Author's response to review
Manuscript: acp-2020-201

Big thanks to the reviewers whose comments and suggestions improved the manuscript. We took the comments into account and did some changes in the manuscript. See below my point-by-point reply to the comments. The marked-up version of the manuscript, which is attached at the end of this document, shows all the changed made in the manuscript with red color. In addition to referee comments, we also did some improvements to the language and shortened the manuscript a bit by modifying the Sect. 2.1 and moving some parts to the supplementary material (this was recommended in the pre-review already). We also included one more co-author, Leena Kangas, as she provided data and helped us with answering to one of the referee comments.

Best,
Krista Luoma

Comments by the Referee #2:

Equivalent BC: Is there a different conversion for optical to mass concentration of BC from vehicle emissions vs. wood combustion?

There was no different conversion factor (mass absorption or attenuation cross section) used at different sites. We added discussion about using the same constant MAC value at all the sites:

As mentioned in the Sect. 2.2, we applied constant mass absorption cross section (MAC) values to convert the optically measured absorption data to eBC concentration. However, the MAC may vary depending on the chemical composition, shape and the mixing state of the PM. The MAC increases for aged BC particles, as the BC particles get coated with a scattering or slightly absorbing coating, which act as a lens increasing the absorption of the BC core (Lack and Cappa, 2010; Yuan et al., 2020). At TR sites, the freshly emitted BC particles from local traffic probably have no coating on the particles, but at the remote sites, however, particles are carried over longer distances and the observed BC at these sites is more aged and likely more coated. Therefore, it is probable that the real MAC at the background sites was higher compared to TR sites. If the differences in the MAC values were taken into account, it could possibly increase the difference between the traffic and background sites. The source of the BC may also have an effect on the MAC, but for example Yuan et al. (2020) and Zotter et al. (2017) did not observe notable difference between the MAC for particles originating from traffic or wood combustion. In addition to spatial variation, the MAC can also vary temporally, which could affect the observed seasonal and diurnal variations and trends (presented in Sect. 3.3) as well. However, determining the variations of MAC would require extensive

long-term measurements of chemical composition of the BC particles in different environments and therefore further analysis of the effect of MAC is omitted here.

Conclusions: Can you make any projections for the future of vehicle related BC emissions considering future changes in regulation, changes in traffic patterns, and changes in vehicle fleet type, either through better combustion, emission control orchanging energy sources, e.g. electric?

A paragraph discussing the future changes in the air quality was added in the conclusions:

This study suggests that the development in vehicle exhaust particle mitigation has been successful, at least from the viewpoint of BC and NOx emissions. With the current development, the pollution concentrations are expected to decrease in the next years as well. In general, the vehicle fleet is renewing, electric and hybrid cars are gaining popularity, and vehicles that run with biofuels or gas are becoming more common. The operator of public traffic in the HMA (HSL) aims to cut more than 90 % of their bus emissions (NOx, PM, CO2) by the year 2025 compared to the year 2010, which will improve the air quality especially at the main roads in the HMA, where several bus lines operate.

The abstract could be shortened and streamlined, see supplemental pdf file suggestions.

Thank you for the suggestions, the abstract was improved according to the supplement.

In presenting ranges throughout the MS, e.g. page 13 line 18, use that format consistently to avoid confusion between minus sign and dash, "The trends at traffic sites varied from -0.31 to -0.15 µg m-3 yr-1 and ... "

This issue was fixed and now the ranges of concentrations or trends are consisted (we used the "from … to …" format).

Page 13, line 15 at a curbside station. A trend study based only in London

This issue was fixed.

Page 17 line 2 concentration has two positive effects: 1) improved air quality and 2) decreased warming effect on the global climate by light absorbing

This issue was fixed.

Page 17 Line 6 relative trends of NOx concentration, which varied between -19.7 – -4.0 % yr-1. However, the relative trends of PM2.5 were did not decrease as rapidly as for eBC and NOx and the relative trends of PM2.5 varied between -3.9 – -2.7 % yr-1.

This issue was fixed.

Fig. 5 Annual trends for the hourly data ...

This issue was fixed.

Fig. S6 Add R value of linear fit. Add confidence limits, of slope, ie., is it significantly different than 1.0? The "trouble" with having so many data points is that one can see only the outline and not the distribution in the densest regions. To help the visual effect, plot the points in a lighter grey with the fits and limits in more intense, overlying colors.

This figure was changed so that the surface of the figure shows the number of data points in each grid point. The R^2 of the fit is presented in the figure and the standard errors of the fit are given in the caption.

Fig. S7 Add R value and statistics.

The *R* values are now presented in the caption.

Fig. S9 Notably R value seem to be much less than for S6 and S7 by my ocular analysis.

I rechecked the calculations of the correlation parameter R, but the values were still the same.

Comments by the referee #3:

The article presents a long-term field study of BC, NOx and PM2.5 at different locations in Finland. Measurements at 4 locations allow for statistical evaluation of long-term trends, which show statistically significant reduction in BC and NOx. The study concludes that the new vehicle emission standards are responsible for the reduction of traffic emissions. The article is well written and can be published in ACP with minor revisions (see below).

1. As noted by the authors, the pollutant concentrations depend both on emission rates and atmospheric dilution. To support the author claims it is critical to quantitatively assess the long-term meteorological trends, especially for wind speed (during winter) and nocturnal mixing height (during summer). The sensitivity of pollutant concentration to the meteorological parameters can be studied using a "BC versus wind speed plot" and "BC versus mixing height plot" (the same for NOx and PM2.5).

We applied the trend analysis to the WS, T and MH, which were shown to be the most important meteorological parameters that affected the PM_1 and BC concentration (Teinilä et al., 2019; Järvi et al., 2008). We added time series and the diurnal variation of the MH (Fig. S1 and S2) in the supplementary material. However, we did not include plot of BC vs. meteorological parameters, since we referred to the work by Teinilä et al. (2019) and Järvi et al. (2008), who already studied the sensitivity of the pollutants to different meteorological parameters in the HMA.

The trend analysis of the meteorological parameters did not show any statistically significant changes during the measurement period and therefore we concluded that the variations in meteorology were not the probable cause for the decreasing trends of the pollutants. We omitted the trend analysis of nocturnal MH during summer, since the MH data represented southern Finland and not the city of Helsinki. Therefore, the MH data can not be used to study if, for example, the heat island effect of the

growing urban area affected the long-term trends. We added a paragraph in the manuscript explaining the trend analysis of the meteorological parameters:

One possible cause for the decreased pollution concentrations could have been the changes in the meteorological parameters that affect the dilution. Teinilä et al. (2019) reported that in HMA the two most important meteorological parameter that affected the PM1 concentrations were wind speed (WS) and temperature (T); Järvi et al. (2008) observed that of the meteorological parameters the WS and mixing height (MH) affected the BC concentration the most. The highest concentrations were observed at low WS and MH conditions and when the T was either very high in summer or very low in winter, which indicates stable and stagnant meteorological conditions. Also, a temperature decrease during colder periods could increase the emissions from residential wood combustion. Therefore, in addition to BC, we ran the trend analysis for the time series of WS, T, and MH (time series in Fig. S1). However, we did not observe statistically significant trends for any of these parameters. We also studied the trends for the different seasons separately to see, for example, if the temperatures had increased in the summer months or decreased in winter months, but this analysis did not yield statistically significant trends either. Therefore, it is likely that the decreasing trends of the eBC concentration were not be explained by the meteorological factors.

2. Authors claim that the detached housing areas are influenced by local wood-burning emissions, the assessment being supported by lower NOx/BC ratio at DH sites compared to TR sites. For the sites where Aethalometer was used (DH4 and DH5) the source contributions should be quantitatively assessed using wavelength dependence of the aerosol absorption (Sandradewi et al., 2008).

We added a citation to Helin et al. (2018), who applied the model by Sandradewi et al. (2008) to AE33 data measured at DH3 and DH4 (note that the stations are named differently at our study than at the study by Helin et al., 2018). We added a paragraph, which discusses the results by Helin et al. (2018):

The effect of wood combustion at DH sites was studied by Helin et al. (2018) who applied AE33 data measured at TR2, DH3, and DH4 in a source apportionment model suggested by Sandradewi et al. (2008). They reported that on average about 41 and 46 % of the eBC observed at the DH4 and DH5, respectively, originated from wood combustion. The fractions were notably higher than observed at the TR2 (about 15 %). They also observed higher eBC fractions from wood combustion in the cool season: for example, eBC fractions from wood combustion were 46 and 35 % at DH3 in winter and summer, respectively. The effect of wood combustion in evenings was also evident in the data by Helin et al. (2018), who observed that both eBC from traffic and wood combustion increased towards the evening at the DH3. A comparison between weekdays and weekends at DH3 showed similar eBC concentrations originating from traffic, but slightly increased eBC concentrations from wood combustion during the weekend.

3. When comparing BC concentrations measured by MAAP at 637 nm and Aethalometer at 880 nm the wavelength dependence of the aerosol absorption should be taken into account (alternatively all measurements can be reported at

the same wavelength). For the reader's convenience, please specify the measurement wavelength of the MAAP on Page 6 Line 12.

The MAAP wavelength was added in the paragraph about MAAP measurements. As the Aethalometer was compared against the MAAP at 880 nm, we decided to stay with that wavelength. However, we added some discussion about the possible effects of the different wavelengths used. Discussion was added in the end of Sect. 3.1:

At DH4, DH5, and RB2 at least part of the measurements were conducted by an Aethalometer, which measured the eBC at 880 nm, which is longer wavelength than on what MAAP operates at (637 nm). This could have caused some difference in the measured eBC concentration in the presence of so-called brown carbon. Brown carbon is organic material, which absorbs light especially at low wavelengths (Andreae and Gelencsér, 2006). However, since the organic carbon absorbs light mainly at wavelengths below 600 nm (Kirchstetter et al., 2004) the difference between the MAAP and Aethalometer wavelengths should not cause a notable effect on the observed eBC concentration.

Spatiotemporal variation and trends of equivalent black carbon in the Helsinki metropolitan area in Finland

Krista Luoma[1], Jarkko V. Niemi[2], Minna Aurela[3], Pak Lun Fung[1], Aku Helin[3], Tareq Hussein[1,5], Leena Kangas[2], Anu Kousa[2], Topi Rönkkö[4], Hilkka Timonen[3], Aki Virkkula[3] and Tuukka Petäjä[1]

[1]Institute for Atmospheric and Earth System Research / Physics, Faculty of Science, University of Helsinki, P.O. Box 68, 00014 Helsinki, Finland
[2]Helsinki Region Environmental Services Authority, P.O. Box 100, 00066 Helsinki, Finland
[3]Atmospheric Composition Research, Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland
[4]Aerosol Physics Laboratory, Faculty of Engineering and Natural Sciences, Tampere University, P.O. Box 692, 33014 Tampere, Finland
[5]Department of Physics, The University of Jordan, 11942 Amman, Jordan

Correspondence to: Krista Luoma (krista.q.luoma@helsinki.fi)

Abstract: In this study, we present results ~~of~~ from 12 years of black carbon (BC) measurements at 14 ~~different measurement~~ sites around ~~the~~ Helsinki metropolitan area (HMA) and at one background site outside the HMA. The main local sources of BC in the HMA are traffic~~,~~ and residential wood combustion in fireplaces and sauna stoves. All the BC measurements were conducted optically and therefore we refer to the measured BC as equivalent BC (eBC). Measurement stations were located ~~at~~ in different ~~types of~~ environments that represented traffic environment ~~(six sites)~~, detached housing area ~~(five sites)~~, urban background ~~(two sites)~~, and regional background. ~~(two sites).~~ The measurements of eBC were conducted ~~during~~ from 2007 – through 2018; however, the times ~~period~~ and the lengths of the time series varied ~~from~~ at each site ~~to site~~. ~~As expected, t~~The largest annual mean eBC concentrations were measured at the traffic sites (from 0.67 –to 2.64 μg m^{-3}) and the lowest at the regional background sites (from 0.16 –to 0.29 μg m^{-3}). The annual mean eBC concentrations at the detached housing and urban background sites varied from ~~in the range of~~ 0.64 –to 0.80 μg m^{-3} and ~~the annual mean eBC concentrations at the urban background sites varied in the range of~~ from 0.42 –to 0.68 μg m^{-3}, respectively.~~.~~ The clearest seasonal variation was observed at the detached housing sites, where ~~the~~ residential wood combustion increased the eBC concentrations during the cold season. D~~Traffic rates and wood burning influenced the d~~iurnal ~~and weekly~~ variation~~s~~ of eBC concentration in different ~~types of~~ environments depended clearly on the local traffic and residential wood combustion. The dependency was not ~~so~~ as clear for the other air pollutants, which were ~~here~~ NO_x and mass concentration of particles smaller than 2.5 μm ($PM_{2.5}$). At four sites, which had at least four-year-long time series available~~, we observed that~~ the eBC concentrations had statistically significant decreasing trends that ~~, which~~ varied from~~in the range of~~ -10.4 to~~–~~ -5.9 % yr^{-1}. Compared to ~~the~~ trends determined at ~~the~~ urban and regional background sites, the absolute trends decreased ~~the~~ fastest at ~~the~~ traffic sites~~, and~~ especially during the morning rush hour. ~~The r~~Relative long-term trends of eBC and NO_x were similar ~~to each other~~, and their concentrations decreased more rapidly than that~~the concentration~~ of $PM_{2.5}$. The results indicated that especially ~~the~~ emissions from traffic have decreased in

the HMA during the last decade. This shows that air pollution control, new emission standards and a newer fleet of vehicles ~~really have~~had an effect ~~in the~~on air quality.

1 Introduction

Air pollution is one of the biggest environmental health risks in the world. Air pollution consists of both gaseous components and particulate matter (PM). Lelieveld et al. (2015) estimated that particles smaller than 2.5 µm in diameter ($PM_{2.5}$) and ozone (O_3) together caused about 3.3 million premature deaths globally in 2010. Majority of these premature deaths were due to $PM_{2.5}$ (approximately 1.9 million). More than 65 % of the premature deaths caused by $PM_{2.5}$ were related to cardiovascular diseases, the second largest cause for the $PM_{2.5}$-related premature deaths were lung and respiratory diseases, and a small fraction ~~of the premature deaths~~ were due to lung cancer (Lelieveld et al., 2015). The causes for the adverse health effects of PM are the PM induced inflammation and toxic materials, which are transported in the respiratory system by the particles.

