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Long-term air quality trends of regulated pollutants in the Helsinki metropolitan area from 1994–2019 and its implications to the Air Quality Index

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Long-term trends of ambient gaseous pollutants and particulate matter in Helsinki metropolitan area were analyzed from 1994 to 2019. Measurement data from ten monitoring stations located in different types of urban environments including traffic, urban background, rural background, and suburban area were included in this study. We analyzed gas-phase air pollutants, such as NO, NO_2, NO_x, O_3, SO_2 and CO; and for aerosol pollutants, we explored mass concentrations for particles smaller than 10 μm and 2.5 μm in diameter (PM_{10} and $PM_{2.5}$, respectively). In order to quantify trends in the data, we deployed a non-parametric Mann-Kendall test and Theil-Sen method. The results were compared with the regional emissions trends and changes in meteorological conditions. Our analysis indicates that SO_2 and CO in all stations have decreased to values corresponding to their regional background concentration levels and their role as urban air pollutants have diminished. Our results from the Helsinki Metropolitan area during the last 25 years show that the air quality improved and all the air pollutant concentrations show a decreasing trend, except ozone. Based on our analysis of the Air Quality Index (AQI) at traffic and urban background environments, NO_2 concentration, which have typically represented the health effects resulting from vehicular traffic, is rapidly decreasing also in traffic environments. The current AQI standard therefore lacks clarity on the potential health risks from other air pollutants emitted from traffic exhaust. In addition, the air quality indicators currently considered in the AQI do not represent well enough residential wood burning and the possible health outcomes from its exposure. We suggest that the current AQI should be revised in a way that new air quality parameters would be considered, which would better represent the health effects resulting from these local combustion sources.

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Introduction

Continuous exposure to high concentrations of gas-phase and particulate pollutants has adverse effects on human health and the environment (WHO 2016, EEA 2019). Both short- and long-term exposure have been associated with several diseases including cardiovascular diseases, respiratory diseases, and lung cancer (Hoek *et al.* 2013, Pope III *et al.* 2002, Silva *et al.* 2013). According to Lelieveld *et al.* (2015), exposure to outdoor air pollution causes around 3.3 million premature deaths globally every year.

The Air Quality Index (AQI) is a communicational tool, which is used to express the health risks associated with certain air pollution exposure level in an easily understandable manner. There are several different ways to calculate AQIs globally, but not a single one that would be universally applicable. Usually, AQI is calculated from several air pollutants (known as air quality indicators), namely particulate matter with diameter less than 2.5 μm ($PM_{2.5}$) and less than 10 μm (PM_{10}) along with gaseous pollutants like ozone (O_3), nitrogen oxides (NO_x), sulfur dioxide (SO_2) (WHO 2005). The AQI is calculated for each parameter based on its limit values and the air pollutant with the highest index value determines the final AQI value (Hewings 2001). The accounted air pollutants in the index and their limit values vary among countries. Currently used AQIs have been criticized, because they do not consider the cumulative effects of air pollutants nor the concentration-response relationship between exposure and the resulting health effects (Stieb *et al.*, 2008).

The Helsinki metropolitan area (HMA) is the most densely populated area in Finland with approximately 1.17 million (as of Dec 2019) habitants. Air quality in HMA and in Finland in general is relatively good for most of the time compared with many other areas around Europe (EEA 2019, Ohtonen *et al.* 2020). Nevertheless, exposure to outdoor air pollution causes more than 1500 premature deaths every year in Finland, with the majority linked to $PM_{2.5}$ exposure (EEA 2019).

The Finnish air quality trends were evaluated in Anttila (2020). Their study included trends for SO_2, CO, NO_2, NO_x, O_3 and PM_{10} from the period between 1994 and 2018. The results indicate that the air pollutant concentrations for which there is international or national regulatory control mechanisms, are decreasing. Removal of sulfur from fuels have led to a drastic reduction of ambient SO_2 concentrations also globally (Aas *et al.* 2019). NO_x emission control has been successful in Finland although NO_2 concentrations have not decreased as expected. This is connected to the increased number of diesel vehicles. O_3 concentrations have been declining in Finland since 2006, most plausibly connected to reduced anthropogenic NO_x and volatile organic compound (VOC) emissions. However, the regional O_3 concentration is influenced by strong natural VOC emissions from the biosphere.

Concentrating into HMA, the main local sources of air pollution are traffic exhaust and residential wood burning (e.g., Pirjola *et al.* 2012, Aurela *et al.* 2015). Additional relevant local sources are traffic-induced road dust episodes especially during the springtime, energy production, at airport and ports. Air pollution episodes occur occasionally in the HMA. These events are typically due to temperature inversions in which the locally emitted air pollutants are trapped within the boundary layer (Kukkonen *et al.* 2005) or long-range transportation (LRT) of air pollution from Eastern Europe (Niemi *et al.* 2009), e.g., smoke emitted from local and transboundary wildfires (Leino *et al.* 2014). Based on the data of campaigns from 2013 to 2015 in the city center of Helsinki, Teinilä *et al.* (2019) explored the impacts of local sources to the PM_1 chemical composition. They showed that the sub-micron aerosol mass load and chemical composition is governed by local traffic, long-range transport, and local aerosol sources.

The objective of this study is to quantify the changes and trends of air pollution indicators ($PM_{2.5}$, PM_{10}, O_3, NO_2, NO_x ($NO+NO_2$), SO_2 and CO) measured in different types of environments in the HMA based on the data from 1994 to 2019. Furthermore, we aim to investigate the effects and the relationship between regional emission trends and meteorological changes in the AQI.

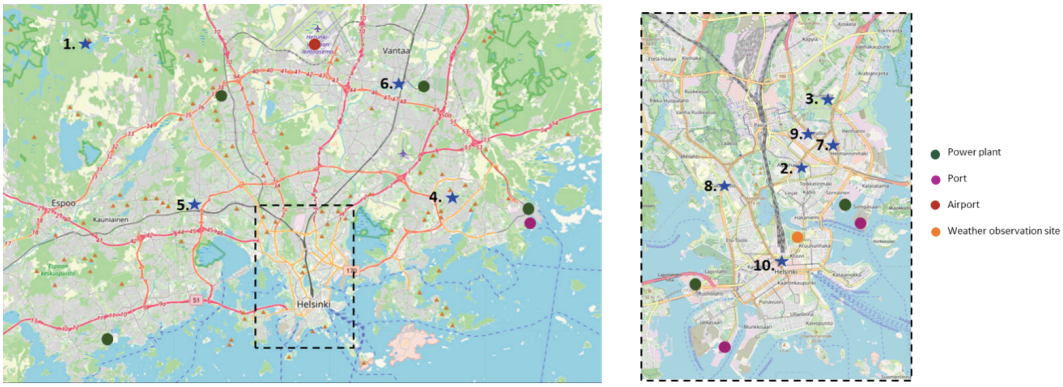

Fig. 1. Left: Locations of the monitoring sites (blue stars) in the HMA region. Right: Location of the stations in downtown area indicated with square. The station names and descriptions are presented in Table 1.

Material and methods

Monitoring sites

The data used in this study were collected from the HMA, which is located in Southern Finland on the shore of the Gulf of Finland (60°10′N, 24°56′E). The HMA consists of four cities: Helsinki, Espoo, Vantaa and Kauniainen. The region has an area of 765 km^2 and a total population of approximately 1.17 million people, which makes it the most densely populated area in Finland.

