(ANT + PHE), FLA/(FLA + PYR) and BaA/(BaA + CHR) during winter did not indicate fossil fuel combustion to be the dominating source in the present study. The fact that contradictory results are obtained with different diagnostic ratios does not necessarily mean that the results are incorrect, since light PAHs are emitted from other sources than heavy PAH (Tobiszewski & Namiesnik, 2012). However, emissions from coal combustion, as suggested by e.g., the BaA/(BaA+CHR) diagnostic ratio, shown in Fig. 6, is essentially absent in Sweden (Swedish Energy Agency, 2022). Only the IcdP/(IcdP + BghiP) and the BaP/BghiP ratios consistently suggested petroleum combustions, both during summer and winter. Due to their strong resistance to photodegradation, H-PAHs are more useful than other PAH categories for source apportionment in long-term sampling, like the 4-week periods used in this study (Famiyeh et al., 2021).

Individual diagnostic ratios varied with the distance from the road despite the limited range (1–80 m from the forest edge) and underline the sensitivity to geographical variation when using diagnostic ratios for source identification. The observed spatial differences in diagnostic ratios may be due to differences in the physicochemical properties between PAH substances included in the ratios, which could influence their dynamics in the environment and, thus, the capacity to accumulate into vegetation. Diagnostic ratios may therefore not be valid as an indicator of PAH sources after advection through vegetation.

In addition, a thin organic film accumulates on impervious surfaces in urban and rural environments, which is derived from chemical molecules deposited from the atmosphere. This film provides a medium into which gas-phase and particle-bound hydrophobic

organic compounds, like PACs, can partition (Csiszar et al., 2012; Law & Diamond, 1998). Consequently, it can act as a temporary sink of chemicals such as PAHs from the atmosphere during dry and cool weather, which can be reemitted during warmer weather, and as a source to storm water and soil during rain events (Csiszar et al., 2012). Differences between PAC species in uptake in both the organic film and by vegetation, as discussed above, can at least partly explain the disparities observed for some diagnostic ratios in a number of studies (Famiyeh et al., 2021; Katsoyiannis & Breivik, 2014; Katsoyiannis et al., 2011; Tobiszewski & Namiesnik, 2012). The results of this study imply limitations in a general application of diagnostic ratios to accurately identify PAH sources in the chemical and meteorological climate of south Sweden, although they can be a useful tool for tracking differences between PAH composition patterns.

Some risk assessments of traffic-related air pollution use distance to major roads to estimate pollution levels (e.g. Hamra et al., 2015; Hoek et al., 2002). Therefore, the understanding of the variation in air pollution concentration roads, as investigated in this study, is important for accurate risk assessments. The results from this study show that the rate of decline in concentration levels with distance from the road is different for different PAC pollutants and also differs between summer and winter. Such knowledge can be used to improve estimates of human exposure to harmful traffic-related air pollutants. In addition, the result from this study provides information that can be used by stakeholders and environmental policymakers to improve air quality management and promote the well-being of the population with respect to PAC pollution.

5 Conclusions

From the present study, it can be concluded that the atmospheric concentrations of the main PAH categories (but not DBT) declined non-linearly in the Norway spruce stand with increasing distance from the forest edge. The decline in concentration, accommodating to a logarithmic relationship, was faster near the highway, which was the dominating emission source of the investigation area. Another important conclusion is that the rate of decline with increasing

distance from the forest edge was higher for PAHs consisting of a larger number of benzene rings, i.e. having a higher molecular mass and being associated with particles to a larger extent than low molecular mass PAHs with fewer benzene rings. This pattern, which was more strongly developed during the summer compared to the winter, means that not only concentrations, but also the mix of PAHs differs between different distances from the source, which has implications for the geographical variation in health risk from PAH pollution. Obviously, the quantitative (concentration) and qualitative (mix of compounds) PAH distribution within the forest stand was influenced both by deposition, affecting particle-bound PAHs more strongly than gaseous, and dilution, as the traffic-exhaust polluted air mass is advected through the forest stand. Notably, the PAH composition was more strongly influenced during the growing season when trees are physiologically active.

Diagnostic ratios were not generally indicated to be informative with respect to the dominating emission source of the investigation area, i.e. road traffic. Furthermore, the diagnostic ratios were affected by season and sensitive to the differential rate of concentration decline of different PAH species with increasing depth into the forest stand, thus not remaining constant over the relatively short distance range from the highway and forest edge covered by the study.

Data Availability   In the supplementary information, the original PAC and NO$_2$ data are available, and the measurement site is further described.

Declarations

Conflict of Interest   The authors declare no competing interests.

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