WHO reported that PM emitted from combustion sources, especially from traffic, is more harmful for health than PM from other sources (Krzyżanowski et al., 2005). Typical combustion sources, which are also discussed in this study, are traffic and domestic wood burning. Traffic emits a complex mixture of gaseous and fine particulate compounds (Rönkkö and Timonen, 2019). Domestic wood burning also emits fine particles including toxic compounds, such as benzo(*a*)pyrene (Hellén et al., 2017). Black carbon (BC), which is defined as black carbonaceous particulate matter, is a good indicator of pollution from combustion because it is a side product of incomplete combustion. Therefore, it is useful to measure BC alongside with the other air quality parameters since BC concentration can give additional information about the health effects of aerosol particles than just PM, which does not only originate from combustion ~~sources~~ (Janssen et al., 2011). BC is not just an indicator on bad air quality, but the BC particles themselves also have adverse health effects. BC particles, which are around the size range of ~100 nm, can penetrate deep in the respiratory system and all the way in the alveolar region, from where the particles can be transported in the blood circulation system and further on in the organs.

Due to its black appearance, BC absorbs solar radiation and decreases the albedo of reflecting surfaces (i.e., snow and ice sheets). BC has been estimated to be one of the largest warming agents in the climate change (Stocker et al., 2013). Since BC is emitted in the air as particles, its lifetime in the atmosphere is relatively short (few weeks) compared to greenhouse gases (tens of years). Therefore, in addition to improving air quality, cutting down the BC emissions would have rather fast effects in the radiative forcing slowing down the rate of global warming. In order to reduce BC emissions there has been, for example, a suggestion of implementing a BC footprint similar to the CO_2 footprint ~~-~~(Timonen et al., 2019).

Several recent studies have reported decreasing long-term trends of BC concentration in different types of environments including urban areas (e.g., Sun et al., 2020; Kutzner et al., 2018; Singh et al., 2018; Font and Fuller, 2016). In Finland, the

concentration of BC and its trends has been studied especially in background sites (Hienola et al., 2013; Hyvärinen et al., 2011), but not that much in urban areas. The previous studies about BC concentration in the Helsinki ~~M~~metropolitan ~~A~~area (HMA) are mainly from shorter campaigns (Aurela et al., 2015; Dos Santos-Juusela et al., 2013; Helin et al., 2018; Järvi et al., 2008; Pakkanen et al., 2000; Pirjola et al., 2017).

The objective of this study is to investigate the spatio-temporal variability of BC in different types of environments in the HMA. Our objective is also to clarify how the proximity of combustion sources, which in HMA are mainly traffic and ~~domestic~~ residential wood ~~burning~~ combustion (Helin et al., 2018; Savolahti et al., 2016), affected the BC concentrations. In this study, we utilize BC data measured at various different locations in the HMA and at one site outside the HMA. The measurements were conducted during 2007~~–~~–2018. To study how the variability of BC differed~~s~~ from the variability of the monitored and regulated air quality parameters, we also included parallel measurements of $PM_{2.5}$ and NO_x in this study. ~~Measuring BC in addition to $PM_{2.5}$ gives a better view on the effect of local pollution sources and more hazardous air pollutants. Understanding the anthropogenic sources of pollution helps overcoming the problems related to poor air quality.~~

2 Measurements and methods

2.1 The field sites

The HMA consists of four different cities (Helsinki, Espoo, Vantaa, and Kauniainen) and it is the most densely populated area in Finland. ~~T~~In 2020, the total population in the HMA wa~~i~~s about 1.4~~2~~ million. Helsinki is the capital of Finland and it is located in the southern coast of the country (60°10'N, 24°56'E). The HMA locates on the seaside of the Gulf of Finland and therefore the climate in the area is a transition zone between oceanic and continental climate and typically defined as humic continental climate. On average, the ~~The~~ coldest month~~ly mean temperature (-5 °C)~~ during the measurement period (2009~~–~~2018) ~~occurred~~ was ~~in~~ February (-5 °C) and the warmest month ~~ly mean temperature (19 °C) in~~was July (19 °C). The mean wind speed over the whole measurement period was about 4 m s^{-1}. For more detailed information about the meteorological parameters, see Fig. S1.

The measurements of BC concentration were conducted at 15 sites. We classified the stations into four categories: traffic environment (TR), detached housing area (DH), urban background (UB), and regional background (RB). The locations of the measurement sites are presented in ~~-~~Fig. 1; the exact coordinates and aerial photos of the stations are provided in Sect. S2. Fourteen of the measurements sites were located in the HMA and one of the RB sites was located circa 200 km north of the HMA in Hyytiälä.

~~One of the sites, which represented the RB, was located circa 200 km north of the HMA in Hyytiälä.~~ At some of the stations, the measurements have been repeated or conducted on a long-term basis and at some of the stations, the measurements lasted

only for one year. The measurement periods for each site are shown in Table 1. ~~Most of the stations were operated by the Helsinki Region Environmental Services Authority (HSY), which is the authority monitoring the air quality in the HMA.~~

Six traffic stations, ~~(~~TR1 – TR6~~)~~, located close to a busy street or road (~~Fig. S3~~Sect. S2.1~~), were categorized as traffic stations~~). At these sites, traffic was the dominating source of pollution. Detailed information, such as traffic counts (TC), fraction of heavy-duty (HD; i.e., trucks and buses), and the distance of the station to the closest traffic lines are presented in Table 2.

Detached housing s~~S~~tations, DH1~~, DH2, DH3, DH4, and~~ – ~~-~~DH5, ~~were~~ located in residential areas that consisted of separate one-family houses, small streets, and some forests and parks (Sect. S2.2). ~~Vartiokylä, Ruskeasanta, Lintuvaara, Rekola, and Itä Hakkila, respectively (Fig. S4). These residential areas consisted of separate one-family houses, small streets, and some forests and parks.~~ The traffic rates at these areas were ~~small streets next to these stations were~~ low~~;~~. ~~F~~ for example, the traffic rates on the streets next to DH1 and DH5 were ~~estimated to be~~ about 2 600 vehicles per day, which was ~~, which is notably~~way less than ~~close to~~at the TR sites (Table 2). According to a survey study, up to 90 % of the one-family households burned wood to warm up houses and/or saunas, but less than 2 % of the households used wood combustion as the main heating source ~~In 2014, the HSY conducted a large survey about wood combustion in the detached houses in the HMA~~ (HSY, 2016, in Finnish only; Hellén et al., 2017). ~~In the study, HSY estimated that 90 % of the households burn wood to warm up houses and/or saunas. However, less than 2 % of the households use wood burning as the main heating source.~~

Background sites were categorized as urban and regional background sites. Urban background stations, UB1 and UB2, located close to the central area but not in vicinity of busy roads (Sect. S2.3). UB2 is also known as SMEAR III (Station for Measuring Ecosystem-Atmosphere Relations; Järvi et al., 2009). Regional background stations, RB1 and RB2, located in rural areas, about 20 and 200 km away from Helsinki, respectively (Sect. S2.3). RB sites represented the concentration levels outside the urban area without local pollution sources. RB2 is also part of the SMEAR network, better known as SMEAR II (Hari and Kulmala, 2005).

~~**2.1.1 Traffic stations**~~

~~Six stations (TR1 – TR6), located close to a busy street or road (Fig. S3), were categorized as traffic stations. At these sites, traffic was the dominating source of pollution. Detailed information, such as traffic counts (TC), fraction of heavy-duty (HD; i.e., trucks and buses), and the distance of the station to the closest traffic lines are presented in Table 2.~~

~~TR1 located right in the city center and the station represented the concentration level that pedestrians are exposed to in the city center. TR2 and TR3 located in street canyons, where tall buildings frame the streets. TR2 was located next to Mäkelänkatu, which is one of the main streets leading to Helsinki city center, and TR3 was located in the northern part of~~

Mannerheimintie, another main street to Helsinki city center. In addition to passenger cars, many busses (heavy duty) bypass the TR2 and TR3.

While, TR1, TR2, and TR3 were in or close to the Helsinki city center, TR4 was located next to a ring road, Kehä I, which is a highway going around the Helsinki city. The station represented the concentration level on walkways and bus stops that are next to major roads.

TR5 and TR6 were located close to busy streets and intersections in the city centers of Vantaa and Espoo, respectively. These city centers were not as large as the city center of Helsinki, but there were still busy roads and apartment buildings. As seen in Fig. 1, TR5 and TR6 were closer to the detached housing areas than the other TR stations.

2.1.2 Stations in the detached housing areas

Stations DH1, DH2, DH3, DH4, and DH5 were located in Vartiokylä, Ruskeasanta, Lintuvaara, Rekola, and Itä-Hakkila, respectively (Fig. S4). These residential areas consisted of separate one family houses, small streets, and some forests and parks. The traffic rates at the small streets next to these stations were low. For example, the traffic rates on the streets next to DH1 and DH5 were estimated to be about 2 600 vehicles per day, which is notably less than close to the TR sites (Table 2). In 2014, the HSY conducted a large survey about wood combustion in the detached houses in the HMA (HSY, 2016, in Finnish only; Hellén et al., 2017). In the study, HSY estimated that 90 % of the households burn wood to warm up houses and/or saunas. However, less than 2 % of the households use wood burning as the main heating source.

2.1.3 Urban background stations

The UB stations represented the concentration levels that people are generally exposed to in urban areas in Helsinki. The UB stations, UB1 and UB2, were located in the close to the central area but not in vicinity of busy roads (Fig. S5a-b). UB1 was located in the Kallio district next to a sports field and close to Helsinginkatu, a moderately busy street, where the average working day traffic count was about 5 000 vehicles per day (5 % heavy duty). The area consisted mostly of apartment buildings. UB2 was located in Kumpula, in the campus of the University of Helsinki. UB2 is also known as SMEAR III (Station for Measuring Ecosystem-Atmosphere Relations; Järvi et al., 2009) and it is run in collaboration by the University of Helsinki and Finnish Meteorological Institute (FMI). There was a busy street, Hämeentie, located after 200 m wide forest band. The traffic count at Hämeentie was about 40 000 vehicles per day.

~~2.1.4 Regional background stations~~

~~The RB stations represented the concentration levels outside the urban area without any main local sources (Fig. S5c-d). The RB stations were mainly affected by regional and long-range transported pollutants. RB1 was located in Luukki, which is a rural area in Espoo about 23 km away from the Helsinki city center. RB2 was located almost 200 km away from Helsinki in Hyytiälä. RB2 was stationed in a boreal forest, far away from any pollution sources. RB2 was included in this study to compare the pollution levels in a city to those observed in a remote countryside station. RB2 is part of the SMEAR network and it is also known as SMEAR II .~~

~~2.1.4 Meteorological station~~

~~The meteorological station measuring wind direction (WD), wind speed (WS), temperature (T), pressure (p), relative humidity (RH), and precipitation, was located in a rooftop (78 m a.s.l) in the Pasila district close to central Helsinki. In this study, we used the measurements of meteorological parameters conducted in Pasila to represent the meteorological conditions of all the stations located in the HMA. Also, the air quality stations measured the meteorological parameters, but four meters above the ground where the local factors, such as trees and buildings, may affect the measurements.(Karppinen et al., 2000)~~

2.2 Measurements of equivalent black carbon

~~Most of the stations were operated by the~~The Helsinki Region Environmental Services Authority (HSY), which is the authority monitoring the air quality in the HMA,~~.~~ ~~HSY~~ arranged the measurements at 13 of the sites.~~,~~ ~~University of Helsinki~~Institute for atmospheric and Earth system research (INAR) ~~conducted~~ arranged the measurements at the RB2, and the Finnish Meteorological Institute (FMI) together with ~~the University of Helsinki~~INAR conducted the measurements at the UB2.

All the BC measurements were conducted optically, meaning that ~~and~~ the BC concentration was derived from the light absorption of the particles and hence we refer to the measured BC as equivalent black carbon (eBC; Petzold et al., 2013). At 11 of the stations, the measurements of eBC were conducted by using only a Multi-angle absorption photometer (MAAP; Thermo Fisher Scientific, model 5012), and at ~~four~~three of the stations (DH4, DH5, and RB2) all or at least part of the measurements were conducted by using an Aethalometer (Magee Scientific, models AE31 and AE33). The instruments used at different sites are listed in Table S1.

At all of the ~~measurement~~ stations, the head of the sampling line was located 4 m above the ground. The concentration of eBC was measured for particles smaller than 1 µm (PM_1). However, at DH1 the eBC concentration was measured for $PM_{2.5}$ for first half of the year, but since most of the eBC mass concentration falls in the PM_1 (Vallius et al., 2000), the cut-off size should

not have caused big difference in the results. Sample air was dried with an external dryer or by warming up the sample to 40 °C at most of the stations, but at TR1, UB2, DH5 and DH4 (only the first half of year) the sample air was not dried. Even if there was no drier, the sample air warmed up to the room temperature, which decreaseds the RH, when the outdoor temperature wais lower than the indoor temperature (i.e., the sample air wais dried passively during the cold period, however, in summer, when the temperature difference wais smaller, the RH does did not necessarily decrease). At 11 of the stations, the measurements of eBC were conducted by using only a Multi-angle absorption photometer (MAAP; Thermo Fisher Scientific, model 5012), and at four of the stations (DH4, DH5, and RB2) all or at least part of the measurements were conducted by using an Aethalometer (Magee Scientific, models AE31 and AE33). The instruments used at different sites are listed in Table S1.

The MAAP determines the absorption coefficient of aerosol particles by collecting the particles on a filter medium and by measuring the intensity of light penetrating the filter and the intensity of light that is scattered from the filter in two different angles (Petzold and Schönlinner, 2004). The absorption coefficient is determined from these measurements by using a radiative transfer scheme. The eBC concentration is obtained from the absorption coefficient by using a mass absorption cross-section (MAC) value of 6.60 $m^2 g^{-1}$ (Petzold and Schönlinner, 2004). The MAAP operates only at one wavelength, which is 637 nm.