The climate in Southern Finland is a mixture of continental and maritime climates. The weather is highly dependent on the tracks of low- and high-pressure systems. Due to the location in the northern latitudes, temperature has a distinct seasonal cycle. The coldest month from 1997–2019 was February, with a mean temperature of –4ºC and the warmest month was July, with a mean temperature of 18ºC. The mean wind speed for the whole measurement period (1997–2019) was 3.5 m/s and the most common wind direction was from south-west. For more information on changes in the meteorological parameters, see Figs. S1–S3 in the Supplementary Information.

Air quality data were collected from ten sites (i.e., air quality stations) within the HMA. The stations were in different types of environments including urban traffic, detached housing areas, urban and regional background areas (Fig. 1 and Table 1). In addition, we used reference data from SMEAR II (Station for Measuring Ecosystem — Atmosphere Relations II) station, which is a rural background station located in Hyytiälä, Southern Finland (61º51′N, 24º17′E), (Hari and Kulmala 2005) and SMEAR III in Kumpula Science Campus (Järvi *et al.* 2009). Helsinki Region Environmental Services (HSY) operated all the monitoring stations, except for SMEAR II and SMEAR III stations, which are operated by the University of Helsinki and the Finnish Meteorological Institute (FMI).

Meteorological data

In this study, we used meteorological data from Kaisaniemi station operated by FMI. The monitoring site was in a botanical garden in the downtown area, three meters above the sea level. The measured parameters included temperature, air pressure, relatively humidity, precipitation, wind speed and wind direction. The hourly data were available starting August 1996 for temperature, wind speed and relative humidity. Wind direction and daily rainfall data were available starting from September 1996 and from January 1994, respectively. Mixing layer height for southern Finland was calculated by a meteorological pre-processing model developed at FMI (MPP-FMI; Karppinen *et al.* 2000).

Emission data

Five-year mean air pollution emissions from the HMA region are presented in Table 2 (Korhonen

Table 1. Air quality monitoring sites in the HMA region and their classification, data coverage, distance to the closest road and traffic counts (vehicles/weekday) on the closest roads at the start and end of the measurement period. The number before the station name is used in Fig. 1 to show the location on the map.

Station	Type	Measurement period	Closest road(s)	Distance from street (m)	Traffic count (vehicles/weekday)	
					Measurement start	Measurement end
1. Luukki	Rural background (RB)	1994–2019	Vihdintie	800	7700	6000
2. Kallio	Urban background (UB)	1999–2019	Helsinginkatu	80	7700	4800
			Sturenkatu	300	31 000	25 600
3. SMEAR III	Urban background (UB)	2004–2019	Kustaa Vaasan tie	150	44 680	40 670
4. Vartiokylä	Suburban detached (DH)	2009–2019	Riskutie	60	2400	2400
			Itäväylä	500	16 800	19 300
. 5. Leppävaara	Traffic (TR)	2010–2019	Turuntie	30	29 300	23 000
			Lintuvaarantie	30	15 500	16 000
			Ring I	250	74 900	89 000
6. Tikkurila	Traffic (TR)	1996–2019	Tikkurilantie	7	12 000	8100
			Ratatie	27	n.a.	3800
7. Vallila	Urban traffic (TR)	1994–2014	Hämeentie	14	16 000	10 900
			Mäkelänkatu	200	25 000	21 000
			Sturenkatu	300	20 000	16 200
8. Töölö	Urban traffic (TR)	1994–2004	Nordenskiöldinkatu	303	14 000	14 400
			Mechelininkatu	10	24 000	24 800
			Topeliuksenkatu	8	19 000	16 900
			Linnankoskenkatu	25	18 000	11 800
9. Mäkelänkatu	Urban traffic (TR)	2015–2019	Mäkelänkatu	0.5	28 300	27 800
10. Mannerheimintie	Urban traffic (TR)	2005–2019	Mannerheimintie	3	16 500	14 900
			Kaivokatu	35	15 300	18 400

n.a. = not available

Table 2. Five-year mean emissions of NO, PM, CO and SO_2 in the HMA expressed in tons yr^{-1} and their trend during the study period from different sectors.

Pollutant	Source	Average annual emissions (tons yr^{-1})					Change (% yr^{-1})
		1995–1999	2000–2004	2005–2009	2010–2014	2015–2019	1994–2019
NO_x	Total	20 710	19 342	16 655	14 032	11 473	–2.4
	Energy production	8748	7744	7736	7285	5689	–2.1
	Vehicular traffic	11 763	9470	7013	4763	3803	–2.9
	Industry	199	47	31	54	8	–1.6
	Harbours	–	1504	1258	1332	1253	–1.1
	Airport	–	577	617	598	720	+1.9
PM	Total	1561	1026	622	389	260	–3.5
	Energy production	854	522	274	181	137	–3.5
	Vehicular traffic	647	443	288	171	112	–3.4
	Industry	60	16	9	7	3	–3.6
	Harbours	–	–	50	29	7	–5.6
	Airport	–	–	1	1	1	0
CO	Total	47 936	36 072	21 976	12 270	7187	–3.4
	Vehicular traffic	47 936	35 179	20 664	11 240	6161	–3.5
	Harbours	–	–	151	130	128	–0.3
	Airport	–	–	1161	900	898	–1.1
SO_2	Total	7508	6251	5145	4598	4017	–2.8
	Energy production	7100	5806	4755	4375	3911	–2.7
	Vehicular traffic	93	23	8	7	5	–3.8
	Industry	315	83	41	17	2	–3.7
	Harbours	–	293	289	151	43	–3.4
	Airport	–	46	52	48	56	+2.6

et al. 2020). Reported emission data is divided into six emission sectors: vehicular traffic, energy production, industry, airport, harbours and residential wood burning. The vehicular traffic emissions includes of traffic exhaust emissions, but the street dust resuspension was not included. Unfortunately, long-term trends of the residential wood burning emissions in the HMA are not available because the emission factors and the method used have changed over the years.

Air Quality Index

In this study, we used European AQI standards to calculate AQI for five main air pollutants ($PM_{2.5}$, PM_{10}, O_3, NO_2 and SO_2), which are legislated by the European Union (EEA 2017). Based on the calculated AQI, the air quality is classified into six index levels (Good, Fair, Moderate, Poor, Very poor and Extremely poor):

$$\mathrm{AQI} = \frac{I_{\mathrm{high}} - I_{\mathrm{low}}}{C_{\mathrm{high}} - C_{\mathrm{low}}}(C - C_{\mathrm{low}}) + I_{\mathrm{low}}, \qquad (1)$$

where C is the ambient concentration of each air pollutant; C_{high} and C_{low} are the upper and lower limit of the pollutant concentration of the index level C belongs to, respectively; I_{high} and I_{high} are the upper and lower index value of the corresponding index level, respectively. For the gases, hourly concentrations were used as inputs to AQI. For $PM_{2.5}$ and PM_{10}, the running mean for the past 24 hours were used. Table 3 shows the concentration range of each air pollutant corresponding to each Air Quality Index level in the European AQI used in this paper (EEA 2017).