The Aethalometer measures the eBC concentration at seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm). Here we chose to use the 880 nm channel, since it is the recommended and most commonly used wavelength to report the eBC measured by an Aethalometer. and in this study the wavelength 880 nm was used. Similarly to MAAP, tThe Aethalometer collects the sample aerosol particles on the filter material similarly to the MAAP, but unlike the MAAP, the Aethalometer only measures the attenuation of light through the filter. Therefore, the Aethalometer is prone to error caused by the increasing filter loading. In the newer model, AE33, this is automatically taken into account in real time as the instrument applies the so-called "dual-spot correction" to the data (Drinovec et al., 2015). For AE33, the recommended MAC value of 7.77 $m^2 g^{-1}$ at 880 nm is recommendedwas used in this study (Drinovec et al., 2015). For the older model, AE31, a correction algorithm needs to be applied by the user (e.g., Collaud Coen et al., 2010). AE31 determines the BC concentration from the so-called attenuation coefficient and it uses mass attenuation cross-section value of 16.62 $m^2 g^{-1}$ at 880 nm.

At DH4, model AE33 Aethalometer was used for the first half of the measurement period (1 January 2017 – 5 May 2017) and at DH5, the whole data set was measured with an AE33. At these two stations, HSY corrected the eBC concentration by multiplying the concentration by 0.75, according to a comparison with MAAP. At RB2, an older model AE31 was used. The AE31 was first corrected for the filter loading error by using the correction algorithm suggested by Virkkula et al. (2007). After the filter loading correction, a comparison with MAAP showed that the AE31 data had to be multiplied by 1.08 to acquire similar concentrations (Sect. S3).

2.3 Measurements of NO_x and $PM_{2.5}$

NO_2 and the mass of particles smaller than 2.5 µm ($PM_{2.5}$) are regulated pollutants based on the Air Quality Directive (2008/50/EC) and therefore they are always measured at the air quality stations. In this study, we used NO_x ($NO + NO_2$) data instead of the regulated NO_2, since NO_x describes better the primary traffic emissions. Even though there was much more NO_x ~~($NO + NO_2$)~~ and $PM_{2.5}$ data available compared to eBC data, ~~in this study~~ we used only NO_x and $PM_{2.5}$ data that was measured ~~during the same period as~~parallel to eBC ~~was measured~~ in order to make the comparison and trend analysis systematic.

The $PM_{2.5}$ concentration was measured with various different instruments, which are listed for each station in Table S1. The instruments were based on four different methods: 1) attenuation of β-radiation (Thermo model FH 62 I-R); 2) tapered element oscillating microbalance (TEOM; Thermo different models); 3) optical detection (Grimm 180); and 4) collecting the particles in a cascade impactor and manually weighting the collected particles. The instruments, which use the methods 1 – 3, measure the concentration continuously. At RB2, where the $PM_{2.5}$ concentration was measured by collecting the particles in a cascade impactor and weighting the collected particles about three times a week so the time interval of these measurements varied from two to three days. To compare the $PM_{2.5}$ measurements to BC concentration at RB2, the $PM_{2.5}$ concentration was interpolated to match the timestamps of the eBC ~~timestamps~~measurements.

NO_x mass concentration [NO_x] was derived from the measurements of NO and NO_2 so that

$$[NO_x] = 1.533 \cdot [NO] + [NO_2]. \quad (1)$$

The mass concentration of NO and NO_2 were measured by instruments, which are based on the chemiluminescence method. The instruments used at each station are listed in Table S1.

2.4 Meteorological measurements

The meteorological station measuring wind direction (WD), wind speed (WS), temperature (T), pressure (p), relative humidity (RH), and precipitation, was located in a rooftop (78 m a.s.l) close to central Helsinki (Fig. 1). In this study, we used the measurements of meteorological parameters conducted at the rooftop station to represent the meteorological conditions of all the stations located in the HMA. We also included data of the mixing height (MH) for southern Finland. The MH was calculated by MPP-FMI, a meteorological pre-processing model developed by the FMI (Karppinen et al., 2000).

2.4~~-5~~ Data processing

The data quality were assured by the data producer and invalid data were omitted from further analysis. ~~At TR1, all the $PM_{2.5}$ data between 8 – 9 a.m. and 9 - 10 p.m. had to be omitted due to technical issues caused by the disturbance of the air conditioning.~~ The concentrations were converted to ambient outdoor temperature and to normal atmospheric pressure (1013.25

hPa). In this study, we used one-hour averages for all the variables (only exception is the $PM_{2.5}$ data at RB2). The hourly mean values were calculated if the hour had at least 75 % of valid data. The hour of day always refers to the local time (note that winter and summer times are used in Finland) and the time stamp of the measurements are reported in the middle of the averaging period.

2.45.1 Trend analysis

We used seasonal Mann-Kendall test and Sen's slope estimator (Gilbert, 1987) in determining the statistical significances and the slopes of the long-term trends. Mann-Kendall test and Sen's slope estimator are non-parametric statistical methods, which allow missing data points. The method determines the trends for each season (here we used months) separately and tests if the trends for different seasons are homogeneous. We used monthly median values in the trend analysis and we required at least 14 days of valid data for each month, otherwise the month was not taken into account in the trend analysis. Similar analysis has been used in various trend studies (e.g., Collaud Coen et al., 2007; Collaud Coen et al., 2013; Li et al., 2014; Zhao et al., 2017).

3 Results and discussion

3.1 Spatial variation

The statistics of eBC, $PM_{2.5}$, and NO_x concentrations from each site are presented in Fig. 2 ~~Figure 2~~. The figure includes all the data and the statistics were determined by using the 1 h mean values. Figure 2 shows that the arithmetic mean values differed from the median values, which means that the data of the air pollutants was not normally distributed at any station and that the data was skewed to the right (i.e., small concentrations occurred more often and therefore the median values were smaller than the means).

As expected, the highest mean eBC concentrations were observed at the TR sites, where the mean eBC concentration varied from 0.77 to 2.08 µg m^{-3} (~~measured~~ at ~~the~~ TR6 and TR3, respectively). At the DH sites, the mean eBC concentration varied from 0.64 to 0.80 µg m^{-3}, which were rather similar mean values as observed at the TR1, TR5, and TR6 (0.84, 0.83, and 0.77 µg m^{-3}, respectively). At the UB sites, the mean eBC concentrations were around 0.52 µg m^{-3}, which ~~were~~ was clearly lower than at the TR and DH sites. The lowest mean eBC concentrations, which were ~~(~~0.28 µg m^{-3}~~,)~~ were observed~~, as expected,~~ at the RB sites that had no local BC sources in vicinity.

Previous studies have shown that in addition to the traffic count, the BC concentration near traffic lines depends on various factors: the distance to the traffic lines (Enroth et al., 2016; Massoli et al., 2012; Zhu et al., 2002); the speed limit (Lefebvre et

al., 2011); and the fraction of heavy-duty vehicles (Clougherty et al., 2013; Weichenthal et al., 2014). The surrounding buildings, vegetation and the wind conditions affect the dilution and therefore the BC concentrations as well (Abhijith et al., 2017; Brantley et al., 2014; Pirjola et al., 2012). Also, close by intersections may affect the BC concentrations if they induce traffic build-ups; BC emissions from vehicles that accelerate are higher than the emissions from a steadily moving vehicle (Imhof et al., 2005).

The abovementioned factors are probably the reason for the relatively big differences between the different TR stations and explain, for example, why the eBC concentration at TR3 was notably higher than at the other TR sites. TR3 was located in a street canyon right next to a very busy traffic line~~., which also has a rather high fraction of heavy-duty vehicles.~~ According to Table 2, the traffic count on the closest street next to TR3 was around 44 400 vehicles per weekday and the fraction of heavy-duty vehicles was 14 %. The traffic count and fraction of heavy-duty vehicles wa~~i~~s higher than for example on the street next to TR1 or at the street next to TR2. The area around TR3 consisted~~s~~ also of a few busy intersections and the ~~station is located~~location in a street canyon ~~, which~~ probably increased the eBC concentrations even further.

The effects of total traffic count and traffic count of heavy-duty vehicles (HD) on the eBC concentration were studied in more detail in the supplementary material (Fig. S9), where the annual mean eBC concentrations were compared against the estimated weekday traffic counts of all vehicles and of ~~heavy duty~~HD vehicles in the nearest street or road. The annual means of eBC concentration correlated better with the number of ~~heavy duty~~HD vehicles passing the station ~~in a day~~ (R = 0.82) than with the total traffic count ~~during the day~~ (R = 0.71). In general, the number of ~~heavy duty~~HD vehicles passing the TR1, TR5, and TR6 per day was low compared to TR2, TR3, and TR4, and this was also seen in the mean concentration of eBC, which was the highest for the TR2 – TR4. This result~~e effect of heavy duty vehicles~~ was expected, since the BC emissions from ~~heavy duty~~HD vehicles are higher compared to light-duty vehicles (Imhof et al., 2005).

~~It must be noted, that t~~The distance from the stations to the edge of nearest street ~~varies~~varied, which has an ~~a~~effect~~s~~ on the measured eBC concentration (Massoli et al., 2012; Zhu et al., 2002). A study by Enroth et al. (2016) estimated that eBC concentration ~~decreases~~ decreased by ~~to a half~~50 % at 33 m distance from the road. TR2 and TR3, where we observed rather high concentrations, were located right next to the street (in a 0.5 m distance), whereas the other stations had a longer distance to the nearest street (3 – 20 m). Previous studies have also shown that tall trees in street canyons may deteriorate the air quality by preventing dispersion (Abhijith et al., 2017) and this may also have affected the higher measured concentrations at TR2 and TR3~~., since i~~In Mäkelänkatu street, which is next to TR2, there are two lines of trees framing the tram lines in middle the street (~~see~~ Fig. ~~S2b~~S3b) and in Mannerheimintie street, which is next to TR3, there are trees planted between the traffic lines and pavements (~~see~~ Fig. ~~S2c~~S3c).

Compared to the HMA, ~~H~~higher concentrations of eBC ~~than observed at this study~~ have been reported at other urban areas in Europe. For example, ~~reported mean eBC values of 3.7 μg m^{-3} at a traffic site and 2.33 μg m^{-3} at an urban background site measured in Madrid in 2015.~~ Sun et al. (2019) reported median values of 2.0, 0.9, and 0.4 μg m^{-3} at several German TR, UB and RB sites, respectively, measured during 2009–2018. Singh et al. (2018) reported on average eBC concentrations of 1.83 and 1.34 μg m^{-3} measured at several urban center and ~~urban background~~UB sites in the United Kingdom during 2009–2011. Krecl et al. (2017) observed mean eBC concentration of 2.1 μg m^{-3} at a street canyon site in Stockholm during weekdays in spring 2013. Becerril-Valle et al. (2017) reported mean eBC values of 3.7 μg m^{-3} at a TR site and 2.33 μg m^{-3} at an UB site measured in Madrid in 2015. The BC concentrations reported by other studies at different environments are higher than the eBC concentrations measured at corresponding environments ~~around~~ in the HMA. Already the concentrations at the RB sites are lower than those measured at other European sites. Generally, the air quality in the HMA and in southern Finland wa~~i~~s good. Finland is isolated from the more populated continental Europe by the Baltic sea and therefore long-range pollution from the more polluted continental area affects southern Finland less. ~~d~~Due to its coastal location, the HMA affected by the sea breeze and is therefore well diluted. ~~which enhances the dilution, and~~Compared to other European metropolitan areas, the HMA ~~relatively~~is small ~~size, since~~and the area is not as densely populated ~~than~~ as many other European capital areas~~.~~

In addition to eBC, we also studied the variations of ~~NO$_x$ and~~ PM$_{2.5}$ and NO$_x$, presented in Figs. 2b and 2c, respectively~~, which were measured at all of the stations~~. The spatial variation of NO$_x$ was partly similar to that of eBC; the highest concentrations were measured at the TR sites and the lowest at the RB sites. At the TR sites, the mean concentrations varied between from 44 ~~–~~to 147 μg m^{-3} (~~lowest concentration~~ at ~~-~~TR5 and ~~highest at~~ TR3, respectively) and the variation between the stations was rather similar to the variation of eBC. Like BC, NO$_x$ is highly dependent on the traffic related parameters such as the traffic count, the fraction of heavy traffic, the speed limit etc., which explain the similar variation observed. For the RB sites, however, the mean NO$_x$ (2 μg m^{-3}) was relatively low compared to eBC at RB sites. Another difference to eBC was that, the NO$_x$ concentration at the DH sites was relatively lower, which was expected, since the NO$_x$ emissions from residential wood combustion are low. The correlation between eBC and NO$_x$ concentrations at each station are presented in Fig. ~~S7a~~S8a. As the likeness in the spatial variability already suggested, the eBC and NO$_x$ had rather similar sources and therefore they were expected to correlate. The correlation coefficient (R) between these variables was the highest at the TR stations ($0.80 \leq R \leq 0.90$) and lower at the DH stations ($0.63 \leq R \leq 0.73$). At the background sites, the correlation coefficient varied more ($0.55 \leq R \leq 0.83$).

For ~~the~~ PM$_{2.5}$, there were not as clear patterns between different station categories as there were for eBC and NO$_x$. At the ~~traffic~~ TR sites, the mean PM$_{2.5}$ concentration varied from 5.6 to 11.3 μgm^{-3} (at TR6 and TR3, respectively) and at the ~~detached housing~~DH sites, the variation was rather similar: from 5.6 to 11.3 μgm^{-3} (at DH4 and DH2, respectively). ~~For PM$_{2.5}$, t~~The mean concentration of PM$_{2.5}$ at the background sites w~~ere~~as not as clearly lower as it was for eBC and NO$_x$. These results show that PM$_{2.5}$ did not depend on local primary pollution sources as much as eBC or NO$_x$ did. PM$_{2.5}$ includes all different kind of

aerosol particles, especially secondary aerosol, which may be anthropogenic or biogenic in origin. In this size range, non-anthropogenic particles (e.g., secondary particles of biogenic origin; Dal Maso et al., 2005) are also contributing. The differences in the sources of eBC and $PM_{2.5}$ concentrations were also seen in the correlation between these two variables; the *R* between these two variables at different stations were notably lower ($0.36 \leq R \leq 0.67$) than the *R* between eBC and NO_x concentrations (Fig. S7b).