Data Processing

In this study we used hourly averaged data for air pollution data. Data from HSY was quality assured by the data producer and invalid data were omitted from further analysis. At Mannerheimintie (see Table 1 station 10), the traffic site data was between 8:00–9:00 and 21:00–22:00 (local time). The data quality was poor due to technical issues caused by the air conditioning between 2014 and 2019. For SMEAR II and SMEAR III stations, data was downloaded from Smart-SMEAR website as one-hour averages (Junninen *et al.* 2009). To unify the data, the unit for gas-phase pollutants from SMEAR II and SMEAR III stations was changed from ppbv to μg m^{-3}, which is the unit used by HSY and the limit values set by WHO and the European Union. The conversion between the mass concentration and volume mixing ratio was as follows:

$$C(\text{in } \mu\mathrm{g m^{-3}}) = \frac{C(\text{in ppbv}) \times 12.187 \times M}{273.15 + T}, \qquad (2)$$

where C is the mass concentration or volume mixing ratio of the compound, M is the molecular weight of the gas in g/mol and T is the temperature in ºC. The atmospheric pressure is assumed to be 1013 hPa and the temperature 20ºC. To unify the time unit, data from SMEAR stations was changed to local time (UTC+2 hours during winter and UTC+3 hours during summer). In our analysis, we only included the months, which had at least 75% valid data. The minimum length for time series was set to four years. No gap-filling to the data was performed and therefore the averages are based on the available observational data.

Table 3. Limit values for different pollutants ($PM_{2.5}$, PM_{10}, O_3, NO_2, SO_2 in $\mu g\ m^{-3}$) for different index levels (Good, Fair, Moderate, Poor, Very poor, Extremely poor) in the European AQI used in this paper.

Index level	$PM_{2.5}$	PM_{10}	O_3	NO_2	SO_2
Extremely poor	> 75	> 150	> 380	> 340	> 750
Very poor	50–75	100–150	240–380	230–340	500–750
Poor	25–50	50–100	130–240	120–230	350–500
Moderate	20–25	40–50	100–130	90–120	200–350
Fair	10–20	20–40	50–100	40–90	100–200
Good	< 10	< 20	< 50	< 40	< 100

Statistical analysis of trends

Temporal variations of air pollutants were investigated with the *Openair* package in R analysis software ver. 4.0.3 (Carslaw and Ropkins 2012). We used seasonal Mann-Kendall test and Theil-Sen method for the trend analysis. Both are suitable for non-normally distributed data and allow missing data points. The Mann-Kendall test assesses whether the parameter of interest shows a monotonic decreasing or increasing trend in time (Hirsch *et al.* 1982). The Theil-Sen method is used to calculate the slope and strength of the trend. In our analysis, we used monthly mean values, which were adjusted for seasonal variation (Sen 1968, Theil 1992). A similar type of analysis has been used in many previous trend studies (e.g. Olstrup *et al.* 2018)

Results and discussion

Overall trends of pollutants in the HMA

Annual air pollution emissions decreased remarkably in the HMA from 1994–2019 (Table 2 and Fig. S4 in Supplementary Information). Total NO_x, PM, CO and SO_2 emissions declined 62% (2.4% yr^{-1}), 91% (3.7% yr^{-1}), 88% (3.4% yr^{-1}) and 70% (2.7% yr^{-1}), respectively. The highest emission cuts were observed in vehicular traffic and energy production sectors, which are the dominant emissions sources in the HMA together with residential wood burning. Similar trends have been observed in Europe as the emissions were reduced by 60%, 69%, 92% for NO_x, CO and SO_x, respectively from 1990–2018. $PM_{2.5}$ and PM_{10} emissions were cut by 32% and 29% from 2000–2018 (EEA 2020).

Table 4 presents a summary of results for the trends and measurement periods of pollutants (CO, SO_2, NO_2, NO, NO_x, O_3, $PM_{2.5}$ and PM_{10} concentration) at ten stations around the HMA. Six of these were traffic sites (TR), two were urban background (UB), one was a rural background (RB) site, and one was a suburban detached housing (DH) area. Our results show a decreasing trend for all air pollutants in the studied sites, except for O_3. Due to great abatement efforts, CO and SO_2 show the most notable decrease in annual mean concentration which varied between 2.9% yr^{-1} and 5.4% yr^{-1} for CO

Table 4. Summary of the trends and measurement periods for the measured air pollutants in the HMA. The measured air pollutants are on the rows and the station names on the columns of the table. On the first row of each box is the absolute annual change in $\mu g\ m^{-3}\ yr^{-1}$ (for CO, the unit is in $mg\ m^{-3}\ yr^{-1}$) and the statistical significance of the trend (*** = $p < 0.001$, ** = $p < 0.01$, *= $p < 0.05$, += $p < 0.1$). The annual trend in percentages is shown on the second row and the measurement years on the third row. The white boxes with NA indicate that there are no data available, or the measurement period is too short. The boxes are coloured according to the annual change in percentages. Blue colours represent decrease and red colours increase.

	RB Luukki	UB Kallio	UB SMEAR III	DH Vartiokylä	TR Leppävaara	TR Tikkurila	TR Vallila	TR Töölö	TR Mäkelänkatu	TR Mannerheimintie
CO	NA	NA	0*** 0 % 2006-2019	-0.01*** -3.2 % 2009-2013	NA	-0.02*** -3.4 % 1996-2016	-0.01*** -2.9 % 1994-2004	-0.05*** -5.4 % 1994-2004	NA	-0.02*** 4.6 % 2005-2013
SO_2	-0.08 *** -3.3 % 1994-2019	-0.2 *** -13 % 2014-2019	-0.03*** -4.1 % 2006-2019	NA	NA	NA	-0.2*** -3.0 % 1994-2014	-0.6** -8.7 % 1994-1998	NA	NA
NO_2	-0.1 *** -1.7 % 1994-2019	-0.5*** -1.9 % 1999-2019	-0.6*** -3.6 % 2005-2019	-0.5*** -3.0 % 2009-2019	-0.2*** -0.8 % 2010-2019	-0.5*** -1.5 % 1996-2019	-0.4*** -1.4 % 1994-2015	-0.4 * -1.1 % 1994-2004	-3.8 *** -8.8 % 2015-2019	-1.3*** -2.7 % 2005-2019
NO_x	-0.4*** -2.4 % 1994-2019	-0.9*** -2.3 % 1999-2019	-1.0*** -4.3% 2005-2019	-1.0*** -3.9% 2009-2019	-2.3*** -3.9% 2010-2019	-2.4*** -2.6% 1996-2019	-1.5*** -2.4% 1994-2014	-6.7*** -4.6% 1994-2004	-11.1 -10.9% 2015-2019	-3.0*** -3.2% 2005-2019
O_3	-0.09 ** -0.2 % 1994-2019	0.3 *** 0.6 % 1999-2019	0.9 *** 2.0 % 2005-2019	0.4* 0.8 % 2009-2019	NA	NA	-0.5 -1.2 % 1996-1999	0.9 *** 2.8 % 1994-2004	2.1 *** 6.1 % 2015-2019	0.4* 1.1 % 2005-2015
$PM_{2.5}$	-0.3 *** -2.8 % 2004-2019	-0.3 *** -2.3 % 1999-2019	NA	-0.2 ** -2.5 % 2009-2019	-0.3 ** -3.6 % 2010-2019	-0.2 * -2.1 % 2009-2019	-0.2+ -1.9 % 1997-2003	NA	-0.5*** -5.8 % 2015-2019	-0.3*** -2.7 % 2005-2019
PM_{10}	NA	-0.2 *** -1.4% 1999-2019	NA	NA	-0.3* -1.6 % 2010-2019	-0.4*** -2.0 % 1998-2019	-0.3*** -1.5 % 1996-2015	-0.6*** -2.3 % 1994-2004	-1.2*** -5.1 % 2015-2019	-0.6*** -2.1 % 2005-2019

and between 3.0% and 13% for SO_2. Five traffic and suburban detached housing areas show a statistically significant increasing trend for O_3, whereas a statistically significant small increasing trend was detected at the rural background site (Luukki).

Nitrogen oxides and ozone

NO_x ($NO+NO_2$) emissions result from combustion processes, which take place in e.g., car engines (e.g., Vestreng *et al.* 2009) and during energy production (e.g., Oberschelp *et al.* 2019). The World Health Organization and European Environmental Agency have set guideline values for NO_2, as elevated levels are associated with several severe health effects, including reduced lung function and increased inflammation of airways (WHO 2005).

NO_2 is the most abundant pollutant in traffic environments, where its concentration is highly influenced by atmospheric chemical interaction with NO, NO_2 and O_3. Most tailpipe NO_x emissions consist of NO and only small share consists of NO_2. While NO_2 is relatively unreactive in near-road environment, NO reacts quickly with O_3 to form additional NO_2 (Wang 2011). This chemical reaction is dependent on the available ambient O_3 concentration.

From 1994–2019, the dominant source of NO_x in the HMA switched from vehicular traffic to energy production. In total, vehicular traffic and energy production NO_x emissions decreased 62% (2.4% yr^{-1}), 75% (2.9% yr^{-1}) and 71% (2.7% yr^{-1}), respectively. One key reason for the successful reduction of vehicular traffic emissions is that the European Union (EU) started to regulate emissions of new vehicles sold in EU and EEA region in 1992. The first EU-wide standard was known as EURO1, which made three-way catalytic converter (TWCs) compulsory in all new petrol vehicles. TWCs reduce the amount of NO_x, and CO emissions from traffic exhaust (Twigg 2011).

The time series of ambient NO_x and NO_2 concentrations were studied at ten stations around the HMA. Both parameters showed a statistically significant decreasing trend at all these sites. The absolute annual reductions of NO_x were greater for the traffic sites (1.5–11.1 μg m^{-3} yr^{-1}) than for the urban background, rural background and detached housing area (0.4–1.0 μg m^{-3} yr^{-1}). For NO_2, the urban traffic sites Mannerheimintie and Mäkelänkatu showed higher reductions (1.3–3.8 μg m^{-3} yr^{-1}), while for the other sites the annual reduction varied between 0.1–0.6 μg m^{-3} yr^{-1}. For all sites, the relative decrease was higher for NO_x than for NO_2. The decrease varied in the ranges between 2.3–3.5% yr^{-1} and 1.2–1.9% yr^{-1} for NO_x and NO_2, respectively, except for Mäkelänkatu street canyon where the annual reductions were 10.9% yr^{-1} for NO_x and 8.8% yr^{-1} NO_2 (2015–2019; Table 2). The trends for NO_x, NO_2 and O_3 are represented in the two traffic sites, urban background site and rural background site (Fig. 2).

As the vehicular traffic is one of the dominant sources of NO_x and NO_2 in the HMA, the highest concentrations were measured in the traffic environments (Mannerheimintie and Tikkurila). During the 15-year measurement period in Mannerheimintie, the annual mean NO_x and NO_2 concentrations almost halved from 90.7 μg m^{-3} and 42.9 μg m^{-3} , respectively, in 2005, to 43.7 μg m^{-3} and 24.9 μg m^{-3} ,respectively, in 2019. At the same time daily traffic count decreased approximately 10% in Mannerheimintie (3 m from the site) and increased 20% in Kaivokatu (35 m from the site). The Helsinki Regional Transport Authority started to renew their bus fleet to ones that meet the newest EURO6 standard in 2014. This is one of the key reasons, why the NO_x and NO_2 concentrations have decreased rapidly in urban traffic sites. At the rural background site in Luukki, where the closest road is 800 meters away, the annual mean NO_2 concentration decreased from 6.2 μg m^{-3} to 4.9 μg m^{-3} from 2005–2019. The longest time series of NO_x were measured in Luukki (RB), Kallio (UB) and Tikkurila (TR), where the annual decrease varied between 2.3–2.4% yr^{-1} which is consistent with total regional NO_x emissions reduction (2.4% yr^{-1}).

The seasonal and diurnal cycle of air pollutant concentrations at traffic, urban background and rural background sites are shown in Figs. S5 and S6 in Supplementary Information. The monthly mean NO_x and NO_2 concentrations

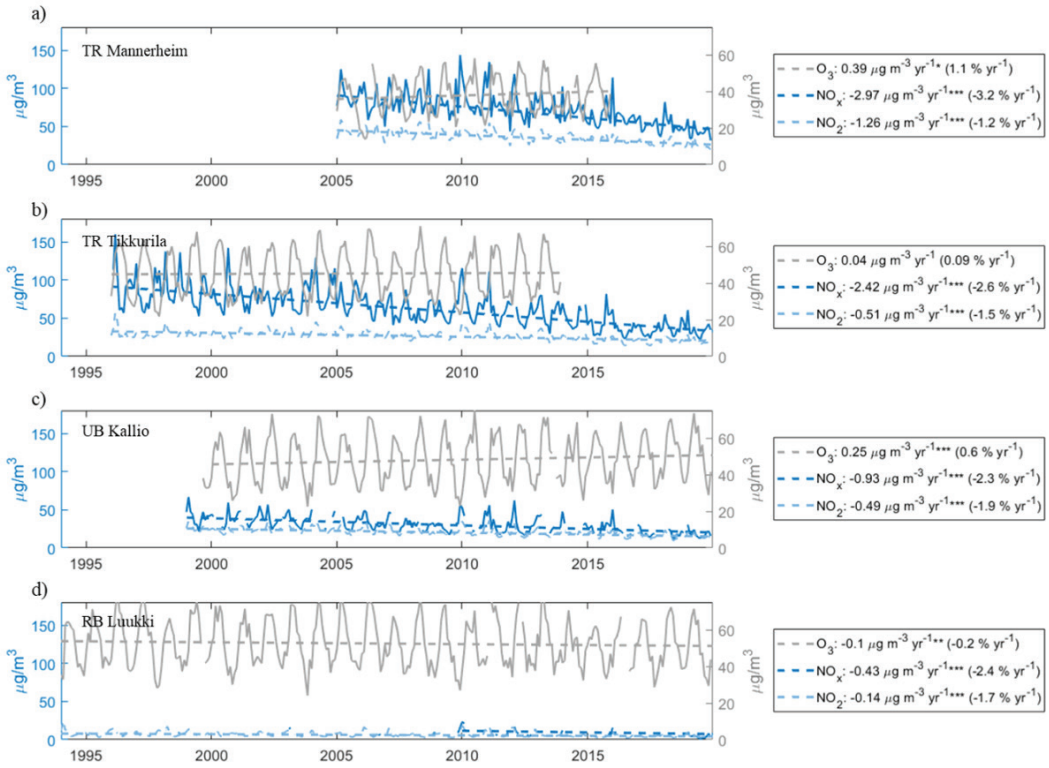

Fig. 2. The monthly average O_3 (grey), NO_x (dark blue) and NO_2 (light blue) concentrations and respective time trends (dashed lines) observed at Mäkelänkatu (TR), Tikkurila (TR), Kallio (UB) and Luukki (RB). The left y-axis is for NO_x and NO_2 and the right y-axis is for O_3.

at urban and rural background sites reached the minimum during summer and the maximum during winter and springtime. At Mannerheimintie traffic site, the seasonal cycles of NO_x and NO_2 were not as distinctive, but rather all seasons were similar. The pattern of hourly averages of both NO_x and NO_2 have remained the same, though the concentration levels have decreased. The diurnal cycles governed by morning and evening peaks from commuter traffic.