The fraction of eBC in the $PM_{2.5}$ is shown in Fig. 3 ~~and it represent how big fraction of $PM_{2.5}$ consists of eBC~~. eBC was measured mostly in PM_1, but since most of the BC particles are smaller than 1 µm in diameter, the difference between the cut off diameters should not have a big effect on the results ~~-~~ (e.g., Enroth et al., 2016). Higher eBC/$PM_{2.5}$ ratio indicate~~d~~s that there wa~~i~~s a larger fraction of PM related to combustion sources. The highest median eBC/$PM_{2.5}$ ratio were observed at the TR sites where the ~~ratio~~ fraction of eBC varied from 10 to 15 %. The second highest median ratios were observed at the DH sites and at UB1 where the ratios varied from 5 to 9 %. At the RB sites, the median fractions were the smallest: less than 5 %.

The results of the spatial variation showed that eBC concentration and eBC/$PM_{2.5}$ ratio depended greatly on the distance to the pollution sources, which ~~are~~were, in this case, traffic and wood ~~burning~~combustion. The NO_x wa~~i~~s very dependent on the distance to the traffic sources only, since NO_x concentration wa~~i~~s not significantly affected by residential wood ~~burning~~combustion. Since the $PM_{2.5}$ has ~~many~~ various sources and generally high background levels, it wa~~i~~s the least dependent component on the contribution of the local sources.

Figures 2 and 3 included all the data that was collected from 2009 to 2018 and that the sizes of the data sets at each station differed. Therefore, the year-to-year variation caused by the meteorological conditions and changes in the emission rates might have affected the results of spatial variability especially at sites that contained only one year of data (all DH sites and TR4).

At DH4, DH5, and RB2, at least part of the measurements were conducted by an Aethalometer. The Aethalometer measured the eBC at 880 nm, which is a longer wavelength than on what MAAP operated at (637 nm). This could have caused some difference in the measured eBC concentration in the presence of so-called brown carbon. Brown carbon is organic material, which absorbs light especially at short wavelengths (Andreae and Gelencsér, 2006). However, since the organic carbon absorbs light mainly at wavelengths below 600 nm (Kirchstetter et al., 2004) the difference between the MAAP and Aethalometer wavelengths should not cause a notable effect on the observed eBC concentration.

As mentioned in the Sect. 2.2, we applied constant mass absorption cross section (MAC) values to convert the optically measured absorption data to eBC concentration. However, the MAC may vary depending on the chemical composition, shape and the mixing state of the PM. The MAC increases for aged BC particles, as the BC particles get coated with a scattering or slightly absorbing coating, which act as a lens increasing the absorption of the BC core (Lack and Cappa, 2010; Yuan et al.,

2020). At TR sites, the freshly emitted BC particles from local traffic probably have no coating on the particles, but at the remote sites, however, particles are carried over longer distances and the observed BC at these sites is more aged and likely more coated. Therefore, it is probable that the real MAC at the background sites was higher compared to TR sites. If the differences in the MAC values were taken into account, it could possibly increase the difference between the traffic and background sites. The source of the BC may also have an effect on the MAC, but for example Yuan et al. (2020) and Zotter et al. (2017) did not observe notable difference between the MAC for particles originating from traffic or wood combustion~~(Yuan et al., 2020; Zotter et al., 2017). (Schwarz et al., 2008)~~ In addition to spatial variation, the MAC can also vary temporally, which could affect the observed seasonal and diurnal variations and trends (presented in Sect. 3.2) as well. However, determining the variations of MAC would require extensive long-term measurements of chemical composition of the BC particles

in different environments and therefore further analysis of the effect of MAC is omitted here.

~~It must be noted, that the and 3 include all the data that was collected from 2009 to 2018 and that the sizes of the data sets for each station differ. Therefore, the year to year variation caused by the meteorological conditions and changes in the emission rates might have affected these results as some sites contained only one year of data (all DH sites and TR4).~~

3.~~3~~ 2 Temporal variation

3.~~3~~2.1 Long-term trends

A quick look in Table 1 already showed that the annual eBC mean values had seemingly decreased. To see if the decreasing eBC trend had a statistical significance, we applied the seasonal Kendal test (see Sect. 2.4.1) to the data sets that were at least four-year-long (TR1, TR2, RB2, and UB1). Even though there was four-years of measurements at the UB2, ~~it~~ the station was omitted from th~~is~~e trend analysis, since the data availability at UB2 in 2016~~–~~2017 was not good enough. The seasonal Kendal test was applied to monthly medians values, which are presented in Fig. 4 for the eBC concentration at TR1, TR2, UB1, and RB2. We could not apply the trend analysis to any of the DH stations, since none of the DH stations had more than one year of eBC measurements.

A statistically significant (p-value < 0.05) decreasing trend was observed for all of the stations included in the trend analysis~~, which had at least four years of data~~ (TR1, TR2, UB1, and RB2) as shown in Fig. 4 and in Table 3. The smallest absolute decrease was observed at the background stations UB1 and RB2, where the slopes of the trends were -0.02 and -0.01 μg m^{-3} yr^{-1}, respectively. At TR1 the concentration decreased more rapidly by -0.04 μg m^{-3} yr^{-1}~~,~~ and at TR2 ~~of~~ the decrease was even greater: -0.09 μg m^{-3} yr^{-1}. In addition to the absolute trend, we also determined the relative trends by dividing the absolute slope of the trend by the overall median concentration. At TR1, UB1, and RB2, the relative trends were rather similar: -6.4, -5.9, and -7.8 % yr^{-1}, respectively. At TR2, the decrease was relatively ~~faster~~steeper: -10.4 % yr^{-1}.

One possible cause for the decreased pollution concentrations could have been the changes in the meteorological parameters that affect the dilution. Teinilä et al. (2019) reported that in HMA the two most important meteorological parameter that affected the PM_1 concentrations were wind speed (WS) and temperature (T); Järvi et al. (2008) observed that of the meteorological parameters the WS and mixing height (MH) affected the BC concentration the most. The highest concentrations were observed at low WS and MH conditions and when the T was either very high in summer or very low in winter, which indicates stable and stagnant meteorological conditions. Also, a temperature decrease during colder periods could increase the emissions from residential wood combustion. Therefore, in addition to BC, we ran the trend analysis for the time series of WS, T, and MH (time series in Fig. S1). However, we did not observe statistically significant trends for any of these parameters. We also studied the trends for the different seasons separately to see, for example, if the temperatures had increased in the summer months or decreased in winter months, but this analysis did not yield statistically significant trends either. Therefore, it is likely that the decreasing trends of the eBC concentration were not be explained by the meteorological factors.

To see how the decrease in eBC concentrations compareds to the trends of other air pollutants, we conducted the trend analysis also for the $PM_{2.5}$ and NO_x data. The resulted trends are also presented in Table 3. The only parameter for which we did not observe a statistically significant decreasing trend was $PM_{2.5}$ at TR2 (p-value = 0.05). The relative trends varied from station to station, but a common trait was that the concentrations of eBC and NO_x decreased relatively faster than the concentration of $PM_{2.5}$. The trends of NO_x concentration varied from -19.7 % yr^{-1} (TR3) to -4.0 % yr^{-1} (RB2) and the trends of $PM_{2.5}$ concentration varied from -3.9 % yr^{-1} (UB1) to -2.7 % yr^{-1} (RB2). Since there was a notable decrease in the eBC concentration at TR3 between the years 2010 and 2015 (Table 1), the trend at TR3 was also studied, which is presented in Sect. S5. The analysis showed that the concentrations of eBC, NO_x, and $PM_{2.5}$ decreased about -12.2, -8.2, and -5.6 % yr^{-1}, respectively. So also at the TR3, the $PM_{2.5}$ concentration decreased relatively the slowest.

Compared to $PM_{2.5}$, tThe concentrations of eBC and NO_x weare more sensitive to the changes in the traffic, since they are more dependent on the local sources than $PM_{2.5}$ as discussed in the Sect. 3.2.1. Therefore, the bigger decrease in eBC and NO_x concentrations indicated that especially the emissions from traffic have decreased. Since $PM_{2.5}$ is not limited as sensitiveonly to changes in primary traffic related emissions, it explains why the slope of the trend for $PM_{2.5}$ was relatively smaller than that of eBC and NO_x. In other words, since the pollutant emissions from traffic sources have generally decreased, it clearly affecteds the trends of eBC and NO_x, which are originating from local traffic sources, but to a lesser extent the trend of $PM_{2.5}$. Even though the decrease in local traffic emissions was the probable explanation for the decreasing trends especially at the TR and UB sites, changes in the long-range transported pollution likely affected the trends as well. Statistically significant trends were observed also at RB2 indicating that the long-range transported pollution and the emissions in the regional area have also decreased.

For eBC and NO_x it is difficult to say which one of the pollutants decreased at faster rate. Their relative decreases were rather similar at TR1 and ~~at~~ UB1. At RB2, the decrease in eBC concentration was a more notable if compared to the decrease in NO_x concentration. However, at RB2, there were no local sources so the situation ~~i~~was probably rather complicated and depend~~s~~ed on the atmospheric chemistry and aging of the pollutants. At TR2, there was a large difference and NO_x seemed to decrease at double rate compared to eBC, which could be caused, for example, by the fast renewal of the city bus fleet. According to the Helsinki Regional Transport Authority (HSL), in 2015 17 % of the HSL buses were Euro VI or Euro VI energy efficient and in 2018 the fraction had increased to 48 %. A study by Järvinen et al. (2019) showed that moving to Euro VI buses from enhanced environmentally friendly vehicles (EEV) efficiently decreases the NO_x emissions. However, ~~it must be noted that here~~at TR2, the short time series cause uncertainty to the trends and for example, the year-to-year variability caused by the meteorological conditions could cause apparent decrease in pollutant concentrations.

The trends at TR1 and UB1 were investigated in more detail, since these stations had the longest time series and they were located closer to local sources in the HMA. To see if the traffic related emissions affected the trends, the trend analysis was conducted separately for each hour so that the monthly median was determined for each hour of the day. Only the data from weekdays was included in the analysis. This trend analysis revealed that there were clear decreases in the eBC and NO_x concentrations around the morning rush hour as shown in Figs. 5a and 5c. The trend of the eBC concentration had a distinctive diurnal cycle in general so that the decrease was more prominent during morning and day and actually there was no statistically significant trend during night time (22 p.m. – 3 a.m.). Similar diurnal pattern was observed for the $PM_{2.5}$ as well, however, the pattern was not as well defined as for the eBC. At TR1, the $PM_{2.5}$ data between 8 – 9 a.m. and 9 – 10 p.m. were not included in the trend analysis due to technical issues caused by a disturbance of the air conditioning. In principle, the diurnal trends~~is~~ indicate~~d~~s that the primary ~~eBC and NO_x~~ emissions from traffic ~~sources~~ have decreased most prominently, i.e., changes in traffic regulations and technological advancements have decreased especially the eBC and NO_x emissions. ~~Oppositely, the times in $PM_{2.5}$ peaks did not correspond directly to traffic hours at either of the stations.~~

According to the report about traffic in Helsinki, the traffic volume in Helsinki increased a bit, by 0.4 % yr^{-1} during the period 2006–2016 (Helsinki, 2017; in Finnish only)~~.~~. However, in general, the number of vehicles entering and exiting the city center (~~including~~ represented by TR1) decreased by 1.5 % yr^{-1} and the number of vehicles entering and exiting the central urban area (~~including~~ represented by TR2, TR3, and UB1) decreased by 0.8 % yr^{-1} during the same period, which obviously decreases the emissions from traffic. The decreases in traffic rates were especially observed in the North-West part of the central urban area (e.g., Fig. S10c), which probably affected the trends observed at TR3. However, at Mäkelänkatu, which is the main street next to TR2 where decreasing trends were also observed, there was no clear decrease in the traffic rate (Fig. S10f).

Since the traffic rates did not decrease at all of the stations, it can be concluded that the decreasing trends were due to the renewal of vehicle and bus fleet as well as cleaner renewable fuels, which have been shown to decrease both BC and NO_x

emissions (Järvinen et al., 2019; Pirjola et al., 2019; Timonen et al., 2017). The exhaust particle number of d~~emission reduction of traffic, taken place mostly due to the efficient regulation of d~~iesel and gasoline vehicles~~car exhaust particle number~~ have been regulated efficiently in order to reduce the traffic emissions. The~~is~~ regulations have~~s~~ enforced the use of diesel particulate filters (DPFs) in new diesel passenger cars and heavy-duty diesel vehicles, reducing their BC and PM emissions even more than 90 % up to 99 % if compared to the diesel vehicles without DPF (Bergmann et al., 2009; Preble et al., 2015). A~~However, a~~lso, the increased use of biofuels and gas as vehicle fuels, increased share of electric vehicles, and ~~as well as~~ improvements of fuel economy in internal combustion engines affected the observed trends. E.g., the increase of fuel injection pressure in diesel combustion can improve the fuel economy of engines and simultaneously decrease the BC emissions of engines (Lähde et al., 2011). ~~The renewal of bus and vehicle fleet as well as cleaner renewable fuels have been shown to decrease both BC and NO_x emissions and this would be expected to be seen in the ambient eBC concentrations in traffic environment.~~

Decreases in the eBC concentration (or absorption coefficient) have also been observed in the Finnish arctic (Dutkiewicz et al., 2014; Lihavainen et al., 2015). In general declining trends in atmospheric aerosol particle number concentration and particulate material has been observed in various different types of environments in Europe (Asmi et al., 2013). Similar results for the trend of eBC concentration have been reported at several cities and countries in Europe. In Stockholm, Sweden, Krecl et al. (2017), reported about 60 % reduction in eBC concentration in a busy street canyon between the years 2006 and 2013 (i.e., -7.5 % yr^{-1}). Singh et al. (2018) observed a statistically significant decreasing trend in eBC concentration at five measurements stations, which operated during 2009–2016 and were located in different types of environments in the United Kingdom. The trends varied from -0.09 µg m^{-3} yr^{-1} (-4.7 % yr^{-1}) at an ~~urban background~~UB site to -0.80 µg m^{-3} yr^{-1} (-8.0 % yr^{-1}) at a ~~curbside~~ TR station. A trend study based only in London reported on average -0.59 µg m^{-3} yr^{-1} (-11 % yr^{-1}) decrease in eBC concentration at three ~~roadside~~ TR sites for the period 2010–2014 (Font and Fuller, 2016). Kutzner et al. (2018) observed statistically significant decreasing BC (both eBC and elemental carbon) trends for the period of 2005–2014 at several sites in Germany. The trends at ~~traffic~~ TR sites varied from -0.31 to -0.15 µg m^{-3} yr^{-1} and the trends at ~~urban background~~UB sites varied from -0.03 to -0.02 µg m^{-3} yr^{-1}. Also, a more recent study by Sun et al. (2020) reported decreasing relative trends of eBC in Germany for the period of 2009–2018: from -0.19 to -0.08 µg m^{-3} yr^{-1} (-11.3 to -5.0 % yr^{-1}), from -0.08 to -0.03 µg m^{-3} yr^{-1} (-8.1 to -2.3 % yr^{-1}), and from -0.03 to 0.00 µg m^{-3} yr^{-1} (-7.8 to -3.2 % yr^{-1}) at TR, UB, and RB sites, respectively.