The EU's air quality directive (2008/50/EC) and World Health Organization (WHO) have set the guideline values of 200 μg m^{-3} for hourly average and 40 μg m^{-3} for annually averaged NO_2 concentration. The hourly guideline should not be exceeded more than 18 times per calendar year according to the EU's air quality directive. The hourly guideline was exceeded at traffic sites in Töölö, Vallila, Mannerheimintie and Mäkelänkatu, though the number of exceedances per calendar year remained always well below 18 times per calendar year. The annual guideline was exceeded at three urban traffic sites: Töölö (1996–2001), Mannerheimintie (2005–2010) and Mäkelänkatu (2015).

Regardless of the significant reductions in regional NO_x emissions, NO_2 concentration levels did not decrease as efficiently as expected. This is consistent with the analysis of Anttila (2020). The share of NO_2 of total NO_x increased at all the ten stations in the HMA (Fig. S7 in Supplementary Information). At the SMEAR II rural background station, the NO_2/NO_x ratio remained close to one due to the absence of local NO_x emissions. In urban background environments, the decrease is slower than expected likely due to changes in the O_3/NO_x equilibrium as NO_x concentrations have reduced and O_3 concentration increased (Keuken *et al.* 2009). On the other hand, in urban traffic environments the slower decrease of NO_2 concentration has been attributed to the increased direct NO_2 emission

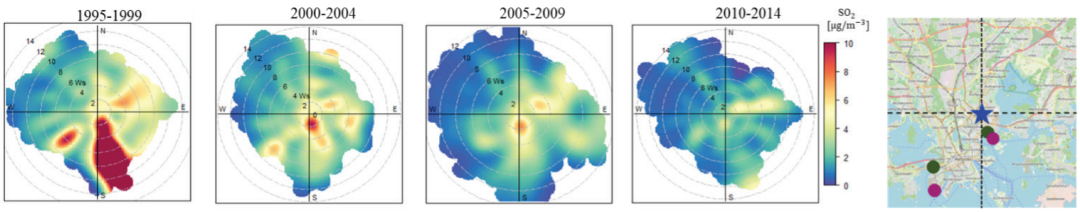

Fig. 3. Five-year mean SO_2 concentration in urban traffic site Vallila in relation to wind speed and direction. The map shows the location of Vallila measurement site (blue star), power plants (dark green spots) and ports (purple spots).

ronments, where the O_3 concentration were typically lower. In the Luukki rural background site, the number of exceedances varied from 0 (2019) to 49 (1996) days per year. However, WHO has not set any lower limit on the number of days when the exceedances occur.

Sulfur dioxide and carbon monoxide

SO_2 is formed during combustion processes of fossil fuels or other substances containing sulfur mainly during energy production and industrial processes (e.g., Zhong *et al.* 2020). High concentrations of atmospheric SO_2 can have severe impacts both on the environment and human health. The increased levels advance the acidification of freshwaters and soils (Schöpp *et al.* 2003, Stoddard *et al.* 1999) and can lead to aggregated asthma or reduced lung function (WHO 2005). SO_2 is also the main precursor of sulfuric acid, which is the main new particle formation agent (e.g., Kulmala *et al.* 2004, Petäjä *et al.* 2009).

Abatement of sulfur dioxide emissions started in the 1980s at the European level in order to prevent acidification of ecosystems (Vestreng *et al.* 2007). This resulted in SO_2 emissions decreasing 86% (4.3% yr^{-1}) in Finland from 1981–2000 (Vestreng 2003). Though the fastest pace of emission reduction was observed during the 1980s, the decline continued into the first and the second decade of the 21st century in the HMA. From 1994 to 2019, the SO_2 emissions were reduced by 72% (2.8% yr^{-1}) and from 2000 to 2019 by 30% (1.5% yr^{-1}). The reduction in emissions have been achieved as an outcome of several measures. Replacing high-sulfur solid and liquid fuels with low-sulfur ones e.g., natural gas in energy sector, flue-gas desulfurization abatement technology in industrial facilities as well as the directives set by the European Union to regulate the sulfur concentration of some liquid fuels have contributed to decrease in SO_2 emissions (Vestreng *et al.*, 2007). The EU introduced a sulfur directive in the Baltic Sea in 2015, which regulates the maximum sulfur content of maritime fuels to be 0.1%, when previously the maximum was 1% (EU, 2012). As a result, SO_2 emissions from harbors in the HMA decreased from 137 tons yr^{-1} in 2014 to 46 tons yr^{-1} in 2015. Energy production remains the largest contributor of SO_2 emissions in HMA covering 97% of the total emissions in 2019.

Due to the extensive abatement measures, SO_2 concentrations have decreased remarkably in Finland (Ruoho-Airola *et al.* 2003, Anttila and Tuovinen 2009) and in Europe. In the HMA, SO_2 concentration declined to background levels in urban traffic (Vallila) and urban background (Kallio and SMEAR III) environments. All five investigated monitoring sites show statistically significant negative trends. Luukki, Vallila and SMEAR III sites have the longest time series, and they showed an annual change in the SO_2 concentration between –3.0 and –4.1% yr^{-1}, which is larger than regional SO_2 emission trend (–2.8% yr^{-1}) in the HMA. Fig. 3 shows the five-year mean SO_2 concentration in Vallila traffic site from 1995–2014. This figure illustrates how the largest sources of SO_2 are from the south-east (Hanasaari power plant, Port of Sörnäinen) and south-west (Salmisaari power plant, Port of Helsinki) from the Vallila measurement station. During the 20-year period, the annual mean SO_2 concentration decreased from 5.1 µg m^{-3} in 1995 to 2.1 µg m^{-3} in 2014 in Vallila, mainly due to addition of flue-gas des-

from diesel vehicles (Carslaw 2005). This might be due to the new CO_2 emission-based tax for passenger cars enacted in the mid-2000s, which increased the number of diesel vehicles, and hence emitted more tailpipe NO_2. Anttila (2020) examined the share of the direct NO_2 emission fraction with the total oxidant model for Mannerheimintie urban traffic site from 2005–2015. Their results show that the annual emission reductions of primary NO_2 reached maximum (21%) in 2009 and has reversed since then, varying between 15–19%, which indicates that the conversion from NO to NO_2 has been larger since 2010.

Tropospheric O_3 is a secondary pollutant, which forms in the atmosphere through complex photochemical reactions between precursor gases like NO_x, CO and VOCs (Monks *et al.* 2015). The main sources of these precursor gases are traffic exhaust, vegetation and biomass burning. Elevated concentrations of O_3 can be harmful to human health by decreasing pulmonary function, and increasing throat irritation and inflammatory response in lungs (WHO 2006). In addition, surface O_3 can have negative effects on vegetation by causing leaf injury, reduced photosynthesis, and reduced growth rates (Karnosky *et al.* 2007).

The precursor gas emissions of O_3 have been greatly reduced in Europe and in the United States. However in Asia, Africa, and Latin America the emissions have remained stable or even increased (Huang *et al.*, 2017, Miyazaki *et al.* 2017, EEA 2020). Despite great efforts to reduce the emissions, observed surface O_3 concentrations in the background sites in the Northern Hemisphere have increased by 1–2% since 1950s (Wilson *et al.* 2012). In addition to regional and global precursor gas emissions, O_3 concentrations are influenced by photochemical production and destruction, stratosphere-troposphere exchange, and changes in air-mass transport patterns (Monks *et al.* 2015).

Increasing trends for O_3 daily average mean concentration have been observed by in many cities around Europe for urban and suburban stations (Colette *et al.* 2011, Yan *et al.* 2018). This is true in the HMA as well; O_3 exhibits a statistically significant positive trend at six monitoring sites including urban background stations (Kallio and SMEAR III), suburban detached housing site (Vartiokylä) and traffic sites (Töölö, Mäkelänkatu and Mannerheimintie). Based on our analysis, the annual increase varied between 0.6–2.8% yr^{-1} within the sites, except for Mäkelänkatu urban traffic station where the increase is 6.1% yr^{-1} (2015–2019). While summer and autumn concentration levels remained relatively steady, there was a clear elevation in wintertime O_3 levels both in the traffic and in the urban background environments, especially during the last five years (2015–2019) of the measurement period. According to previous studies in Europe, the increase during wintertime levels is due to increased long-range transportation of O_3 and decreased titration by NO (Colette *et al.* 2011, Sicard *et al.* 2016). Springtime ozone slightly increased in the traffic environment and remained stable in urban background (Fig. S8 in Supplementary Information). Table 2 shows a small, statistically significant decreasing trend of 0.09 μg m^{-3} yr^{-1} (0.2% yr^{-1}) for O_3 at Luukki rural background site from 1994–2019. Yan *et al.* (2018) showed negative trends for O_3 peak concentrations (95th percentile) and a positive trend for the 5th percentile at urban background sites in Europe, which led to insignificant trends for the mean concentrations. At urban background station, the decline was observed during spring, summer, and autumn, whereas wintertime ozone seemed to be increasing similarly to traffic and urban background sites (Fig. S8 in Supplementary Information).

The guideline value for O_3 concentration set by EEA is 120 μg m^{-3} for the maximum daily 8-hour running mean and this value should not be exceeded more than 25 days per year averaged over three years. These conditions were not met at any of the investigated measurement stations in the HMA from 1994–2019, although the guideline value was exceeded on several occasions, most often in the rural background site in Luukki. The WHO decreased their guideline value from 120 μg m^{-3} to 100 μg m^{-3} for the maximum daily 8-hour running mean O_3 concentration in 2005, since new epidemiological evidence was published on the association between mortality and O_3 levels (WHO, 2005). This new guideline was exceeded at all the stations in the HMA, also in the traffic envi-

ulfurization in power plants and the regulation of the maximum sulfur content of the maritime fuels. The port of Sörnäinen was closed in 2008.

The guideline limits set by EEA for SO_2 are 350 μg m^{-3} for one hour average and 125 μg m^{-3} for 24-hour average (EEA 2019). These limits were not exceeded at any of the sites during the period of 1994 and 2019. As the concentrations decrease, SO_2 is becoming less important as an air pollutant, but there is still great scientific interest towards the component due to its role as a precursor of sulfur aerosols (e.g., Petäjä *et al.* 2009, Sipilä *et al.* 2011).

CO is a primary pollutant, which forms during incomplete combustion processes of fossil fuels and biomass and via atmospheric volatile organic compound oxidation (e.g., Yin *et al.* 2015). In urban environments, the dominant source of CO is road traffic (e.g., Vimont *et al.* 2017). Exposure to high CO levels increase risks for mortality due to cardiovascular diseases and can weaken the amount of oxygen transported in bloodstream to critical organs (e.g., Liu *et al.* 2018).

With the introduction of emission controls, particularly three-way catalysts in vehicles, CO emissions from all sources decreased 89% (3.4% yr^{-1}) and from vehicular traffic 91% (3.5% yr^{-1}) from 1994 to 2019 (Table 2, Fig. S4 in Supplementary Information). Ambient concentration of CO was studied at six sites, of which the four traffic and suburban detached housing area showed a statistically significant negative trend varying between 2.9 to 5.4% yr^{-1} (Table 2). The longest continuous time series of CO was measured at Tikkurila traffic site from 1996–2016, during which the annual mean concentration decreased 3.4% yr^{-1}, which agrees with the regional CO emission reduction from all sources (–3.4% yr^{-1}). Similar decreasing trends for CO have been reported in Europe (Zellweger 2009, Angelbratt 2011). Since CO concentration in the urban environments decreased to background levels, the role of CO as air pollutant has become less important and CO measurements have been discontinued at many sites in the HMA. CO is still important from the climate change perspective as it produces indirect positive radiative forcing (IPCC, 2013).

Particulate matter mass concentration

Atmospheric PM consists of directly emitted primary particles and secondary particles, which are formed in the atmosphere through chemical reactions. These reactions depend on precursor gases such as SO_2, NO_x and volatile organic compounds (VOCs). PM size and chemical composition are dependent on sources and on both atmospheric and geographical conditions (e.g., Hopke *et al.* 2020). PM is identified as one of the most harmful air pollutants (Lelieveld *et al.* 2015) as it can enter to the thoracic region of the respiratory system. Exposure to high levels can increase the risk of aggravated asthma, cardiovascular and respiratory diseases and lung cancer (WHO 2013).

$PM_{2.5}$

Regional emissions of $PM_{2.5}$ and its precursor gases in Finland are relatively low compared with more polluted areas in Europe (EEA 2020). As a result, the $PM_{2.5}$ concentrations in the HMA remain typically on a relatively low level Fig. S9 in Supplementary Information. Nevertheless, high levels occur occasionally as a result from long-range transport of PM from more polluted areas in Eastern Europe (Niemi *et al.* 2009). According to Karppinen *et al.* (2004), long-range transport contributed up to 64–76% of $PM_{2.5}$ concentration in urban areas in Helsinki and less than 10% of the measured $PM_{2.5}$ concentrations in Luukki regional background site was emitted from local sources. Important local sources of $PM_{2.5}$ are energy production, traffic and residential wood burning. $PM_{2.5}$ concentration attributable to residential wood burning was estimated to be 31–66% in suburban areas and 18–29% in urban areas during heating season (Saarnio *et al.* 2012).

$PM_{2.5}$ had decreasing trend at all the eight investigated stations, seven of which were statistically significant. The annual reduction varied between 0.2 and 0.3 μg m^{-3} yr^{-1} within the stations, which corresponds to 2.3–3.6% yr^{-1}, except for Mäkelänkatu urban street canyon where the annual reduction was 0.5 μg m^{-3} yr^{-1} (5.8% yr^{-1}) (Table 2). The declining trend was observed