These studies reported higher absolute trends compared to the absolute trends observed at our study, which wa~~i~~s probably due to ~~higher~~ lower eBC concentrations in southern Finland~~at these sites~~. However, the relative trends were rather similar. Similarly to this study, ~~O~~other studies also observed steeper absolute trends at TR and UB~~urban~~ sites ~~compared to~~than at RB~~background~~ sites. Sun et al. (2020) observed similar diurnal eBC trends at TR and UB site as we did so that the decreasing trends were the highest during the morning rush hour. In addition to eBC, Krecl et al. (2017) studied also the trend of NO_x, and Font and Fuller (2016) studied the trend of $PM_{2.5}$. Contradictory to our study, Krecl et al. (2017) did not observe a decreasing trend for NO_x

and. Font and Fuller (2016) reported decreasing trends for $PM_{2.5}$ concentration, which were relatively similar to the trends of eBC concentration.

The abovementioned studies, which were conducted in urban environments, also suggested that the dominant reason for thelinked the decreased in BC concentrations were to traffic regulations. For example: Krecl et al. (2017) drew a connection between the decreasing eBC concentration and the renewal of the vehicle fleet.; Sun et al. (2020), Singh et al. (2018), and Kutzner et al. (2018) proposed that the reductions in eBC were linked to the local and national air quality policies;. and Font and Fuller (2016) suspected that the eBC concentration decreased due to effective filters in diesel vehicles.

3.32.2 Seasonal, weekly and diurnal variation of BC

The diurnal variation of eBC was investigated separately for the cold and the warm seasons. According to Fig. S1, the coldest 5 months typically extended from November to March and the warmest 5 months from May to September. April and October were omitted from this analysis as transition months. The seasonal dependencies for each station separately are presented in Figs. S11 and S12.

The seasonal and diurnal variations of eBC were rather similar between the stations that belong in the same category (Fig. S13). Instead of studying the variation at each station separately, we determined a mean diurnal variation for different station categories to study the variation more generally. The figures were plotted by calculating the mean concentration each hour of each day of the week for the cold and the warm seasons separately. All the available data were taken into account when the diurnal variation from different stations were averaged together.

The mean seasonal, weekly and diurnal variation of eBC for different station categories are presented in Fig. 6. At the TR1, TR2, TR3, and TR4 (i.e., TR1–4) the seasonal and diurnal variation was different from TR5 and TR6 (i.e., TR5–6) and therefore the mean diurnal variations for these sets of stations were plotted separately in Figs. 6a and 6b. Fig. 6c presents the mean diurnal variation averaged over all the DH sites. Since UB1 had a notably longer time series compared to UB2, these two time series were not combined and only the diurnal variation from UB1 is used in Fig. 6d. The diurnal variation at RB1 was also presented without combining it with RB2 data in Fig. 6e.

Fig. 6 shows two common traits observed at all of the TR, DH, and UB sites during both seasons: 1) eBC concentration peak appeared each weekday morning around 8 a.m. because of the morning rush hour; and 2) the lowest eBC concentrations were measured each day during the night around 3 a.m. when there were not much anthropogenic activities. In addition to these common trends, each station category had their own traits in the seasonal, weekly, and diurnal variation. We also did a similar

variation analysis for the eBC/$PM_{2.5}$ ratio presented in Fig. S14. In general the eBC/$PM_{2.5}$ ratio seemeds to follow the variation of eBC (i.e., the eBC varies relatively more than $PM_{2.5}$).

At the TR1–4, the seasonal variation of eBC was not strong and in Fig. 6a the lines for the cold and the warm season follow each other. The lack of seasonal variation in traffic environment was also observed previously in the HMA (Helin et al., 2018; Teinilä et al., 2019) and elsewhere (Kutzner et al., 2018; Reche et al., 2011). During weekdays, the morning concentration peak of eBC occurred around 8 a.m. and the afternoon peak around 4 p.m. The variation of eBC concentration correlated with the diurnal variation of the traffic counts (see examples of traffic rates from Mannerheimintie and Mäkelänkatu in Fig. S10). eBC concentration was notably lower during weekend, when the traffic rates were also lower. Järvi et al. (2008) also reported that the close by traffic rates are the most important factors explaining the variation in eBC concentration at a TR site.

During the warm season at TR1–4 sites, the morning eBC concentration peak was notably higher than during the cold season. Since seasonal variation in the traffic rates were not expected, the observation iwas probably explained by the variation in WS and MH. When the MH and WS are higherWS, the concentrations of air pollutants decrease is one of the most important meteorological parameter that affects the air pollution concentrations so that the concentrations decrease with increasing WS due to more effective dilution (Järvi et al., 2008; Teinilä et al., 2019). A study by Teinilä et al. (2019) showed that the diurnal variation of WS depended on the season: in summer, Fig. S2 shows that the MH and WS in the morning (before 9 a.m.) during the cool season were lower than in the warm season. WS and MH had a clear diurnal cycle in the warm season, but had its minimum around 6 a.m. and it reached its maximum around 3 p.m. with the growing boundary layer; in winter the cool season the WS or MH had no variation whatsoever. Similar diurnal variation in WS was seen in our data (Fig. S2). Since theThe lower WSMH and WS in the morning during the warm season is typically lower during the warm season than during the cold season, the differences in the WS and the dilution could explain why the eBC concentrations peak during the warm season mornings.

The seasonal variation of eBC was the most pronounced at the DH sites, where the lowest concentrations occurred in June and July and the highest concentrations in December and January (Figs. S11gS12g–k), which is probably likely explained by the residential wood combustion during the cold season. At the DH sites, the highest concentrations were measured during the evening (Fig. 6c), when people returned back home and started to warm up their houses and saunas. after the workday. This was different to other station categories, where the morning concentration peak was higher or similar compared to the afternoon peak. Domestic wood combustion also increased the concentrations during the weekend and unlike at the TR and UB sites, the eBC concentration at the DH sites were rather similar compared to the weekdays.

The effect of wood combustion at DH sites was studied by Helin et al. (2018), who applied AE33 data measured at TR2, DH3, and DH4 (note different names of the sites between our study and the study by Helin et al., 2018) in a source apportionment model suggested by Sandradewi et al. (2008). They reported that on average about 41 and 46 % of the eBC observed at the

DH4 and DH5, respectively, originated from wood combustion. The fractions were notably higher than observed at the TR2 (about 15 %). They also observed higher eBC fractions from wood combustion in the cool season: for example, eBC fractions from wood combustion were 46 and 35 % at DH3 in winter and summer, respectively. The effect of wood combustion in evenings was also evident in the data by Helin et al. (2018), who observed that eBC from wood combustion increased towards the evening at the DH3 and DH4. A comparison between weekdays and weekends at DH3 showed similar eBC concentrations originating from traffic, but slightly increased eBC concentrations from wood combustion in the weekend.

The effect of residential wood combustion on the diurnal variation was also observed at TR5–6 (Fig. 6b), which were closer to the detached housing areas than the other TR sites (see Fig. 1). The temporal variation at TR5–6 was a mix between the variation observed at the other traffic sites (TR1–4) and at the DH sites. At TR5–6, the concentration of eBC was notably higher in the evenings during the cold season compared to the warm season, which is similar to the DH stations. ~~Also~~In addition, the maximum of the afternoon concentration peak occurred around the same time as at the DH sites. However, the effect of traffic is seen in the afternoon peaks, since the peaks grew more rapidly around the afternoon rush hour. The traffic also affected the morning concentration peak, which is similar to the afternoon concentration peak, whereas at the DH sites, the morning peak was notably lower. At TR5–6, the eBC concentration during weekend was lower than in weekdays, which was observed at the other TR sites as well. However, the difference was not as pronounced, which is again due to the effect of wood combustion (e.g., increased eBC concentration during Saturday evening).

In the cold season, the diurnal variation of eBC at the UB1 was rather similar to the diurnal variation at the TR1–4, with the rush hour peaks occurring in the morning and afternoon (Fig. 6d). During the warm season, however, the afternoon rush hour peak was missing. Also in weekends, the concentrations during the warm season were considerably lower. Since the UB1 was not in vicinity to pollution sources, the effect of dilution, which is governed by meteorological parameters, ~~the meteorological parameters~~ became more important. In the daytime during the warm season, the pollutants were diluted in the convective and more windy (Fig. S2) boundary layer ~~more~~ effectively, whereas during the cold season the pollutants accumulated in the boundary layer, if it is shallower and does not grow ~~-~~during the day as much as in the warm season (Pohjola et al., 2004). Also, the transported wood combustion emissions from the DH areas can slightly increase the concentrations during the cool season even closer to the city center (Helin et al., 2018).

At the RB sites, there was a notable seasonal variation as the eBC concentrations are higher during the cold season (Fig. S13n–o), since regional ~~and long-range transported~~ wood combustion emissions and long-range transported pollutants also elevates eBC concentrations at background sites in winter (Luoma et al., 2019). The diurnal and weekly variation, however, were not as clear (e.g., Fig. 6e) as there were no anthropogenic activities nearby. Diurnal variation was mainly caused by the variation in the convective boundary layer height that caused mixing and dilution and not by local anthropogenic sources, which is expected for a regional background station.

It is important to note that the measured variation in the ~~concentration of~~ eBC concentration wa~~i~~s affected by both: the BC emissions, and the atmospheric processes determining the dilution of the BC. For instance, the diurnal variation of the eBC concentration at a traffic site is affected by traffic rate at the nearby streets, roads and highways, but it is affected also by changes of local weather conditions possibly having a diurnal variation. Such atmospheric parameters are e.g., ambient temperature, prevailing wind direction, wind speed, and height of the boundary layer. Furthermore, especially the ambient temperature can directly affect the emissions also; e.g., slightly higher wintertime morning concentrations in detached housing areas can be due to the effects of ambient temperature to the cold start emissions of passenger cars.

Conclusions

This study analyzed the time series of atmospheric eBC concentrations in traffic, detached house, urban background and regional background environments measured in years 2007–~~–~~2018 in the Helsinki metropolitan area (HMA) and at one background site outside the HMA. The overall mean eBC concentration varied in the range ~~of~~ from 0.77 –~~–~~to 2.08 μg m^{-3} at TR stations, from 0.64 –~~–~~to 0.80 μg m^{-3} at DH stations, and from 0.51 –~~–~~to 0.53 μg m^{-3} at UB stations. At the RB sites the mean eBC concentrations were the lowest, about 0.28 μg m^{-3}.

The study highlights~~s~~ the importance of local sources in respect of urban air quality; it shows that the local traffic and ~~local~~ residential wood combustion~~burning~~ in residential areas formed~~s~~ the main sources of atmospheric eBC in the urban area studied here. The influence of traffic was clearly seen at the traffic stations (TR) and the effect of domestic wood combustion at the detached housing sites (DH). At the TR sites, which located closer to residential areas (TR5–~~–~~6), the diurnal variation showed signs that the air quality in the area was heavily affected both by traffic and domestic wood burning emissions. The concentrations at the TR sites reached their peak in the weekday mornings and stayed elevated until the afternoon rush hour had~~s~~ passed. In the evening, after the workday, the eBC concentrations reached their maxima at the DH sites, where the residents ~~started to~~ warmed up their houses partly by~~with~~ wood combustion. At the DH sites, the eBC concentrations did not decrease during the weekend like at the ~~concentration decreased at the~~ TR sites.

The trend analysis conducted in this study showed ~~very~~ statistically significant~~clearly~~ decreasing trends for atmospheric BC. Decreasing eBC concentration has two positive effects: 1) improved air quality and 2) decreased warming effect on the global climate by light absorbing aerosols. The absolute trends of eBC concentration were most notable at the ~~traffic~~ TR sites, where the trends varied from -0.04 to -0.09 μg m^{-3} yr^{-1}. At an UB station and at an RB station, the absolute trends were -0.02 and -0.01 μg m^{-3} yr^{-1}, respectively. The relative trends of eBC concentration varied ~~between~~ from -10.6 –~~–~~to -5.7 % yr^{-1}, which was

rather similar to the relative trends of NO_x concentration, which varied ~~between~~ from -19.7 ~~—~~to -4.0 % yr^{-1}. ~~However, t~~The relative trends of $PM_{2.5}$, which varied from -3.9 to -2.7 % yr^{-1}, did not decrease as rapidly as the trends of eBC and NO_x ~~were did not decrease as rapidly as for eBC and NO_x and the relative trends of $PM_{2.5}$ varied between -3.9 — -2.7 % yr^{-1}~~. For the eBC and NO_x the most notable decrease was observed for the hours of morning rush hour, when traffic has the biggest effect on the air quality. The difference between the relative trends of eBC, NO_x and $PM_{2.5}$ concentrations, and the most notable decrease of eBC and NO_x concentrations during the morning rush hour indicated that especially the emissions from traffic have decreased.

~~This study suggest that the development in vehicle exhaust particle mitigation has been successful, at least from the viewpoint of BC and NO_x emissions.~~This study suggests that the development in vehicle exhaust particle mitigation has been successful, at least from the viewpoint of BC and NO_x emissions. With the current development, the pollution concentrations are expected to decrease in the next years as well. In general, the vehicle fleet is renewing, electric and hybrid cars are gaining popularity, and vehicles that run with biofuels or gas are becoming more common. The operator of public traffic in the HMA (HSL) aims to cut more than 90 % of their bus emissions (NO_x, PM, CO_2) by the year 2025 compared to the year 2010, which will improve the air quality especially at the main roads in the HMA, where several bus lines operate.

~~Simultaneously, this~~This study ~~clearly~~ also shows the need for regulation and mitigation of emissions from residential wood combustion, which is, according to the emission inventories, actually the most significant BC source in Finland (Rautalahti and Kupiainen, 2016). The research material of this study did not allow the assessment of long-term trends in DH areas but, in principle, any similar technology changes have not been observed in wood combustion than has been in vehicles.

Acknowledgements This work was supported by the Academy of Finland Centre of Excellence in Atmospheric Science (grant no. 307331) and NANOBIOMASS (207537), ACTRIS-Finland (329274), the Regional innovations and experimentations funds (project HAQT, AIKO014) and Business Finland (BC Footprint -project 528/31/2019, and MegaSense Smart City-project 6884/31/2018). In addition, the work was financially supported by European Commission through ACTRIS2 (654109) and ACTRIS-IMP (871115) and through SMart URBan Solutions for air quality, disasters and city growth (689443), ERA-NET-Cofund and by University of Helsinki (ACTRIS-HY).

Data availability All the data presented in this study is open access. The air quality data collected by the HSY is available from their website (https://www.hsy.fi/fi/asiantuntijalle/avoindata/Sivut/default.aspx, last access: 27 February 2020). The data collected from the SMEAR sites (UB2 and RB2) has been accessed by the Smart-SMEAR online tool (Junninen et al., 2009).

Author contribution Krista Luoma did the data analysis and wrote the manuscript. All the other authors helped with analysing the results and they ~~also~~ reviewed and commented the manuscript.

References

Abhijith, K. V., Kumar, P., Gallagher, J., McNabola, A., Baldauf, R., Pilla, F., Broderick, B., Di Sabatino, S., and Pulvirenti, B.: Air pollution abatement performances of green infrastructure in open road and built-up street canyon environments–A review, Atmos. Environ., 162, 71-86, 10.1016/j.atmosenv.2017.05.014, 2017.

Andreae, M. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 10.5194/acp-6-3131-2006, 2006.

Asmi, A., Collaud Coen, M., Ogren, J., Andrews, E., Sheridan, P., Jefferson, A., Weingartner, E., Baltensperger, U., Bukowiecki, N., Lihavainen, H., Kivekäs, N., Asmi, E., Aalto, P. P., Kulmala, M., Wiedensohler, A., Birmili, W., Hamed, A., O'Dowd, C., Jennings, S., Weller, R., Flentje, H., Fjaeraa, A. M., Fiebig, M., Myhre, C. L., Hallar, A. G., Swietlicki, E., Kristensson, A., and Laj, P.: Aerosol decadal trends–Part 2: In-situ aerosol particle number concentrations at GAW and ACTRIS stations, Atmos. Chem. Phys., 13, 895-916, 10.5194/acp-13-895-2013, 2013.

Aurela, M., Saarikoski, S., Niemi, J. V., Canonaco, F., Prevot, A. S. H., Frey, A., Carbone, S., Kousa, A., and Hillamo, R.: Chemical and source characterization of submicron particles at residential and traffic sites in the Helsinki metropolitan area, Finland, Aerosol Air Qual. Res., 15, 1213-1226, 10.4209/aaqr.2014.11.0279, 2015.

Becerril-Valle, M., Coz, E., Prévôt, A. S. H., Močnik, G., Pandis, S. N., de la Campa, A. M. S., Alastuey, A., Díaz, E., Pérez, R. M., and Artíñano, B.: Characterization of atmospheric black carbon and co-pollutants in urban and rural areas of Spain, Atmos. Environ., 169, 36-53, 10.1016/j.atmosenv.2017.09.014, 2017.

Bergmann, M., Kirchner, U., Vogt, R., and Benter, T.: On-road and laboratory investigation of low-level PM emissions of a modern diesel particulate filter equipped diesel passenger car, Atmos. Environ., 43, 1908-1916, 10.1016/j.atmosenv.2008.12.039, 2009.

Brantley, H. L., Hagler, G. S., Deshmukh, P. J., and Baldauf, R. W.: Field assessment of the effects of roadside vegetation on near-road black carbon and particulate matter, Sci. Total Environ., 468, 120-129, 10.1016/j.scitotenv.2013.08.001, 2014.

Clougherty, J. E., Kheirbek, I., Eisl, H. M., Ross, Z., Pezeshki, G., Gorczynski, J. E., Johnson, S., Markowitz, S., Kass, D., and Matte, T.: Intra-urban spatial variability in wintertime street-level concentrations of multiple combustion-related air pollutants: the New York City Community Air Survey (NYCCAS), J. Expo. Sci. Env. Epid., 23, 232-240, 10.1038/jes.2012.125, 2013.

Collaud Coen, M., Weingartner, E., Nyeki, S., Cozic, J., Henning, S., Verheggen, B., Gehrig, R., and Baltensperger, U.: Long-term trend analysis of aerosol variables at the high-alpine site Jungfraujoch, J. Geophys. Res. Atmos., 112, 10.1029/2006JD007995, 2007.

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J., Jennings, S. G., Moerman, M., Petzold, A., Schmid, O., and Baltensperger, U.: Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms, Atmos. Meas. Tech., 3, 457-474, 10.5194/amt-3-457-2010, 2010.

Collaud Coen, M., Andrews, E., Asmi, A., Baltensperger, U., Bukowiecki, N., Day, D., Fiebig, M., Fjæraa, A. M., Flentje, H., Hyvärinen, A., Jefferson, A., Jennings, S., Kouvarakis, G., Lihavainen, H., Lund Myhre, C., Malm, W., Mihalopoulos, N., Molenar, J., O'Dowd, C., Ogren, J. A., Schichtel, B., Sheridan, P., Virkkula, A., Weingartner, E., Weller, R., and Laj, P.: Aerosol decadal trends–Part 1: In-situ optical measurements at GAW and IMPROVE stations, Atmos. Chem. Phys., 13, 869-894, 10.5194/acp-13-869-2013 2013.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiala, Finland, Boreal Environ. Res., 10, 323-336, 2005.

Dos Santos-Juusela, V., Petäjä, T., Kousa, A., and Hämeri, K.: Spatial–temporal variations of particle number concentrations between a busy street and the urban background, Atmos. Environ., 79, 324-333, 10.1016/j.atmosenv.2013.05.077, 2013.

Drinovec, L., Močnik, G., Zotter, P., Prévôt, A., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T., Wiedensohler, A., and Hansen, A. D. A.: The "dual-spot" Aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation, Atmos. Meas. Tech., 8, 1965-1979, 10.5194/amt-8-1965-2015, 2015.

Dutkiewicz, V. A., DeJulio, A. M., Ahmed, T., Laing, J., Hopke, P. K., Skeie, R. B., Viisanen, Y., Paatero, J., and Husain, L.: Forty-seven years of weekly atmospheric black carbon measurements in the Finnish Arctic: Decrease in black carbon with declining emissions, J. Geophys. Res. Atmos., 119, 7667-7683, 10.1002/2014JD021790, 2014.

Enroth, J., Saarikoski, S., Niemi, J., Kousa, A., Ježek, I., Močnik, G., Carbone, S., Kuuluvainen, H., Rönkkö, T., Hillamo, R., and Pirjola, L.: Chemical and physical characterization of traffic particles in four different highway environments in the Helsinki metropolitan area, Atmos. Chem. Phys., 16, 5497-5512, 10.5194/acp-16-5497-2016, 2016.

Font, A. and Fuller, G. W.: Did policies to abate atmospheric emissions from traffic have a positive effect in London?, Environ. Pollut., 218, 463-474, 10.1016/j.envpol.2016.07.026, 2016.

Gilbert, R. O.: Statistical methods for environmental pollution monitoring, John Wiley & Sons, New York, 1987.

Hari, P. and Kulmala, M.: Station for measuring ecosystem–atmosphere relations, Boreal Environ. Res., 10, 315-322, 2005.

Helin, A., Niemi, J. V., Virkkula, A., Pirjola, L., Teinilä, K., Backman, J., Aurela, M., Saarikoski, S., Rönkkö, T., Asmi, E., and Timonen, H.: Characteristics and source apportionment of black carbon in the Helsinki metropolitan area, Finland, Atmos. Environ., 190, 87-98, 10.1016/j.atmosenv.2018.07.022, 2018.

Hellén, H., Kangas, L., Kousa, A., Vestenius, M., Teinilä, K., Karppinen, A., Kukkonen, J., and Niemi, J. V.: Evaluation of the impact of wood combustion on benzo [a] pyrene (BaP) concentrations; ambient measurements and dispersion modeling in Helsinki, Finland, Atmos. Chem. Phys., 17, 3475-3487, 10.5194/acp-17-3475-2017, 2017.

Helsinki: Liikenteen kehitys Helsingissä 2016, Helsingin kaupunki, kaupunkiympäristön toimiala, 2017.

Hienola, A. I., Pietikäinen, J.-P., Jacob, D., Pozdun, R., Petäjä, T., Hyvärinen, A.-P., Sogacheva, L., Kerminen, V.-M., Kulmala, M., and Laaksonen, A.: Black carbon concentration and deposition estimations in Finland by the regional aerosol-climate model REMO-HAM, Atmos. Chem. Phys., 13, 4033-4055, 10.5194/acp-13-4033-2013, 2013.

HSY: Tulisijojen käyttö ja päästöt pääkaupunkiseudulla vuonna 2014 (The use of fireplaces and the emissions from small-scale combustion in the Helsinki metropolitan Area in 2014; in Finnish), Helsinki Region Environmental Services Authority, Helsinki, 2016.

Hyvärinen, A.-P., Kolmonen, P., Kerminen, V.-M., Virkkula, A., Leskinen, A., Komppula, M., Hatakka, J., Burkhart, J., Stohl, A., Aalto, P., Kulmala, M., Lehtinen, K. E. J., Viisanen, Y., and Lihavainen, H.: Aerosol black carbon at five background measurement sites over Finland, a gateway to the Arctic, Atmos. Environ., 45, 4042-4050, 10.1016/j.atmosenv.2011.04.026, 2011.

Imhof, D., Weingartner, E., Ordóñez, C., Gehrig, R., Hill, M., Buchmann, B., and Baltensperger, U.: Real-world emission factors of fine and ultrafine aerosol particles for different traffic situations in Switzerland, Environ. Sci. Technol., 39, 8341-8350, 10.1021/es048925s, 2005.

Janssen, N. A. H., Hoek, G., Simic-Lawson, M., Fischer, P., Van Bree, L., Ten Brink, H., Keuken, M., Atkinson, R. W., Anderson, H. R., Brunekreef, B., and Cassee, F. R.: Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM10 and PM2. 5, Environ. Health Perspect., 119, 1691-1699, 10.1289/ehp.1003369, 2011.

Junninen, H., Lauri, A., Keronen, P., Aalto, P., Hiltunen, V., Hari, P., and Kulmala, M.: Smart-SMEAR: on-line data exploration and visualization tool for SMEAR stations, Boreal Environ. Res., 14, 447-457, 2009.

Järvi, L., Junninen, H., Karppinen, A., Hillamo, R., Virkkula, A., Mäkelä, T., Pakkanen, T., and Kulmala, M.: Temporal variations in black carbon concentrations with different time scales in Helsinki during 1996–2005, Atmos. Chem. Phys., 8, 1017-1027, 10.5194/acp-8-1017-2008, 2008.

Järvi, L., Hannuniemi, H., Hussein, T., Junninen, H., Aalto, P. P., Hillamo, R., Mäkelä, T., Keronen, P., Siivola, E., Vesala, T., and Kulmala, M.: The urban measurement station SMEAR III: Continuous monitoring of air pollution and surface–atmosphere interactions in Helsinki, Finland, Boreal Environ. Res., 14, 86-109, 2009.

Järvinen, A., Timonen, H., Karjalainen, P., Bloss, M., Simonen, P., Saarikoski, S., Kuuluvainen, H., Kalliokoski, J., Dal Maso, M., Niemi, J. V., and Rönkkö, T.: Particle emissions of Euro VI, EEV and retrofitted EEV city buses in real traffic, Environ. Pollut., 250, 708-716, 10.1016/j.envpol.2019.04.033, 2019.

Karppinen, A., Joffre, S. M., and Kukkonen, J.: The refinement of a meteorological pre-processor for the urban environment, International Journal of Environment Pollution, 14, 565-572, 10.1504/IJEP.2000.000580, 2000.

Kirchstetter, T. W., Novakov, T., and Hobbs, P. V.: Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon, J. Geophys. Res. Atmos., 109, 10.1029/2004JD004999, 2004.

Krecl, P., Johansson, C., Targino, A. C., Ström, J., and Burman, L.: Trends in black carbon and size-resolved particle number concentrations and vehicle emission factors under real-world conditions, Atmos. Environ., 165, 155-168, 10.1016/j.atmosenv.2017.06.036, 2017.

Krzyżanowski, M., Kuna-Dibbert, B., and Schneider, J.: Health effects of transport-related air pollution, WHO Regional Office Europe, 2005.

Kutzner, R. D., von Schneidemesser, E., Kuik, F., Quedenau, J., Weatherhead, E. C., and Schmale, J.: Long-term monitoring of black carbon across Germany, Atmos. Environ., 185, 41-52, 10.1016/j.atmosenv.2018.04.039, 2018.