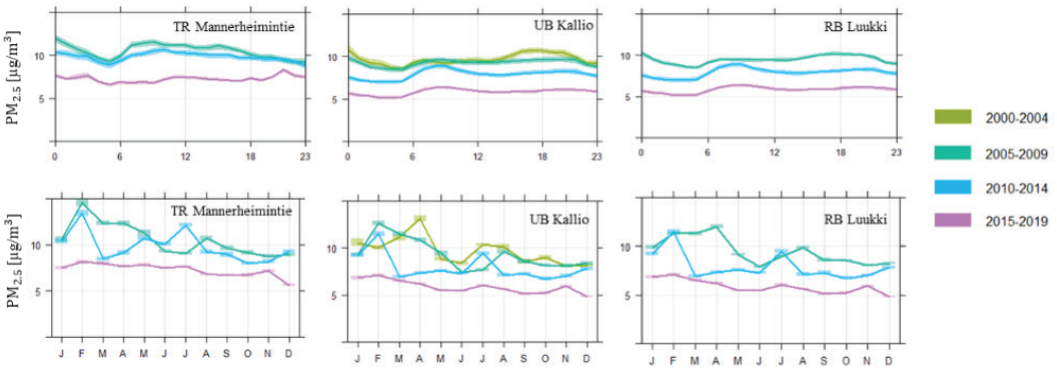

Fig. 4. Five-year mean diurnal (top row) and seasonal (bottom row) cycle of $PM_{2.5}$ concentration in Mannerheimintie, Kallio and Luukki measurement sites. The boxes on bottom row show the 95th confidence interval.

during all seasons at all sites with the largest reductions typically observed during winter and springtime. The change of $PM_{2.5}$ diurnal and seasonal cycle in traffic, urban background and rural background stations is presented in Fig. 4. The concentration levels and shape of the diurnal cycle are fairly similar at all three investigated sites. The shape of the diurnal cycle remained relatively similar though the concentrations decreased at all sites. Similarly to the diurnal cycle, also the seasonal cycle followed the same pattern at three sites, which reinforces that regional and long-range transport contribute a large fraction to $PM_{2.5}$ concentration at the HMA.

The decrease in $PM_{2.5}$ concentration was achieved through emission cuts both in direct PM emissions as well as emissions of the precursor gases. Total, energy production and vehicular traffic PM emissions were reduced by 91% (3.5% yr^{-1}), 91% (3.5% yr^{-1}) and 89% (3.4% yr^{-1}) respectively from 1994–2019 in the HMA (Table 2) At the same time, the concentrations of precursor gases SO_2 and NO_x were reduced by 72% (2.8% yr^{-1}) and 62% (2.4% yr^{-1}) respectively.

De Jesus *et al.* (2020) studied the $PM_{2.5}$ and particle number concentration trends and how changes in meteorological parameters and legislation influenced the concentration trends in Helsinki, Augsburg, Brisbane, London and Rochester. Their results showed that the $PM_{2.5}$ concentration in Helsinki (Kallio urban background site) decreased at a rate of 0.2 µg m^{-3} yr^{-1} from 2001–2017, which was the lowest of the significant trends in their study. They also identified four change points in the trend analysis for the $PM_{2.5}$ concentration in Helsinki in 2003, 2006, 2010 and 2014. The first change point they assumed to be due to reduced sulfur content in gasoline and diesel, the second and the third due to long-range transported particles from wildfires and the last due to reduced sulfur content in maritime fuels in Baltic Sea and warm winter in 2015. In the same study, they found out that increased wet deposition and dilution due to increased precipitation and higher wind speeds affected the $PM_{2.5}$ concentration.

PM_{10}

Numerous studies have shown that traffic-induced resuspension is the dominant source of PM_{10} particularly nearby busy roads (e.g. Vallius *et al.* 2000, Hosiokangas *et al.* 2004, Kukkonen *et al.* 2005). Typically, atmospheric PM_{10} concentration remains comparatively low in the HMA, however road dust episodes cause high peaks during springtime. As the snow and ice melts typically in March–April, traffic enhances the resuspension of grit, which is used to reduce the slipperiness of the roads during wintertime. The use of studded tires boosts the formation of road dust (Hosiokangas *et al.* 2004) and dust resuspension.

PM_{10} time series was available at six traffic and one urban background site. A statistically significant decreasing trend was detected for all of these sites. The largest annual reduc-

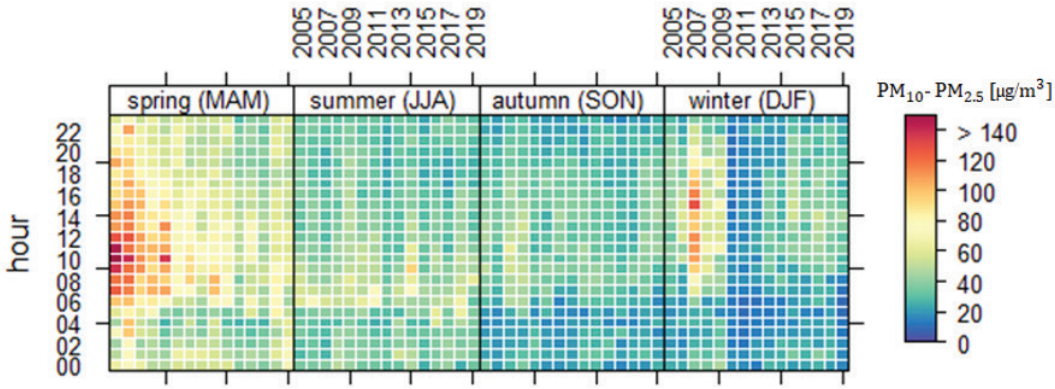

Fig. 5. The 95th percentile of coarse particles (PM_{10}–$PM_{2.5}$) in Tikkurila traffic site during four seasons. Year is on the x-axis and hour is on the y-axis. The colors represent the 95th percentile of coarse particle mass concentration.

tions were measured at three urban traffic sites; 0.6 µg m^{-3} yr^{-1} (2.3% yr^{-1}) in Töölö (1994–2004), 0.6 µg m^{-3} yr^{-1} (2.1% yr^{-1}) in Mannerheimintie (2005–2019) and 1.2 µg m^{-3} yr^{-1} (5.1% yr^{-1}) in Mäkelänkatu (2005–2019). The smallest reduction was measured at the urban background site in Kallio with annual reduction of 0.2 µg m^{-3} yr^{-1} (1.4% yr^{-1}) from 1999–2019.

Kupiainen (2007) showed that most of the road dust, which is formed of traction sand and pavement wear particles, is in coarse size fraction (aerodynamic diameter varying between 2.5 and 10 µm). Figure 5 shows the 95th percentile of hourly coarse particle mass concentration during four seasons at Tikkurila traffic site. As expected, the highest concentrations are measured during springtime due to road dust episodes. The 95th percentile of hourly coarse particles occurring during daytime have clearly decreased during spring (left-hand column). Still, from 2005–2009 the peak concentrations reached up to 140 µg m^{-3}, but they have since decreased to levels of 40–80 µg m^{-3}. In 2007, winter temperatures in the HMA remained mild, which led to earlier and shorter road dust episode than usually. This is seen as higher peak values during winter months during 2007 (right-hand column).

Implications to the Air Quality Index

The distribution of the Air Quality Index between 1995 and 2019 is shown for each air pollution indicator separately in Fig. 6. The left column is for traffic site and the right column is for urban background site. In accordance with our previous results, share of hours of good air quality increased for NO_2, $PM_{2.5}$ and $PM_{10.}$ For O_3, the number of good hours decreased, which led to increase in hours of fair air quality especially in the urban background environment. O_3 has been proven to be the most complex air pollutant to control due to its dependence on other precursor gases.

Elevated NO_2 concentrations have been an issue especially in busy traffic environments. However, during the past decade, exceedances of NO_2 levels decreased in both traffic and urban background site due to improved emission reduction technologies and renewal of vehicle fleet.