Lack, D. and Cappa, C.: Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, Atmos. Chem. Phys., 10, 4207-4220, 10.5194/acp-10-4207-2010, 2010.

Lefebvre, W., Fierens, F., Trimpeneers, E., Janssen, S., Van de Vel, K., Deutsch, F., Viaene, P., Vankerkom, J., Dumont, G., Vanpoucke, C., Mensink, C., Peelaerts, W., and Vliegen, J.: Modeling the effects of a speed limit reduction on traffic-related elemental carbon (EC) concentrations and population exposure to EC, Atmos. Environ., 45, 197-207, 10.1016/j.atmosenv.2010.09.026, 2011.

Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525, 367, 10.1038/nature15371, 2015.

Li, J., Carlson, B. E., Dubovik, O., and Lacis, A. A.: Recent trends in aerosol optical properties derived from AERONET measurements, Atmos. Chem. Phys., 14, 12271-12289, 10.5194/acp-14-12271-2014, 2014.

Lihavainen, H., Hyvärinen, A., Asmi, E., Hatakka, J., and Viisanen, Y.: Long-term variability of aerosol optical properties in northern Finland, Boreal Environ. Res., 20, 526-541, 2015.

Luoma, K., Virkkula, A., Aalto, P., Petäjä, T., and Kulmala, M.: Over a 10-year record of aerosol optical properties at SMEAR II, Atmos. Chem. Phys., 19, 11363-11382, 10.5194/acp-19-11363-2019, 2019.

Lähde, T., Rönkkö, T., Happonen, M., Söderström, C., Virtanen, A., Solla, A., Kytö, M., Rothe, D., and Keskinen, J.: Effect of fuel injection pressure on a heavy-duty diesel engine nonvolatile particle emission, Environ. Sci. Technol., 45, 2504-2509, 10.1021/es103431p, 2011.

Massoli, P., Fortner, E. C., Canagaratna, M. R., Williams, L. R., Zhang, Q., Sun, Y., Schwab, J. J., Trimborn, A., Onasch, T. B., Demerjian, K. L., Charles, E., Worsnop, D., and Jayne, J.: Pollution gradients and chemical characterization of particulate matter from vehicular traffic near major roadways: Results from the 2009 Queens College Air Quality Study in NYC, Aerosol Sci. Technol., 46, 1201-1218, 10.1080/02786826.2012.701784, 2012.

Pakkanen, T. A., Kerminen, V.-M., Ojanen, C. H., Hillamo, R. E., Aarnio, P., and Koskentalo, T.: Atmospheric black carbon in Helsinki, Atmos. Environ., 34, 1497-1506, 10.1016/S1352-2310(99)00344-1, 2000.

Petzold, A. and Schönlinner, M.: Multi-angle absorption photometry—a new method for the measurement of aerosol light absorption and atmospheric black carbon, J. Aerosol Sci, 35, 421-441, 10.1016/j.jaerosci.2003.09.005, 2004.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S.-M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X.-Y.: Recommendations for reporting "black carbon" measurements, Atmos. Chem. Phys., 13, 8365-8379, 10.5194/acp-13-8365-2013, 2013.

Pirjola, L., Lähde, T., Niemi, J., Kousa, A., Rönkkö, T., Karjalainen, P., Keskinen, J., Frey, A., and Hillamo, R.: Spatial and temporal characterization of traffic emissions in urban microenvironments with a mobile laboratory, Atmos. Environ., 63, 156-167, 10.1016/j.atmosenv.2012.09.022, 2012.

Pirjola, L., Niemi, J. V., Saarikoski, S., Aurela, M., Enroth, J., Carbone, S., Saarnio, K., Kuuluvainen, H., Kousa, A., Rönkkö, T., and Hillamo, R.: Physical and chemical characterization of urban winter-time aerosols by mobile measurements in Helsinki, Finland, Atmos. Environ., 158, 60-75, 10.1016/j.atmosenv.2017.03.028, 2017.

Pirjola, L., Kuuluvainen, H., Timonen, H., Saarikoski, S., Teinilä, K., Salo, L., Datta, A., Simonen, P., Karjalainen, P., Kulmala, K., and Rönkkö, T.: Potential of renewable fuel to reduce diesel exhaust particle emissions, Appl. Energ., 254, 113636, 10.1016/j.apenergy.2019.113636, 2019.

Pohjola, M. A., Rantamäki, M., Kukkonen, J., Karppinen, A., and Berge, E.: Meteorological evaluation of a severe air pollution episode in Helsinki on 27-29 December 1995, Boreal Environ. Res., 9, 75-87, 2004.

Preble, C. V., Dallmann, T. R., Kreisberg, N. M., Hering, S. V., Harley, R. A., and Kirchstetter, T. W.: Effects of particle filters and selective catalytic reduction on heavy-duty diesel drayage truck emissions at the Port of Oakland, Environ. Sci. Technol., 49, 8864-8871, 10.1021/acs.est.5b01117, 2015.

Rautalahti, E. and Kupiainen, K.: Emissions of black carbon and methane in Finland, Ministry of the Environment, Helsinki, 2016.

Reche, C., Querol, X., Alastuey, A., Viana, M., Pey, J., Moreno, T., Rodríguez González, S., González Ramos, Y., Fernández-Camacho, R., de la Rosa, J., Dall'Osto, M., Prévôt, A. S. H., Hueglin, C., Harrison, R. M., and Quincey, P.: New considerations for PM, black carbon and particle number concentration for air quality monitoring across different European cities, Atmos. Chem. Phys., 11, 6207–6227, 10.5194/acp-11-6207-2011, 2011.

Rönkkö, T. and Timonen, H.: Overview of sources and characteristics of nanoparticles in urban traffic-influenced areas, J. Alzheimer's Dis., 72, 15-28, 10.3233/JAD-190170, 2019.

Sandradewi, J., Prévôt, A. S., Szidat, S., Perron, N., Alfarra, M. R., Lanz, V. A., Weingartner, E., and Baltensperger, U.: Using aerosol light absorption measurements for the quantitative determination of wood burning and traffic emission contributions to particulate matter, Environ. Sci. Technol., 42, 3316-3323, 10.1021/es702253m, 2008.

Savolahti, M., Karvosenoja, N., Tissari, J., Kupiainen, K., Sippula, O., and Jokiniemi, J.: Black carbon and fine particle emissions in Finnish residential wood combustion: Emission projections, reduction measures and the impact of combustion practices, Atmos. Environ., 140, 495-505, 10.1016/j.atmosenv.2016.06.023, 2016.

Schwarz, J. P., Gao, R., Spackman, J., Watts, L., Thomson, D., Fahey, D., Ryerson, T., Peischl, J., Holloway, J., Trainer, M., Frost, G., T, B., Lack, D., de Gouw, J., Warneke, C., and Del Negro, L.: Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions, Geophys. Res. Lett., 35, 10.1029/2008GL033968, 2008.

Singh, V., Ravindra, K., Sahu, L., and Sokhi, R.: Trends of atmospheric black carbon concentration over the United Kingdom, Atmos. Environ., 178, 148-157, 10.1016/j.atmosenv.2018.01.030, 2018.

Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M.: Climate change 2013: The physical science basis, Cambridge University Press, 2013.

Sun, J., Birmili, W., Hermann, M., Tuch, T., Weinhold, K., Spindler, G., Schladitz, A., Bastian, S., Löschau, G., Cyrys, J., Gu, J., Flentje, H., Briel, B., Asbach, C., Kaminski, H., Ries, L., Sohmer, R., Gerwig, H., Wirtz, K., Meinhardt, F., Schwerin, A., Bath, O., Ma, N., and Wiedensohler, A.: Variability of black carbon mass concentrations, sub-micrometer particle number concentrations and size distributions: results of the German Ultrafine Aerosol Network ranging from city street to High Alpine locations, Atmos. Environ., 202, 256-268, 10.1016/j.atmosenv.2018.12.029, 2019.

Sun, J., Birmili, W., Hermann, M., Tuch, T., Weinhold, K., Merkel, M., Rasch, F., Müller, T., Schladitz, A., Bastian, S., Löschau, G., Cyrys, J., Gu, J., Flentje, H., Briel, B., Asbach, C., Kaminski, H., Ries, L., Sohmer, R., Gerwig, H., Wirtz, K., Meinhardt, F., Schwerin, A., Bath, O., Ma, N., and Wiedensohler, A.: Decreasing trends of particle number and black carbon mass concentrations at 16 observational sites in Germany from 2009 to 2018, Atmos. Chem. Phys., 20, 7049-7068, 10.5194/acp-20-7049-2020, 2020.

Teinilä, K., Aurela, M., Niemi, J. V., Kousa, A., Petäjä, T., Järvi, L., Hillamo, R., Kangas, L., Saarikoski, S., and Timonen, H.: Concentration variation of gaseous and particulate pollutants in the Helsinki city centre — observations from a two-year campaign from 2013–2015, Boreal Environ. Res., 24, 115-136, 2019.

Timonen, H., Karjalainen, P., Saukko, E., Saarikoski, S., Aakko-Saksa, P., Simonen, P., Murtonen, T., Dal Maso, M., Kuuluvainen, H., Bloss, M., Ahlberg, E., Svenningsson, B., and Pagels, J.: Influence of fuel ethanol content on primary emissions and secondary aerosol formation potential for a modern flex-fuel gasoline vehicle, Atmos. Chem. Phys., 17, 5311-5329, 10.5194/acp-17-5311-2017, 2017.

Timonen, H., Karjalainen, P., Aalto, P., Saarikoski, S., Mylläri, F., Karvosenoja, N., Jalava, P., Asmi, E., Aakko-Saksa, P. i., Saukkonen, N., Laine, T., Saarnio, K., Niemelä, N., Enroth, J., Väkevä, M., Oyola, P., Pagels, J., Ntziachristos, L., Cordero, R., Kuittinen, N., Niemi, J., and Rönkkö, T.: Adaptation of black carbon footprint concept would accelerate mitigation of global warming, Environ. Sci. Technol., 53 (21), 12153-12155, 10.1021/acs.est.9b05586, 2019.

Vallius, M. J., Ruuskanen, J., Mirme, A., and Pekkanen, J.: Concentrations and estimated soot content of PM1, PM2. 5, and PM10 in a subarctic urban atmosphere, Environ. Sci. Technol., 34, 1919-1925, 10.1021/es990603e, 2000.

Weichenthal, S., Farrell, W., Goldberg, M., Joseph, L., and Hatzopoulou, M.: Characterizing the impact of traffic and the built environment on near-road ultrafine particle and black carbon concentrations, Environ. Res., 132, 305-310, 10.1016/j.envres.2014.04.007, 2014.

Virkkula, A., Mäkelä, T., Hillamo, R., Yli-Tuomi, T., Hirsikko, A., Hämeri, K., and Koponen, I. K.: A simple procedure for correcting loading effects of Aethalometer data, J. Air Waste Ma., 57, 1214-1222, 10.3155/1047-3289.57.10.1214, 2007.

Yuan, J., Modini, R. L., Zanatta, M., Herber, A. B., Müller, T., Wehner, B., Poulain, L., Tuch, T., Baltensperger, U., and Gysel-Beer, M.: Variability in the mass absorption cross-section of black carbon (BC) aerosols is driven by BC internal mixing state at a central European background site (Melpitz, Germany) in winter, Atmos. Chem. Phys. Discuss., 2020, 1-36, 10.5194/acp-2020-41, 2020.

Zhao, B., Jiang, J. H., Gu, Y., Diner, D., Worden, J., Liou, K.-N., Su, H., Xing, J., Garay, M., and Huang, L.: Decadal-scale trends in regional aerosol particle properties and their linkage to emission changes, Environ. Res. Lett., 12, 054021, 10.1088/1748-9326/aa6cb2, 2017.

Zhu, Y., Hinds, W. C., Kim, S., Shen, S., and Sioutas, C.: Study of ultrafine particles near a major highway with heavy-duty diesel traffic, Atmos. Environ., 36, 4323-4335, 10.1016/S1352-2310(02)00354-0, 2002.

Zotter, P., Herich, H., Gysel, M., El-Haddad, I., Zhang, Y., Močnik, G., Hüglin, C., Baltensperger, U., Szidat, S., and Prévôt, A. S.: Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol, Atmos. Chem. Phys., 17, 4229-4249, 10.5194/acp-17-4229-2017, 2017.

Table 1: Annual mean values of eBC concentration for each station in units of µg m^{-3}. Color-coding indicates the magnitude of the mean concentration; darker color refers to higher concentration. The annual means at UB2 in 2016—2017 are bracketed, since there was less than 50 % of valid data.

Station	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
TR1					1.27		0.90	0.81	0.72	0.80	0.68	0.73
TR2									1.35	1.24	1.08	0.99
TR3				2.64					1.55			
TR4						1.58						
TR5								0.91		0.76		0.83
TR6									0.88		0.67	
DH1			0.80									
DH2								0.80				
DH3										0.65		
DH4											0.64	
DH5												0.74
UB1						0.68	0.59	0.53	0.52	0.50	0.42	0.49
UB2									0.54	(0.56)	(0.44)	0.50
RB1										0.26		0.29
RB2	0.32	0.41	0.37	0.43	0.37		0.21	0.25	0.21	0.19	0.16	0.19

Table 2: The traffic counts for working days at the nearest streets to the TR ~~stations~~sites. The traffic counts and the fraction of the heavy-duty are from ~~from~~ the yearly Helsinki traffic reports. ~~The traffic rates are given for working days only.~~ The streets and roads mentioned here are marked in ~~the~~ Fig~~s~~. S3.