PM_{10} caused more hours of moderate, poor, or very poor air quality than other considered air pollutants in traffic environment. The exceedances of the guideline values occurred typically due to road dust episodes during springtime. In urban background environment, the air quality was classified mostly as good or fair due to $PM_{10.}$ Though the number of hours of fair, moderate, and poor air quality due to $PM_{2.5}$ have been decreasing in traffic and urban background environments, exposure to $PM_{2.5}$ still causes more than 1500 premature deaths annually in Finland (EEA 2019).

The European AQI break point value between good and fair air quality for hourly

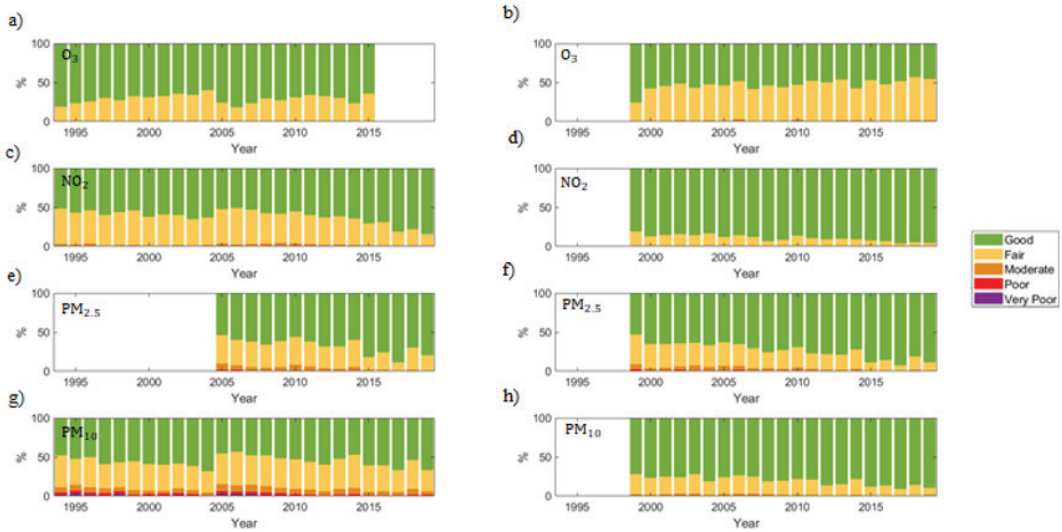

Fig. 6. The distribution of calculated air quality indices based on historical data classified into good, fair, moderate, poor, and very poor categories. The left column is for traffic site and the right column is for urban background site. The left-hand side column is for traffic site (Töölö 1994–2004, Mannerheimintie 2005–2019) and the right-hand side is for urban background site (Kallio 1999–2019).

SO_2 concentration is 100 µg m^{-3}, which is significantly higher than observed concentrations in the HMA. The mean concentrations at traffic site Töölö (1994–1998) was 5.2 µg m^{-3}, at the traffic site Mannerheimintie (2013–2014) it was 1.9 µg m^{-3} and at the urban background site in Kallio it was 1.2 µg m^{-3} (2014–2019). The limit value for fair air quality was exceeded 20 times in Töölö. There were no exceedances in Mannerheimintie and in Kallio.

Traffic exhaust emissions and residential wood burning are the major sources of air pollution in the HMA together with energy production. In the current AQI standard, the effect of locally emitted traffic exhaust on air quality is represented mainly by NO_2 concentration, since the majority of $PM_{2.5}$ is long-range transported also in urban areas (Karppinen *et al.* 2004). Exceedances due to NO_2 concentration are becoming less and less frequent also at urban traffic station, due to rapidly decreasing NO_x emissions from vehicular traffic (Fig. 6e). However, several previous studies have shown that vehicular traffic emits also ultrafine particles (UFPs) with diameter less than 0.1 µm (Rönkkö *et al.* 2017). UFPs are identified as harmful air pollutants to human health (e.g., Schraufnagel 2020), due to their efficient lung deposition and ability to translocate to other parts of the body. In addition, they have high surface-to-mass ratio, ultrahigh reactivity, and smaller size than the dimensions of cellular structures, which makes them extremely toxic (e.g., Sukhanova *et al.* 2018). Similarly to traffic, residential wood burning would need a better indicator to represent the resulting health. In the report by WHO (2012), the experts suggested the use of black carbon (BC) as an additional air quality indicator to highlight the population exposure to combustion PM. Currently, UFPs and BC are not considered in air quality regulations.

Conclusions

In this work, we explored long-term trends of ambient gaseous pollutants and particulate matter mass in the Helsinki metropolitan area (HMA) from 1994 to 2019. The sites considered were characterized as urban background, suburban detached housing area or traffic dominated environments. As a rural background, we utilized data from Luukki in the HMA region and SMEAR II in Hyytiälä as a regional reference.

We explored gas-phase concentration (NO_2, NO_x, O_3, SO_2 and CO) and aerosol particle mass concentrations ($PM_{2.5}$ and PM_{10}).

The statistical trend analysis showed that both NO_x and NO_2 concentrations decreased at all the sites in the HMA between 1994 and 2019. NO_x levels decreased more rapidly than NO_2 levels, which might be partly due to increased number of diesel vehicles operating in the Helsinki area (Anttila 2020), which increases direct NO_2 emissions. Due to the decrease in NO_x emissions in the traffic environment, there is even more O_3 that can efficiently convert NO to NO_2. The overall effect is the faster decrease of NO_x than NO_2 close to the traffic sources (e.g., Krecl *et al.* 2021; Fig. 2). The greatest drop in concentration levels was seen for SO_2 and CO. With the introduction of emission controls and improved technology, SO_2 and CO concentrations decreased to background levels at all the sites. Aerosol particle mass concentration, both $PM_{2.5}$ and PM_{10}, decreased at all investigated sites. For PM_{10}, the largest declines were detected at traffic sites while $PM_{2.5}$ showed more uniform decrease within the sites. The 95th percentile of hourly coarse sized particle mass concentration, which form most of the street dust, showed a negative trend during springtime at the Tikkurila traffic site.

The secondary air pollutant, O_3, remains a challenge in the perspective of air pollution control. O_3 concentration showed increasing trends at traffic, urban background, and suburban sites. While the spring and summertime levels remained relatively stable, there was a clear elevation in wintertime O_3. Conversely, at the rural background site in Luukki, the monthly mean O_3 concentration decreased 0.2% yr^{-1} from 1994–2019.

The share of hours of good air quality calculated with the Air Quality Index (AQI) has increased for NO_2, $PM_{2.5}$ and $PM_{10.}$ O_3 shows a clear increase in hours of fair air quality at urban background site and moderate increase at the traffic site. $PM_{10,}$ which typically exceeds the guideline limit of good air quality for the period of street dust season during springtime, remains a challenge at busy traffic site. NO_2 concentration has decreased due to reduced NO_x emissions from vehicles.

Since NO_2 concentration, which have typically represented the health effects resulting from vehicular traffic, is rapidly decreasing, the current AQI lacks information on the resulting health effects from exposure to traffic exhaust, which contains harmful components such as ultrafine particles (UFPs). Similarly to traffic, health effects from residential wood burning are not sufficiently represented in the current AQI. Residential wood burning is an important source of combustion PM and black carbon (BC), which have been shown to be more harmful than PM from other sources. We suggest that the current AQI should be revised to consider new air quality parameters, which would represent the health effects resulting from vehicular traffic and other local combustion sources such as residential wood burning. Based on results from previous exposure studies and reports, we propose to include UFPs and BC in the AQI.

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