Station	Street name	Traffic count (vehicles/weekday)	Heavy-duty (%)	Distance to street edge (m)	Reference year
TR1	Mannerheimintie	15 800	11	3	2017
	Kaivokatu	20 100	8	40	2017
TR2	Mäkelänkatu	28 100	11	0.5	2017
TR3	Mannerheimintie	44 400	14	0.5	2010
	Reijolankatu	19 400	-	25	2010
TR4	Kehä I	69 200	8	5	2012
	Tattariharjuntie	13 700	13	120	2012
TR5	Tikkurilantie	9 500	-	7	2016
TR6	Turuntie	29 300	4	20	2017
	Lintuvaarantie	15 400	5	30	2017
	Kehä I	68 900	4	250	2017

Table 3: Results of the trend analysis of the eBC, $PM_{2.5}$, and NO_x concentrations. The values without brackets are the absolute trends in units of µg m^{-3} yr^{-1}, the values in the square brackets are the 5^{th} and 95^{th} uncertainty limits of the trend in units of µg m^{-3} yr^{-1}, and the bracketed values are the relative trends in units of % yr^{-1}. The values for $PM_{2.5}$ at TR2 are italicized, since they were statistically not significant (p-value = 0.05).

Station	eBC (µg m^{-3} yr^{-1})	$PM_{2.5}$ (µg m^{-3} yr^{-1})	NO_x (µg m^{-3} yr^{-1})	Measurement years
TR1	-0.04 [-0.06; -0.02] (-6.5 % yr^{-1})	-0.24 [-0.38; -0.01] (-3.7 % yr^{-1})	-3.11 [-4.18; -1.87] (-7.1 % yr^{-1})	2011, 2013–2018
TR2	-0.09 [-0.11; -0.05] (-10.6 % yr^{-1})	*-0.46* *[-0.71; 0.02]* *(-7.1 % yr^{-1})*	-11.00 [-12.47; -8.70] (-19.7 % yr^{-1})	2015–2018
UB1	-0.02 [-0.03; -0.01] (-5.7 % yr^{-1})	-0.20 [-0.34; -0.05] (-3.9 % yr^{-1})	-0.80 [-1.08; -0.50] (-5.0 % yr^{-1})	2012–2018
RB2	-0.01 [-0.02; -0.01] (-7.6 % yr^{-1})	-0.10 [-0.20; -0.03] (-2.7 % yr^{-1})	-0.05 [-0.08; -0.03] (-4.0 % yr^{-1})	2006–2018

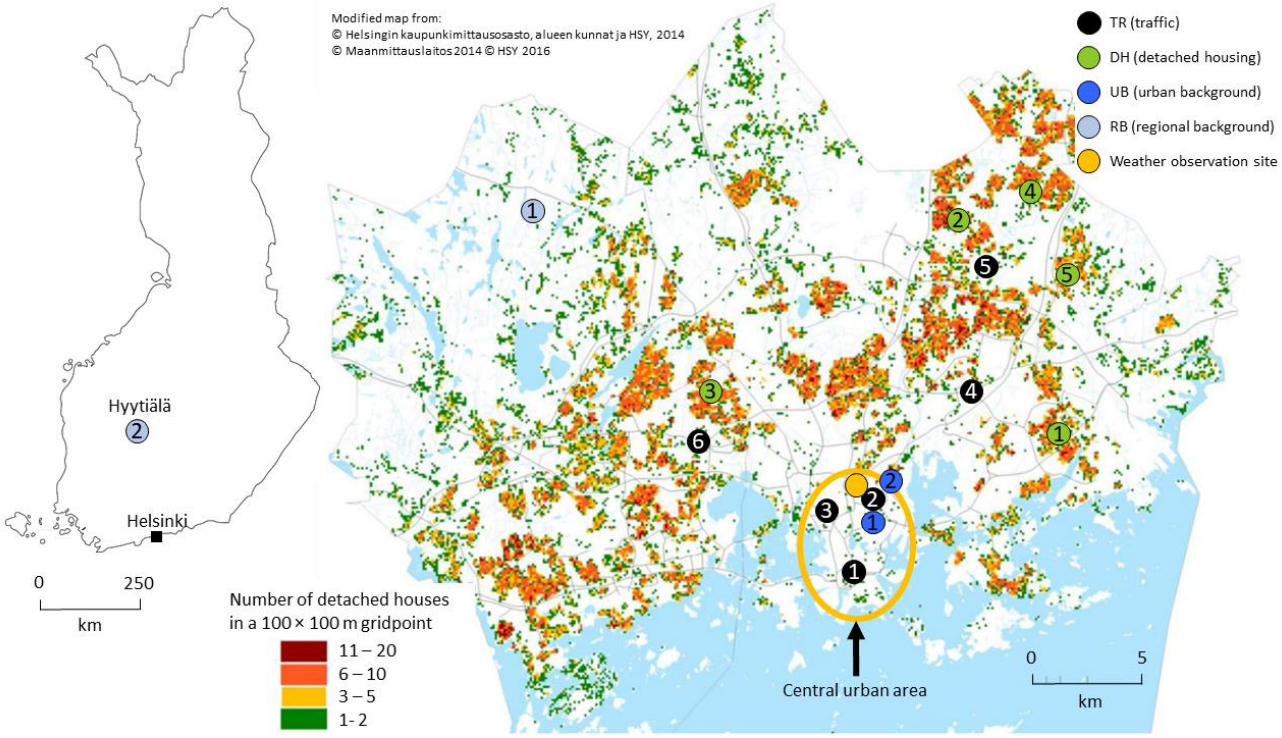

Figure 1: The locations of the stations and the density of the detached houses. Differently colored markers indicate different station categories. The central urban area is marked to the map with an orange circle.

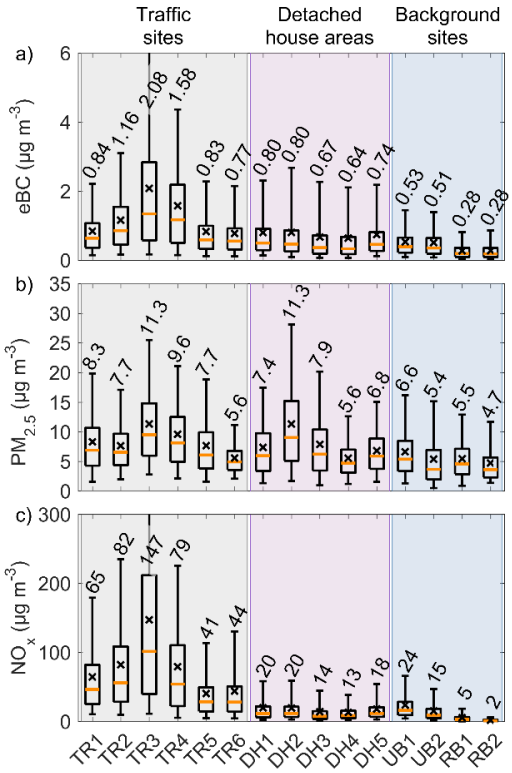

Figure 2: The statistics of the a) eBC, b) $PM_{2.5}$, and c) NO_x concentrations at each station. The boxplots are presented for 1 h mean values. The orange line in the middle of each box represents the median, the edges of the boxes represent the 25th and 75th percentiles, and the whiskers represent the 5th and 95th percentiles. The black cross is the arithmetic mean, and its numerical value is reported above or below each box. The background color represents the station type: gray for traffic sites (TR), purple for the detached housing sites (DH), and blue for the background sites (UB for urban and RB for regional background). The 75th percentiles of eBC and NO_x concentration at TR3, which are not visible at the figure, were 6.7 and 440 μg m^{-3}, respectively.

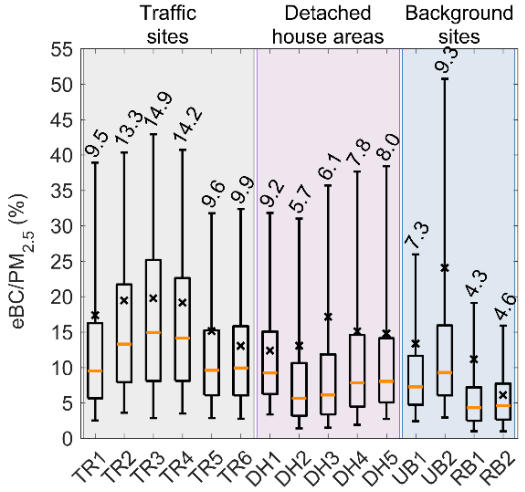

Figure 3: The statistics of eBC/$PM_{2.5}$ fraction at each station. The explanation for the markers is the same as in Fig. 2, except here the values reported above each box are the median values.

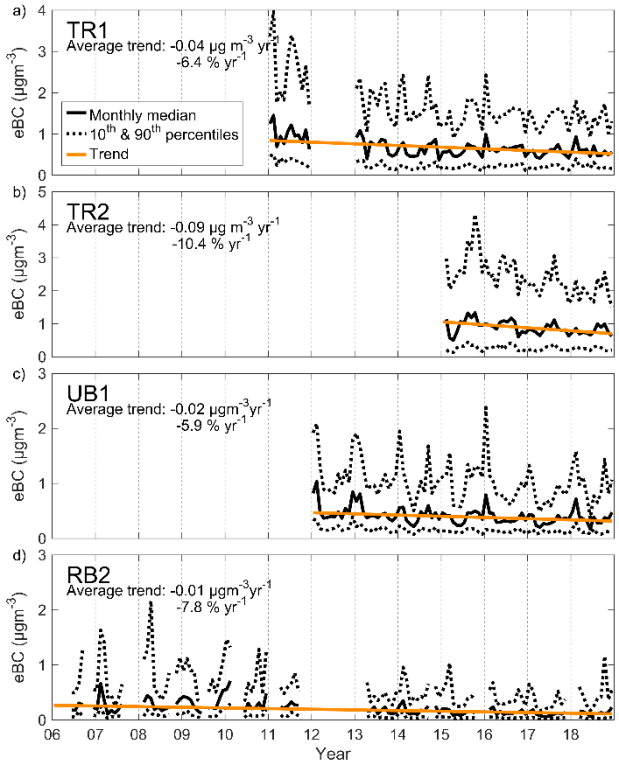

Figure 4: Time series and the trends of eBC concentration at TR1, TR2, UB1, and RB2. The solid black line represents the monthly medians, the dashed lines represent the 10th and 90th monthly percentiles, and the orange line is the fitted long-term trend.

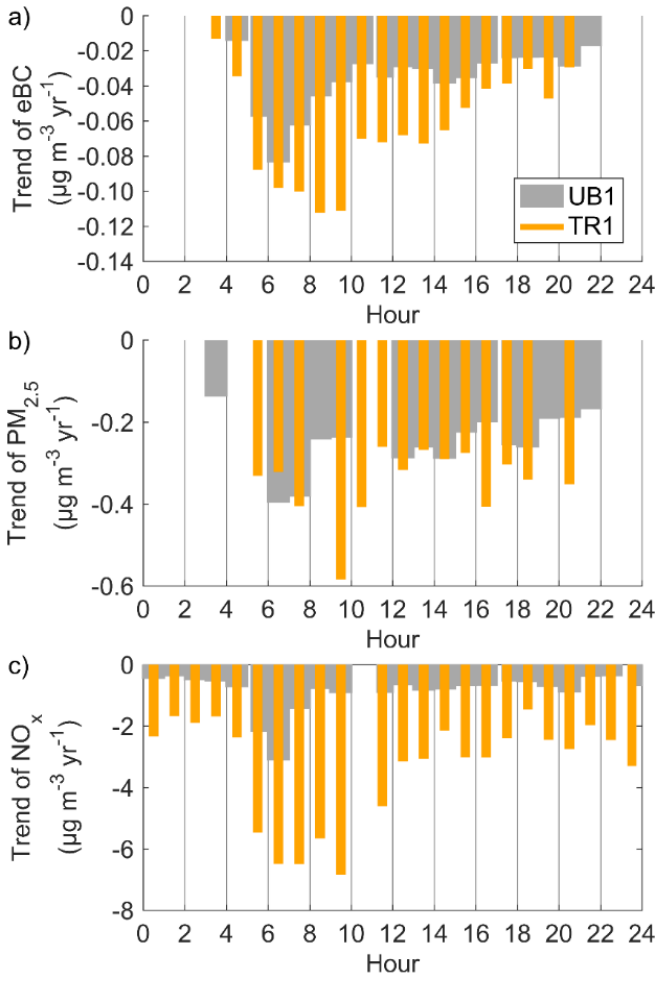

Figure 5: Annual t~~T~~rends for the hourly data. Here, only data from weekdays were used. None of the pollutants had a positive trend for any time of the day.

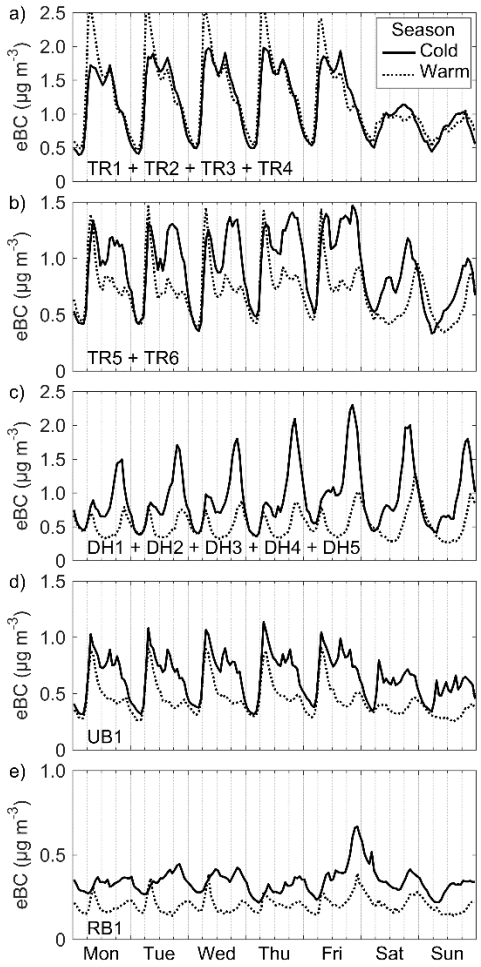

Figure 6: Diurnal variation of eBC for different station categories: a) traffic sites that we~~a~~re not influenced by wood burning (TR1 –~~–TR~~4), b) traffic sites that ~~a~~were influenced by wood burning (TR5–~~–TR~~6), c) detached housing sites (DH1–~~–DH~~5), d) urban background (UB1), and e) regional background (RB1). The diurnal variation is determined separately for the cold (from November to March) and warm (from May to September) periods.